DESULPHURIZATION OF NIGERIAN LAFIA COAL FOR METALLURGICAL COKE PREPARATION

BY

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ABSTRACT

The Government-owned integrated iron and steel plant under construction in Ajaokuta has been designed to operate through the blast-furnace process. The blast-furnace requires metallurgical coal. It therefore became necessary and relevant to the national economy to scientifically work on Nigerian coals with a view to using same as metallurgical coal. Lafia coal is the only Nigerian coal that produces coke lump after dry distillation (carbonization). The property of coke-ability therefore attracted attention towards Lafia coal. The main interest was to so prepare the coal that as much as possible of it will be usable for metallurgical coke preparation for use in Ajaokuta blast-furnace.

Lafia coal was subjected to chemical characterization and petrological studies. It became clear that it was high in ash and sulphur. Silicates, pyrites and carbonates of various elements constituted the inorganic mineral inclusions. Analytical data gave the sulphur varieties in Lafia coal to be organic (0.1 - 0.7%), pyritic (1.4 - 4.4%) and sulphate (0.01 - 0.07%).

Pyritic sulphur which predominates in Lafia coal can be removed by physical means. Chemical desulphurization which is reducto-oxidative type of chemical reaction impares coal cokeability. A plunge was thus made for physical desulphurization techniques to bring down the total sulphur in Lafia coal to the level that will make it acceptable as a metallurgical coal with regards to its sulphur content.

Trace elements in Nigerian coal ash determined using the Atomic Absorption Spectrophotometer (AAS) and confirmed with the Inductively Coupled Plasma Emission Spectrophotometer (ICPES) showed that the relative abundance of the elements in the coals depended on the biogeo-chemistry of the area of the deposit as follows:

Lafia coal; F>Zn>Mn>>V Cr>Ni>Cu>B>Pb>Ca>Be>Mo>Cd>Hg
Enugu coal; B>Mn>V F>Cr>Zn>Cu>Pb Ni>Co>Be>Mo>Hg
Okaba coal; Mn>>B>>V>Cr>Cu>Pb>F=Ni>Zn Co>Be>Mo>Hg

Trace element content of coal was considered in connection with the impact of coal utilization on the environment.

From the study it was concluded that either the wet concentrating table or the froth flotation method could be used to substantially desulphurize Lafia coal to meet the requirement of metallurgical coal.

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DEDICATION

TO

EL SHADDAI

CERTIFICATION

I certify that this work was carried out by MR. WILLIAM IBITOYE ATIKU ADERONPE in the Department of Chemistry, University of Ibadan, Ibadan, Oyo State, Nigeria

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ABBREVIATIONS

equ. = equation

SME = Society of Mining Engineers

AIME = American Institute of Mining and Metallurgical Engineers

Contd. = Continued

Trans. = Transactions

Jnl. = Journal

edn. = Edition

Geol. = Geological

Bull. = Bulletin

Surv. = Survey

Div. = Division

Dept. = Department

Sa/No = Sample Number

Se/No = Serial Number

M = Moisture

VM = Volatile Matter

db = dry basis

daf = dry ash free

Chem. = Chemistry

Ind. = Industry

ASTM = Amercian Society for Testing and Materials,

mtce = Million metric tonnes coal equivalent.

CHAPTER ONE:- INTRODUCTION

1.1. THE ORIGIN AND TYPES OF COAL

1.1.1. Introduction

Coal is a fossil fuel deposit of vegetable matter (i.e. plants, mosses, ferns, etc) which grew in the carboniferous era (i.e. the period or system of the paleozoic era characterized by coal bearing rocks) rendered compact and hard by pressure and heat. Coal originated from the remains of ferns, spores, seeds, mosses, vines, bushes, trees and other plants which grew and fluorished in extensive swamps and bogs some 300 million years ago during prolonged periods of humid, tropical climate and abundant rainfall. Bacterial and chemical action converted these plant debris over time into organic matter called peat.

The overlying water burden on the peat formed, excluded oxygen thereby retarding the rate of decay of peat and its further transformation into other products. The accumulation of more plant debris and sediments on top of the peat compressed and solidified it, converting it into coal by bio-chemical and physical processes.

Thus peat is the precursor of coal and the processes converting peat into coal is known as COALIFICATION. The extent of the Coalification process which depends significantly on the prevailing temperatures determines the RANK OF COAL. The latter will be discussed in greater detail later on. However when the prevailing temperature was low at depths of only a few hundred metres beneath the surface, coalification was relatively slow and resulted in the formation of LOW RANK COAL such as LIGNITE AND SUBBITUMINOUS COAL. The rate of coalification has been shown to be catalysed by intrusion of igneous rocks[1].

Further studies have shown that the major chemical changes that occur during coalification from a low-rank coal e.g. Lignite to a high-rank coal e.g. Anthracite, are the decrease in hydrogen and oxygen content accompanied by increase in carbon content. Pressures on the coal deposits due to overburden have also been reported to have little effect on the chemical coalification process but having significant effect on the physical-structural development of coal[1].

1.1.2. Coal Types

Physical characterization of coal through microscopic studies shows that it is characterized by layers or bands having glossy or dull appearances. The glossy layers are composed mainly of VITRINITE which was formed from woody parts of plants (e.g. trunks, branches, roots, etc). The dull bands consists of finely divided material formed from leaves, pollen, spores, seeds, resins and are called EXINITE. Other dull bands of coal include FINE EXINITE and more coalified MICRINITE and FUSINITE.

Banded, stratified coal types are described as BRIGHT, SEMI-SPLINT and SPLINT. The United States Bureau of Mines[2] defines BRIGHT COAL as one containing less than 20% opaque matter; SEMI-SPLINT COAL is one containing between 20% and 30% opaque matter while SPLINT COAL is made up of more than 30% opaque matter.

Non-banded, non-stratified coals are called SAPROPELIC COALS and there are two types namely: CANNELS AND BOGHEADS; and are characterised by a small percentage of VITRAIN. Bogheads possess a high percentage of volatile oils and gases, and contains an abundance of algae remains.

Cannels or Candle Coals are so named because they burn with unusual brilliance. Cannel coal is non-coking, often contains large quantities of spores and pollen materials similar to bog-head.

Cannel Coal also has a high content of volatile oil and gas. A hybrid Coal with composition midway between the two exists and is called CANNEL-BOGHEAD or BOGHEAD-CANNEL. Stopes and wheeler[3] and later Fox[4] have proposed a definition of coal as follows: "Coal is a compact stratified mass of metamorphosed plants which have in part suffered arrested decay to varying degrees of completeness". This definition excludes the various oil shales, canneloid shales, and carbonaceous shales, all of which are deposits in which inorganic material is dominant. The definition also excludes isolated coalified material such as stems, pinnules, and fonds. The definition however includes: brown coal, lignite, sub-bituminous, bituminous and anthracite coal; and boghead, cannel, pseudo-cannel and splint coal, some of which have already been described. Peat, which is the parent material from which humic coals are formed, has not undergone metamorphosis at that stage and so it is not covered by the definition.

1.1.3. Theories of Coal Formation

Apart from the foregoing concise description of the nature of coal, some of the various theories on the nature of coal will now be examined. Deluc[5] as early as the later part of the eighteenth century believed that coal was formed from plant debris that had accumulated in a peat swamp, had been covered by later sediments and had undergone later changes that produced a sequence of coals beginning with peat and terminating with anthracite. This rationalization of the origin of coal which has been described earlier-on is called the IN SITU PEAT SWAMP OR ANTOCHTHONOUS THEORY of origin of coal. In support of this theory Rogers[6] and Potonie advanced the arguments that:- (i) the generally low content of mineral matter in coal precludes large-scale transportation of the plant material, otherwise transportation would have been accompanied by transportation of equally great amounts of mineral matter; (ii) the presence beneath most normal banded coals of underclays, which are believed to be ancient soils, indicates growth of the plants IN SITU; (iii) the wide lateral extent of many coal beds could have developed only in areas of essentially coextensive plant growth;

(iv) the arrangement and condition of the plant particles are not those of transported material; (v) the presence of rooted tree trunks and roots both in underclays and coals is indicative of plant growth in situ; (vi) large modern peat deposits have formed in situ, cover comparatively large areas, and if converted to coal would form deposits comparable in size, shape, and purity to many coals; and (vii) the deposition of the large quantities of plant debris in open water would leave the deposits subject to destructive bacterial decay.

Many students of coal, throughout most of the nineteenth century, contended that the parent material of coal was not formed by degradation of plant material in situ in peat swamps but rather resulted from the deposition of transported plant material in lakes, seas, and estuaries. To this school of thought belonged Hutton[7] who believed that particles of root from smoke, soluble organic matter carried by streams, and remains of plants and animals both native to the sea and transported from land were deposited in the sea, buried by inorganic debris, and later converted to coal. This theory is termed ALLOCHTHONOUS or TRANSPORTATION THEORY OF COAL FORMATION.

As to conditions of deposition of the parent materials the normal banded (or layered) coals are believed to have formed from peat-swamp types of deposits. Potonie [8] divided the coal-forming materials into three groups, namely, HUMIC COALS, SAPROPELITHS and LIPTOBIOLITHS. He believed that the parent material of humic coal was formed by the partial decay of land plants and swamp plants in a moderately moist habitat. Such coal is called BRIGHT COAL (or GLANZKOHLE) and is rich in carbohydrates. The Sapropeliths were formed in stagnant water from the remains of aquatic organisms that were primarily planktonic. The most important of these were the oil-bearing algae, certain crustaceans, and a subordinate admixture of pollen grains and other drifted materials. Liptobioliths consist primarily of resins, waxes, and spores, and represent the more stable residue of decomposed land plants. Potonie further explained the banding of normal coal by postulating alternation of moist conditions which yielded bright coal with periods of pronounced dryness that permitted more complete destruction of humic materials and the formation of dull bands of liptobiolithic materials and occasional periods of flooding that produced sapropels.

Several factors were observed to come into play at the dynamochemical (or metamorphic) stage of peat to coal transformation. As soon as peat is buried by later sediments, it is subjected to pressure by the load of over-burden and from that time onward through its history, the organic residue is undergoing changes in response to the two agencies of heat and pressure. The difference in composition of original materials was, however, found to be the largest single factor determining the variations in fixed carbon content in coals. Furthermore, White has developed the thesis that horizontal thrust pressure due to deformation of the earth's crust is the dominant agency in effecting the metamorphosis and increase in rank of coal. He realised that the increase in rank of coal under the influence of thrust pressure was the result of a combination of the direct effect of pressure with increased temperature resulting from compressions and friction induced by the pressure, both agencies operating over a long time period.

Fusain (mineral charcoal) is one of the common constituents of coal, but some workers do not believe pure fusain to be coal.

The mode of origin of fusain has been a source of much speculation. In general, two schools of thought have existed, the first attributing the origin of fusain to charring of plant material in forest fires, [9] and the second to some form of chemical alteration of plant material prior to burial [10]. The general objections to the forest-fire theory that were expressed were: i) lack of ash layers and ash mixed with fusain such as resulted from modern fires; ii) inprobability of extensive fires in a typical peat swamp; iii) the fact that very fragile plant structures, such as finely veined fern pinnules were fusainized; iv) the existence of plant stems, such as cordaites, in which pithy core is fusainized and the outerpart converted to vitrain; v) the fact that the temperature required to produce fusain in forest fires would have been so great that resins would have disappeared and also spores, waxes, and other hydrogenous substances would have been fused or destroyed; and vi) the transition from vitrain to fusain in individual wood fragments. On the other hand, the arguments advanced in favour of the forest-fire origin of fusain are:

i) that there is at least apparent similarity between fusain and charcoal formed by fire; and ii) that fragments of charcoal occur in concretions, coal balls, etc, and must have existed as charcoal prior to deposition. Hendricks [11] believes most fusain to have been formed by dehydration of plant bebris before burial and further loss of volatile constituents after burial. The general absence of fusain as such in peat together with its general presence in normal banded coal seems to preclude a forest-fire origin for most fusain.

Cannel coals, which are typically made up chiefly of spores and spore exines, are believed to be formed from accumulation of spores, spore exines, and pollen in areas of open water. These materials are believed to have been transported either by water or wind to the site of deposition. The generally high mineral-matter content of cannel coal, the occurence of most cannel coals in lenticular, cannel-shapped bodies, the presence of invertebrate fossils, and even fishes in some cannels, and the spore content of the cannels all suggest transportation of the parent material. After deposition of the spore-rich material the formation of cannel coal apparently is parallel to and a result of the same agencies of heat and pressures that form and modify the normal banded coals.

Boghead coals are characterized by abundant alginite and very finely dispersed inertinite (fusain type not fusible at high temperature) and vitrinite. In open-water areas associated with peat swamps or other areas of abundant decaying humic debris, the toxic fundamental jelly was present near the bottom and served as a matrix into which the spores of cannel deposits and the algae of boghead deposits settled and were preserved.

Mineral matter content of coal presents a problem as to its origin. However, the nature and distribution of mineral matter in coal give some information bearing on the conditions of deposition of the coal-forming materials. Lessing [12] classified the mineral constituents of coal into a) the residue of the mineral constituents of the plants from which the coal is derived; b) detrital matter, blown, washed, or settled into the deposit; c) saline deposits from water with which the plants or plant residues were in contact before and during coal formation; d) crystalline deposits from waters which percolated the coal seams through cracks and fissures during and after coal formation; e) products of decomposition of the above substances and of interaction between themselves and the coal substances.

Lessing analysed ash remains of the four main coal macroscopic constituents, namely, FUSAIN, DURAIN, CLARAIN, and VITRAIN and obtained the result shown in Table 1. Lessing considered the ash of Clarain and Vitrain largely inherent plant ash, that of Durain largely detrital clay particles, and that of Fusain together with that found in cleats and partings largely material deposited from infiltrating solutions. In Lessing's opinion the data supplied by his studies (Table 1) so far as they bear on the origin of coal, indicated that clarain and vitrain consist of pure plant remains, whereas durain contains much clay mixed with small plant entities. Lessing believed this to indicate that durain which contained high silica was formed in deeper water than vitrain and clarain and was subjected to contamination by transported mineral debris.

TABLE I

Composition of the Ashes from the Four Main Constituents of Hamstead "thick coal" according to Lessing

Parameters	FUSAIN (weight %)	DURAIN (weight %)	CLARAIN (weight %)	VITRAIN (weight %)
SiO_2	8.84	50.54	9.44	6.08
Al_2O_3	8.66	42.34	16.58	15.49
Fe_2O_3	3.37	1.36	3.31	3.09
MnO	0.51	0	0.23	0.13
TiO_2	0.04	0.44	0.50	0.24
CaO	57.00	3.69	12.98	15.22
MgO	1.30	0	10.52	1.87
Na_2O	3.24	0	15.71	17.67
K_2O	0.67	0	---	0.20
SO_2	14.65	3.23	32.18	30.89
P_2O_5	---	---	0.01	trace
CO_2	2.98	---	---	6.69
	101.26	101.60	101.46	97.57
% ash soluble in H_2O	16.57	3.48	65.24	69.52
% ash soluble in HCl	71.38	23.81	17.86	20.46
% ash insoluble in HCl	12.05	72.71	16.90	10.02
	100.00	100.00	100.00	100.00

1.2. Nature of Coal

Coal is highly heterogeneous.

It is opaque but when a thin section of it is cut and light is shone through it on one side, then on the other side it acquires such transluscence that displays the fine structural boundaries of constituent particles. Coal is non-crystalline and varies in colour from light brown to black, of dull to glossy lustre according to rank, petrographic composition (inclusive of minerals) and techtonic factors.

Coal has specific gravity varying from 1.0 to 1.8 depending on the maceral composition. It has fairly weak tensile and impact strengths and a force of 0.5 to 2.5kg/cm^2 is enough to break or shatter the coal structure. It contains, in varying amounts, essentially all elements of the periodic table combined into nearly all of the minerals normally encountered in the earth's crust. The organic matrix comprises most of the coal weight and consists mainly of carbon, with smaller amounts of hydrogen, oxygen, nitrogen and sulphur. Coal can be considered a rock structure containing both macroscopic and microscopic petrographic features.

It has also been viewed as an organic chemical substance containing the classical organic functional groups, like the carbonyl and the hydroxyl, the aromatic and heterocyclic ring units and aliphatic bridges.

From another standpoint, coal might be defined as "a solid colloid which has a large volume porosity and can absorb gases and vapours as well as liquids". Furthermore, the organic coal matrix may be characterized as a cross-linked polymer (formed from the cellulosic polymer present in plant material which, in the absence of degradation, is essentially insoluble and non-volatile.

1.2.1. The Organic Coal Matrix

Given [13] has proposed an admittedly arbitrary model (Figure 1) for the organic coal matrix involving methylene bridges of the 9, 10-dihydroanthracene type, aromatic structures including benzo-pyridine, benzoquinone and benztropolone as major constituents, and non-aromatic units such as cyclohexanone, cyclohexane and the like. The major feature of this model involves the bonding of aromatic nuclei by two methylene linkages.

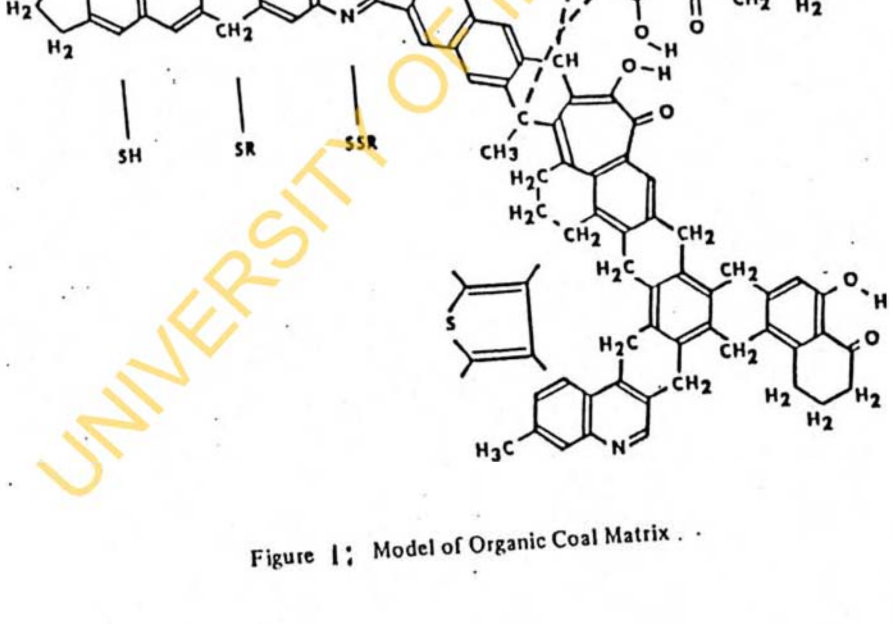


Figure 1: Model of Organic Coal Matrix .

This structure was suggested on the basis of spectroscopic and carbon/hydrogen ratio data as well as knowledge of the chemical reactions of coal with bromine and oxygen. A complete determination of the actual structural formula for coal is an elusive goal, since coal structure must be highly variable among various coals and may include all known organic structures, as well as some yet to be defined.

In the organic structure of coal matrix mercaptan, sulphide, disulphide, thiophene and thiopyrone ring were suggested as the functions containing organic sulphur.

1.2.2. The Inorganic Coal Structure

The major minerals typically present in bituminous coals [12] are listed in Table 2 below.

Table 2: Minerals Associated with Bituminous Coals

Mineral Group	Species	General Formula for the Group
Shale	Muscovite	$(K,Na,H_3OCa)_2(Al,Mg,Fe,Ti)_4(Al,Si)_8O_{20}(OH,F)_4$
	Hydromuscobite Illite Bravaisite Montmorillonite	
Kaolin	Kaolinite Levisite Metahalloysite	$Al_2(Si_2O_5)(OH)_4$
Carbonate	Ankerite Ankeritic calcite Ankeritic dolomite Ankeritic chalybite	$(Ca,Mg,Fe,Mn)CO_3$
Sulphide	Pyrite Marcasite	FeS_2
Chlorides	Sylvine Halite	KCL NaCl

It is very difficult to totally separate the minerals from the coal for analysis since the mineral component of coal is intimately associated with the organic coal matrix.

The list above is based on optical microscopy, spectroscopy and analysis of the ash content obtained after combustion of the organic coal matrix. Coal ash composition ranges for oxides of the above minerals are listed below in Table 3.

Table 3: Coal ash Composition of bituminous Coals

Constituent Oxide	Present
Silica (SiO_2)	20-60
Aluminium oxide(Al_2O_3)	10-35
Ferric oxide (Fe_2O_3)	5-35
Calcium oxide (CaO)	1-20
Magnesium oxide	0.3-4
Titanium oxide (TiO_2)	0.5-2.5
Alkalies (Na_2O + K_2O)	1-4
Sulphur oxides (SOx)	0.1-12

The trace element composition of coal is highly variable, depending on the origin of the coal and the method of mining. Many of the trace elements are associated with both the inorganic and organic portions of the coal matrix.

The sulphur content of the inorganic portion of coal occurs mainly in the minerals pyrite and marcasite. However, a small amount of inorganic sulphur is also present as sulphate minerals such as melanterite ($FeSO_4.7H_2O$), jarosite $KFe_3(SO_4)_2(OH)_6$ and gypsum ($CaSO_4$). In freshly-mined coal, the total sulphur content, as sulphate, rarely exceeds 0.1 percent by weight of coal, but in weathered coal, it may rise to 0.2 percent or more. The minerals, pyrite and marcasite have the same chemical composition (FeS_2) but differ in crystal form, pyrite is isometric (cubic), while marcasite is orthorhombic. Pyrite is the most commonly found form in coal, with marcasite occuring in lesser amounts. Marcasite, slightly less stable than pyrite and more reactive chemically, is converted to the pyrite form on heating.

1.2.3. MOLECULAR STRUCTURE OF COAL

Coal is not a uniform simple molecular species but may be regarded as a statistical structure made up of small, condensed aromatic units or layers with a variety of substituent groups around the perimeter and some cross-linking between adjacent units. Some of the units may not be strictly plannar because of the presence of hetero-atoms (oxygen, nitrogen or sulphur) or hydro-aromatic portions, but they approximate to planarity and consequently show some tendency to pack parallel to one another, although this parallelism is not very extensive. The structure of graphite consists of extensive parallel layers of carbon-atoms arranged in condensed, aromatic arrays, the layers being mutually orientated and separated by a distance of 0.335nm (the carbon-carbon van der waals' distance) [9] The parallel stacking of layers in coal is similar, but lacks mutual orientation between layers, and the average spacing between layers is somewhat larger.

The sketch below approximates the molecular structures of the commoner three 'ranks' of coal. The low rank coal has small layers more or less randomly orientated connected by cross-links, so that the structure is highly porous (Figure 2).

Coking coal shows a greater degree of orientation and consequently a greater tendency towards parallel stacking. There are less cross-links, fewer pores and the structure has some similarities to that of a liquid (Figure 3).

High rank coal or anthracites shows a growth of the individual layers, a marked increase in the degree of orientation and the development of a new type of pore elongated parallel to the stacks of layers (Figure 4).

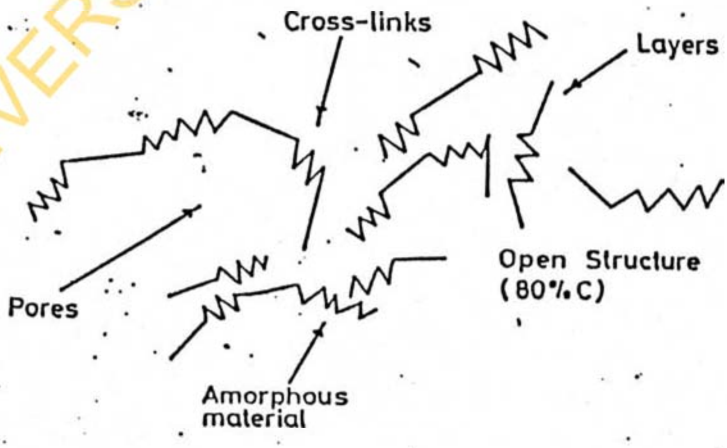


Fig.2: Low rank coal type of Structure

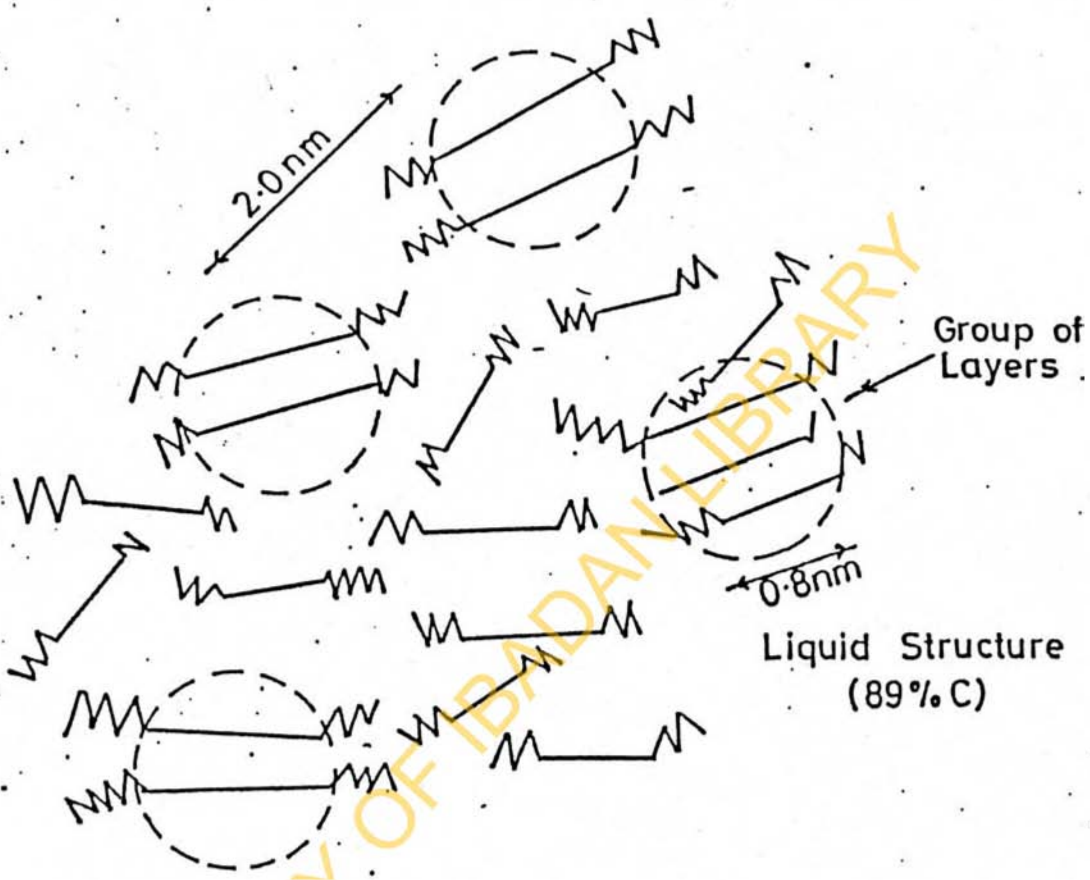


Fig. 3: Coking coal type of Structure.

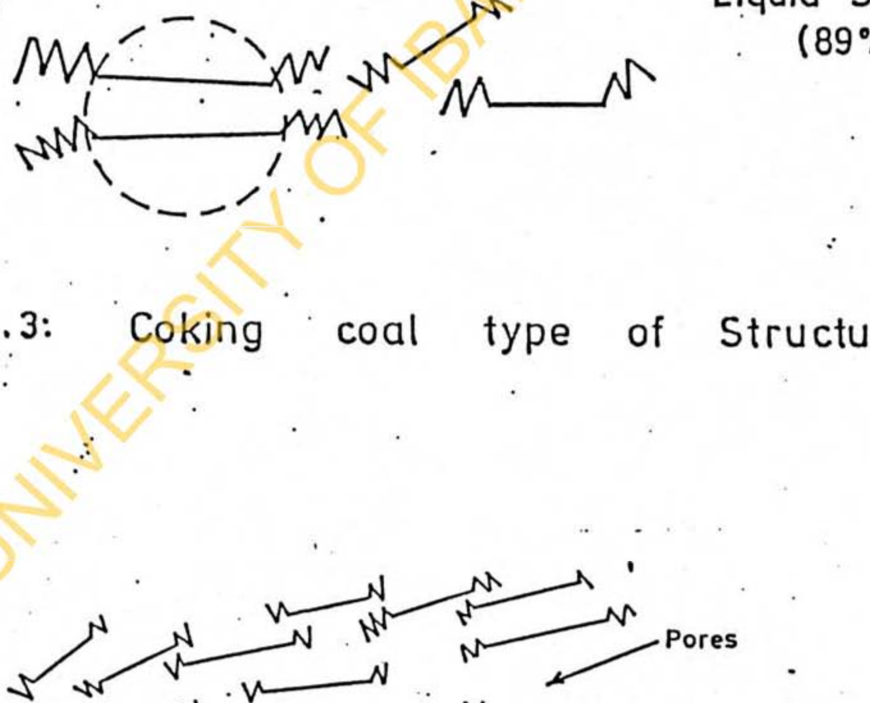


Fig.1: High rank coal type of Structure

When talking about the nature of coal its behaviour on heating is typical and important from the practical stand-point. When heated, most coals evolve tarry vapours, gas and moisture and some coals soften and fuse into a cake residue.

In the presence of air, the combustible products ignite. The tarry vapours burn with a smoky flame, the gases with non-smoky flame and the solid residue glows, leaving ash derived from the mineral matter. The calorific value of coal varies with rank. The low rank coals generate less heat than the medium rank or the high rank because the former contains higher oxygen and moisture contents. Even then the low rank coals are widely burned because of their greater availability and lower cost.

Heated in the absence of air coal gives the principal products of metallurgical coke, domestic coke, coal tar, town's gas, oxides of sulphur and nitrogen and other products including ammonia. The tar is derived from aromatic layer molecules in the original structure which are only weakly held by cross-links and can therefore be evolved as a result of minor decomposition. This process is limited because the breakage of cross-links generate free valencies and these tend to satisfy themselves by recombination.

Gaseous products such as methane and hydrogen are derived from the breakage of bonds to peripheral substituent groups and combinations of the resulting radicals. This also leaves free valencies and increases the incidence of recombination so that the coke remaining becomes more cross-linked and non-volatile.

1.3. CLASSIFICATION OF COAL

Coal has been classified in different ways based on various parameters and criteria [14]. Simplistically, coal has been classified as mined-coal, charcoal, lamp-black or soot. Blackness of colour is the factor used to call all these substances coal. However, a common origin attributable to carbonaceous organic origin could justify this simple approach regarding these three different substances but having common characteristics as coal. Industrially, mined-coal is of course, the substance to be classified. Charcoal is charred wood. Soot is in-completely combusted fuel carbon particles.

In coal science, vitrain, clarain, durain and fusain, are sometimes referred to as different classes of coal. However, in the real sense these four types of coaly materials are banded components of humic coal.

At other times, coal has been classified as peat, lignite, sub-bituminous and bituminous coal and anthracite.

The most desirable classification of coal is one that suggested definitive maturation stages in coalification.

Ideally, classification of coal has to be done scientifically from observable features and for a purpose. Classification of coal is done to enable us understand at once the nature, type, physico-chemical properties and maturation stage of the coals. Such classification makes it easy to shop for desired coals for specific end uses.

Most systems of coal classification are based upon some characteristic property of the coal series that change uniformly and progressively with increase in 'maturity' or "rank" (or degree of coalification) as the series is ascended. The parameters generally used for this purpose are the "Ultimate Analysis" (often called the "Elementary Composition) and/or the "Proximate Analysis".

The parameters usually considered are the weight percentages of carbon, hydrogen, oxygen and nitrogen. Other parameters used include moisture (free and inherent), volatile matter, (material that is volatilised when the coal is heated in the absence of air at a certain temperature and for a certain length of time), weathering properties, fixed carbon and calorific (heating) value. The reflectivity of the coal substance caused by Vitrinite, being the most prevalent of coal materials is also used as a classification parameter.

The caking and swelling properties of coals are additional important classification parameters. The swelling of coal is measured on very rapid heating in a crucible (about 400°C/min). Another method of determination of coal swelling property is that of percentage of maximum dilatation on slow heating (3°C/min). A sample of coal, compressed to a thin strand of about 1mm diameter is heated at constant rate (3°C/min) while a plunger, resting on the pencil, indicates the swelling of the coal as a function of time. Whereas the determination of the swelling index supplies information about the behaviour of coal on very rapid heating (and thus under conditions resembling those in a fire grate), the dilatation test is suitable for assessing the behaviour during slow heating (as in a coke oven).

These physico-chemical properties have formed a basis for several classification systems. Some of these classification systems and the codes they use to describe coal types will now be discussed.

1.3.1. INTERNATIONAL CLASSIFICATION OF HARD COALS BY TYPE

The International Classification of Hard Coals by Type uses the criteria of volatile matter on the dry-ash-free basis (d.a.f.), calorific value on the moist ash-free basis, caking properties (free swelling index or Roga index), and coking properties (dilatometer test or Gray-King assay) to place hard coals into classes, groups and sub-groups [2,3]. The term "hard coals" is used to refer to coals that have a gross calorific value of more than 5,700 kcal/kg (10,260 Btu/lb) on a moist ash-free basis (m.a.f.). A coal after being tested for these properties is then given a three-figure code number: according to International Standards Organization (ISO). The first figure indicates the class of the coal, the second figure the group, and the third figure the sub-group.

Class:

The class to which a particular coal sample belongs is established on the basis of its volatile matter on the dry-ash-free basis (d.a.f.) up to 33% volatile matter, and on the moist-ash-free (m.a.f.) calorific value. There are ten classes of coal from 0 to 9. Classes 0,1A, 1B, 2, 3, 4, and 5 are based entirely on volatile matter. Classes 6 - 9 are based on volatile matter from 33 to 50% and on calorific values of 7,750 down to 5,700 kcal/kg (13,950 down to 10,260 Btu/lb). This provides the first figure of the code.

Group:

The ten classes are further divided into four groups, 0 to 3, according to caking properties using the Roga Test (ISO 335) or the crucible swelling number (ISO 501). The group number provides the second of this three-figure code system.

Sub-Group:

The four groups are further divided into six sub-groups, 0 to 5, using the coking properties as measured by the Audibert-Arnu Test (ISO 349) or the Gray-King coke test (ISO 502). The last of the three-digit code is the sub-group number.

The class, group and sub-group classification of coal [4,6,7] described above is shown in Table 4.

1.3.2. CLASSIFICATION OF COALS BY RANK

This ASTM Standard Classifies coals according to the degree of fossilised vegetative metamorphism (or the progressive alteration in the natural series from lignite to anthracite) [3,11]. The criteria of rank are fixed carbon and calorific value, expressed in Btu/lb or kcal/kg and calculated on the mineral-matter-free basis. The higher rank coals are classified according to fixed carbon content on the dry mineral-matter-free basis. The lower rank coals are classified according to their calorific value on the "moist" mineral-matter-free basis. Moist refers to coal containing its natural inherent moisture but not including visible water on the surface of the coal.

TABLE 4: International Classification of Hard Coal by Type

GROUPS (determined by coking properties)			CODE NUMBERS										SUBGROUPS (determined by coking properties)		
GROUP NUMBER	ALTERNATIVE GROUP PARAMETERS		The first figure of the code number indicates the class of the coal, determined by volatile-matter content up to 33% V.M. and by calorific parameter above 33% V.M. The second figure indicates the group of coal, determined by coking properties. The third figure indicates the subgroup, determined by coking properties.										SUBGROUP NUMBER	ALTERNATIVE SUBGROUP PARAMETERS	
	Free-swelling index (crucible-swelling number)	Roga Index												Dilatometer	Gray-King
3	>4	>45					435	535	635				5	>140	>G_8
						334	434	534	634				4	>50-140	G_5-G_8
						333	433	533	633	733			3	>0-50	G_1-G_4
						332a 332b	432	532	632	732	832		2	≤0	E-G
2	2½-4	>20-45				323	423	523	623	723	823		3	>0-50	G_1-G_4
						322	422	522	622	722	822		2	≤0	E-G
						321	421	521	621	721	821		1	Contraction only	B-D
1	1-2	>5-20			212	312	412	512	612	712	812		2	≤0	E-G
					211	311	411	511	611	711	811		1	Contraction only	B-D
0	0-½	0-5		100 A B	200	300	400	500	600	700	800	900	0	Nonsoftening	A
CLASS NUMBER			0	1	2	3	4	5	6	7	8	9			
CLASS PARAMETERS	Volatile matter (dry ash-free)		0-3	>3-10 >3-6.5 >6.5-10	>10-14	>14-20	>20-28	>28-33	>33	>33	>33	>33			
	Calorific Parameter[a]		-	-	-	-	-	-	>13.950	>12.960-13.950	>10.980-12.960	>10.260-10.980			

As an indication, the following classes have an approximate volatile-matter content of:

Class		
Class 6	33-41%	volatile matter
7	33-44%	" "
8	35-50%	" "
9	42-50%	" "

CLASSES

(Determined by volatile matter up to 33% V.M. and by calorific parameter above 33% V.M.)

*Gross calorific value on moist, ash-free basis (30°C, 96 pct relative humidity; Btu/lb.

Note: (1) Where the ash content of coal is too high to allow classification according to the present systems, it must be reduced by laboratory float-and-sink methof (or any other appropriate means). The specific gravity selected for flotation should allow a maximum yield of coal with 5-10 percent of ash. (2) 332a ...>14-16 pct volatile matter, 332b ...>16-20 pct volatile matter. (3) Classes determined by volatile matter up to 33 pct volatile matter and by calorific parameter above 33 pct volatile matter.

In order to be able to classify coals according to the ASTM standard it is necessary to carry out proximate analyses (moisture, ash, volatile matter, and calculation of fixed carbon by difference and calorific value) and to determine the equilibrium moisture on lower rank coals that may be classified on the "moist" basis. For certain coals it is necessary to know the agglomerating character of the coal. This physical property is determined by examination of the residue in the silica crucible used to determine volatile matter of the coal. Coals that in this test produce either an agglomerate button that will support a 500g weight without pulverizing, or a button showing swelling or cell structure shall be considered agglomerating. Table 5 shows the classification of high and low rank coals according to ASTM Standard.

Table 5: Classification of HIGH AND LOW RANK COALS according to ASTM

Class	Group	Fixed carbon limits percent (Dry, mineral-matter-free basis)		Volatile matter limits, percent (Dry mineral-matter-free basis)		Calorific value limits (Btu/1b moist, mineral-matter-free basis)		Agglomerating character
		Equal or greater than	Less than	Greater than	Equal or less than	Equal or Greater than	Less than	
I. Anthracite	1. Meta-anthracite	98	-	-	2	-	-	Non-agglomerating
	2. Anthracite	92	98	2	8	-	-	
	3. Semi-anthracite	86	92	8	14	-	-	
II. Bituminous	1. Low volatile bituminous coal	78	86	14	22	-	-	Commonly agglomerating
	2. Medium volatile bituminous coal	69	78	22	31	-	-	
	3. High Volatile A bituminous coal	-	69	31	-	14,000	-	

Table 5 Contd.

	4. High volatile B bituminous coal	-	-	-	-	13,000	14,000	
	5. High volatile C bituminous coal	-	-	-	-	11,500	13,000	
						10,500	11,500	
III. Sub-bituminous	1. Sub-bituminous A coal	-	-	-	-	10,500	11,500	
	2. Sub-bituminous B coal	-	-	-	-	9,500	10,500	
	3. Sub-bituminous C coal	-	-	-	-	8,300	9,500	Non agglomerating
IV. Lignitic	1. Lignite A	-	-	-	-	6,300	8,300	Non-agglomerating
	2. Lignite B	-	-	-	-	-	6,300	

1.3.3. SOFT BROWN COALS AND LIGNITES - CLASSIFICATION BY TYPES ON THE BASIS OF TOTAL MOISTURE CONTENT AND TAR YIELD (ISO 2950)

The parameters of total moisture and low-temperature tar yield are used to divide these low rank coals into classes and groups (Table 6 and 7). The moisture parameter indicates its value as a fuel and the tar yield its value in chemical processing. The total moisture is determined according to ISO 1015 Brown Coals and Lignites - Determination of moisture content - Direct Volumetric method. Tar yield is determined by ISO 647 Brown Coals and Lignites method - Determination of the Yields of Tar, Water, Gas, and Coke Residue by Low-Temperature Distillation.

Table 6: Class Numbers of Soft Brown Coal Types

Class	Total moisture content of run-of-mine coals on the ash-free basis(%)
1	<20
2	20-30
3	30-40
4	40-50
5	50-60
6	60-70

Table 7: Group Numbers of Soft Brown Coal Types

Group	Tar yield on the dry ash-free basis (%)
0	<10
1	10-15
2	15-20
3	20-25
4	>25

Coalification and classification could be followed by results of chemical analysis as shown in the table 8 below:

Table 8: Chemical Composition of Wood, Peat and Various Coals.

	% C (dmmf)	% H(dmmf)	% O(dmmf)	% N(dmmf)
Wood	50.0	6.3	42.7	1.0
Peat	57.0	5.2	36.8	1.0
Lignite	65.0	4.0	30.0	1.0
Low rank coal	79.0	5.5	14.0	1.5
Medium rank coal	88.0	5.3	5.0	1.7
Anthracite	94.0	2.9	1.9	1.2

In West Germany, a classification system for coking coal has emerged based on the Ruhr Dilatometer dilatation measurement and the mean maximum reflectance (in oil) as shown on Figure 5.

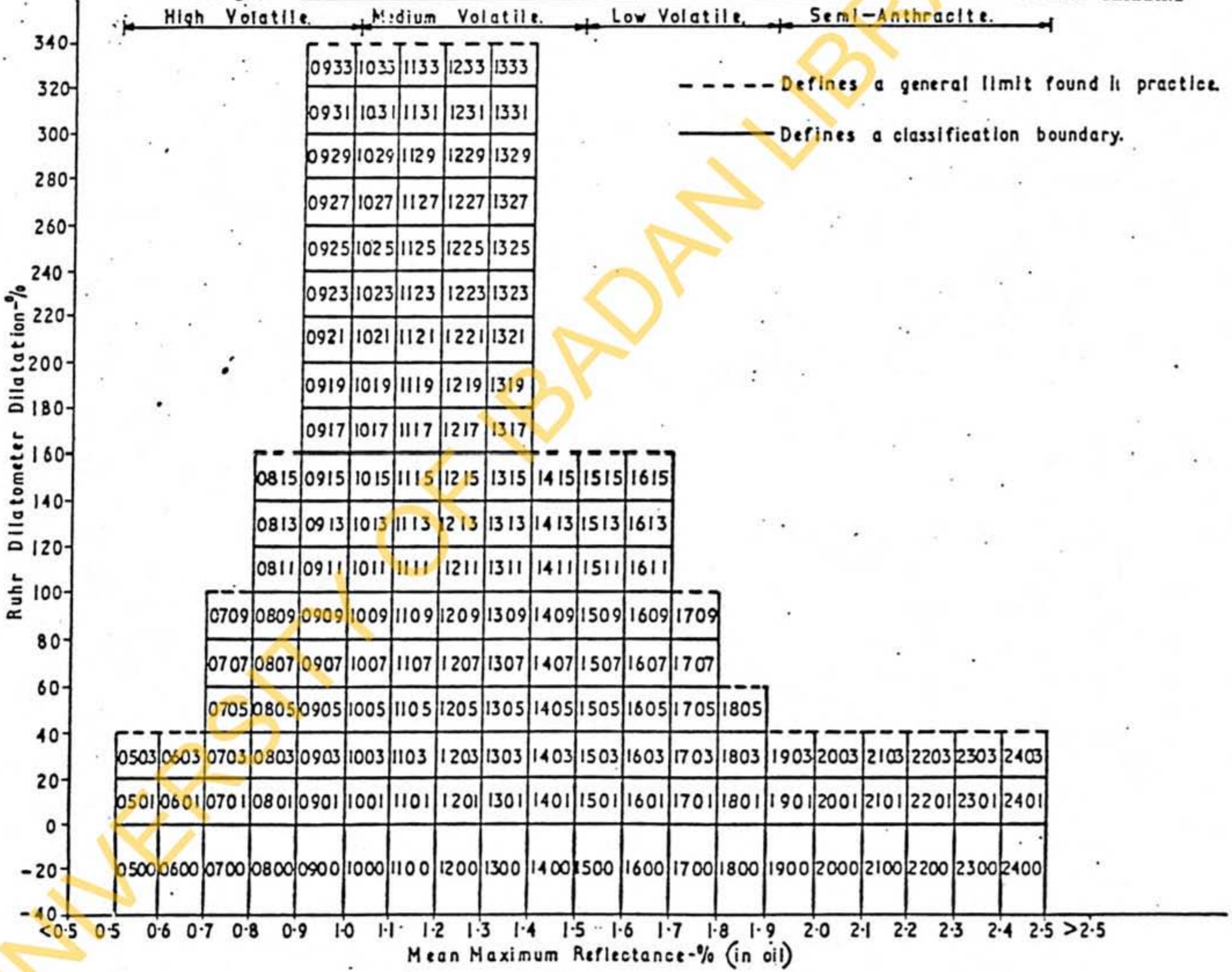
The International Committee for Coal Petrology has produced a tabular schematic analytical outline of coal classification as given in Table 9. The last column to the right of Table 9 is particularly instructive as it gives the applicability of the different parameters for the determination of rank. From the said column it is to be observed that the parameters of the moisture in situ, calorific value (a.f.), reflectance of the vitrinites and carbon content (d.a.f.) are likely to be suitable for use in the classification of Nigerian coals which are tertiary and fall within the immature or low rank coal type.

Table 9:- Outline of Coal Classification (ICCP)

Rank increasing

		Rank Stages	% Reflectance of vitrinite	Important microscopic characteristics	% C in vitrinite	Volatile matter % d.a.f. in vitrite	% H_2O in situ	Calorific value of vitrite (a.f.)	Applicability of the different parameters for the determination of rank
		Peat		Large pores Details of initial plant material still recognizable Free cellulose	50		75~		Calorific value (a.f.) or moisture in situ (moisture-holding capacity)
Brown coal		Soft brown coal		No free cellulose Plant structures still recognizable (cell cavities frequently empty) Marked gelification and compaction takes places	60				
			ca. 0.3			ca. 53	35~	7,200 B.t.u./lb (4,000 kcal/kg)	
	Hard brown coal	Dull brown coal		Plant structures still partly recognizable	70	ca. 49	25~	9,900 B.t.u./lb (5,500 kcal/kg)	Carbon (d.a.f.) Reflectance of the vitrinites
		Bright brown coal		(cell cavities filled with collinite)					Volatile matter (d.a.f.)
			ca. 0.5			ca. 45	8-10~	12,600 B.t.u./lb (7,000 kcal/kg)	
Hard coal		Bituminous hard coal		Exinite becomes markedly lighter in coulour ("Coalification jump") Exinite no longer distinguishable from vitrinite in reflected light	80 90	30			X-ray diffraction (graphite lattice)
			ca. 2.5			10		15,500 B.t.u./lb (8,650 kcal/kg)	H. (d.a.f.)
		Anthracite Graphite	11.0	Reflectance anisotropy	100	0			

FIG. 5 : COKING COAL CLASSIFICATION SYSTEM. FROM WESTERN GERMANY



1.4. Coal Resources and Reserves

The world energy conference (WEC) [15] held in 1978 has defined geological coal resources as hard and brown coal deposits, (regardless of seam thickness), of maximum depths of between 1800 - 2000 metres. The reserves that are technically and economically recoverable are defined as those of hard coal of minimum seam thickness of 0.6 metres and maximum depth of 1,500 metres as well as those of brown coal of minimum seam thickness of 2.0 metres and maximum depth of 600 metres.

1.4.1. Regional Distribution of Coal Resources and Reserves

Although there are quite significant coal resources in many parts of the world, it is apparent from Table 10 below that 10 countries account for 98 percent of the currently estimated world coal resources and 90 percent of the reserves [16,17]. Moreover, 4 countries - the Soviet Union, the United States, the Peoples' Republic of China and Australia - contain about 90 percent of the total resources and 60 percent of the reserves.

Table 10: World Coal Resources and Reserves by major coal-producing countries(mtce)

		Geological Resources	Technically and Economically Recoverable Reserves
1.	Australia	600,000	32,800
2.	Canada	323,036	4,242
3.	People's Republic of China	1,438,045	98,883
4.	Federal Republic of Germany	246,800	34,419
5.	India	81,019	12,427
6.	Poland	138,750	59,600
7.	Republic of South Africa	72,000	43,000
8.	United Kingdom	190,000	45,000
9.	United States	2,570,398	166,950
10.	Soviet Union	4,860,000	109,900
11.	Other countries	229,164	55,711
	Total World	10,750,212	662,932

Source: World Energy Conference and WOCOL Country Reports.

1.4.2. World Coal Production

The 9 largest producers - i) the United States, ii) the Soviet Union, iii) the People's Republic of China, iv) Poland, v) the Federal Republic of Germany, vi) the United Kingdom, vii) Australia, viii) the Republic of South Africa and ix) India - account for about 85 percent of world coal production. Three largest producers - the United States, the Soviet Union and the People's Republic of China - account for about 60 percent of world coal production.

Coal has been the major fuel man depended on until recently when petroleum was discovered and because of its easier transportation and cleaner combustion products the latter displaced coal to the second place. Coal has been regarded as the black diamond that as a fuel has made the industrial revolution of the 18th Century possible. Coal is highly sought after for multifarious purposes such as steam generation in industry and commerce; in transportation in the days of the steam-ship and railway locomotives, in the home for heating and warming up, for generation of town gas, for generation of thermal power, in metallurgy as fuel and chemical reductant.

The pharmaceutical and agricultural world find coal as useful raw material for drugs and chemicals.

Table 11 shows world production of fossil fuels [15] from 1835 to 1960 portraying an ever rising trend in succeeding years. Table 11 illustrates world coal production and reserves [18, 19, 20, 21, 22]. The coal types being exploited all over the world are anthracite, bituminous and lignites (as in Table 12). World total reserves and resources are 11.6 trillion metric tons. Out of the world total North America possesses 3.7trillion metric tons, Europe-Asia has 5.7trillion metric tons, Asia 1.1trillion, Africa 94.4 billion, South Africa 28.7billion, Oceania 350billion, and Europe 0.61 trillion metric tons. It is also apparent from Table 12 that the industrialised nations of the world, the United States, Soviet Union, Western Europe exploit their reserves and resources of solid fuels much more purposefully than do the developing nations. Their coal utilization permeates all facets of human endeavour and more particularly industrial and commercial activities. In other words, coal utilization has become an indicator of techno-economic activity and hence of the level of development of a nation.

Table 11: CONSUMPTION OF FOSSIL FUELS

WORLD PRODUCTION IN 10^6 TONS

Year	Coal	Lignite	Mineral oil	Total in standard tons of coal
1835	36	-	-	36
1855	89	-	-	89
1885	422	-	5	430
1905	953	(100)	31	1049
1913	1213	127	62	1370
1925	1192	180	156	1517
1930	1226	189	193	1611
1935	1147	186	224	1575
1938	1235	241	273	1765
1940	1407	285	289	1985
1945	1199	158	356	1813
1946	1245	223	375	1922
1947	1408	236	412	2164
1948	1406	255	467	2288
1949	1372	283	469	2214
1950	1507	302	523	2438
1951	1587	331	609	2663
1952	1510	414	621	2648
1953	1507	435	656	2709
1954	1480	457	709	2771
1955	1602	490	785	3045
1956	1676	565	841	3273
1957	1710	592	881	3384
1958	1802	611	926	3546
1959	1862	620	964	3618
1960	1971	643	1038	3850

Table 12:

Comparative Data of Coal Mining in Selected Countries, 1968

item	Australia	Canada	U.S.	West Germany	U.K.	South Africa	Soviet Union	Japan	India
Geologic age*									
Anthracite	P	Cr	Pa-Te	none	C	C-Tr	C	none	Pa-C
Bituminous	Tr or P	Cr-C-Te	C-Te	C-Cr-Te	C	C-Tr	C-Ju	Tr-Ju-Te	P-C-Te
Lignite or brown	Te	Cr-Te	Cr-Te	Te-Pl	none	none	Te-Me	Te	Te
Tectonic movement									
Folding (pitch)	90%<10° 1%>20°	heavy in areas	limited areas	22%>20°	heavy in areas	<5°	heavy in areas	heavy+	
Faulting	limited	in the west	limited areas	extensive	extensive	none	prevalent	prevalent	prevalent
Igneous intrusion	extensive	...	very limited areas	limited	limited	heavy in areas	...	heavy in areas	heavy in areas
Dept of overburden									
Average (ft)	520	1,700	<500‡	2,400	1,468	<300	1,130	1,510	490
Maximum (ft)	1,200	2,500	...	10%>3,300	3,765	1,430	3,600	3,025	1,970
Seam thickness									
Average (in)	102	96	63.6#	62.6	51.2	...	60	71.6	118
Minimum (in.)	36	60	24%	19.7	18	24	20	...	31
Maximum (in.)	216	...	7.1%>86#	197	150	240	1,180	...	827
Underground methods									
Longwall	3.3%	32%	1.34%	99%	96.5%	2 faces	85%	87.5%	1.3%
Longwall, with powered roof supports	100%	...	100%	27.5%	760 faces	100%	549 faces	14.7%	-
Room-and-pillar(%)	96.7	68	98.65%	0	3.5%	>98‡	1.7,8 incl. slicing, 3.2 shield, 2.1 other	7.1	98.7
Hand loading (%)	0.5	0	4.55	8.7	8.0@	60	...	...	97
Mechanical loading(%)	99.5	100	95.45	90.3	92	40	2 hydraulic	...	3
Conventional (%)	23.3	0	51	0	9	96	48	...	99.63
Continous	75.7	100	49	100	91	4	50	67	0.37
Productivity^µ									
Ungerground	10.06	6.14	15.07Ɨ	3.89	3.42	2.83	2.19	2.44	-
Surface	52.4	43.4	36.1 Ɨ	...	...	none	13.33	-	-
Total average	30.9	27.4	19.17Ɨ 18.8I	3.44	...	2.83	2.78	2.44	0.65
Largest operation (000,000 tons)	12.0 opencut	...	5.8 strip	20.0 opencut	...	4.27 under-ground	5.5 opencut	...	1.19 opencut

*P-Permian; Tr-Triassic; Te-Tertiary; Cr-Cretaceous; Pa-Pennsylvanian; C-Carboniferous; Ju-Jurassic; Pl-Pleistocene; Me-Mesozoic. + Mining mostly 6-12 degrees pitch seams-access 59% shaft. ‡ Estimated. # 1965. @ 1969.
µ Based on tons per day for all employees to point of shipment. Ɨ Bituminous and lignite, 1967. I Bituminous, lignite, and anthractie, 1967.

Sources: U.S. Department of the Interior, Bureau of Mines, IC, 8345 (August 1967); Coal-Bituminous, Lignite and Anthracties, Bureau of Mines Yearbook (1968 preprints); National Coal Association, World Coal Trade (1969); Mining Congress Journal (December 1969).

A selected list of countries that have developed their coal mining, on a large scale are also chosen on Table 12 to illustrate the age of the coals being mined, tectonic movement in areas of coal mining activity, depth of over-burden, seam thickness, underground mining method, and the productivity with respect to different mining engineering methods [19,23,24,25,26, 27,28]. Coal of all geological ages are being mined from permian through to the tertiary coals. The folded, faulted and igneously intruded coal stratigraphy are variously mined by the opencut, strip or underground longwall, longwall with powered roof supports or the roof-and-pillar mining engineering techniques. The depth of overburden reaches close to 1219.2m in some countries like the United Kingdom and the Soviet Union. Seam thickness averages, for example, 259.1cm in Australia, 243.8cm in Canada, 161.5cm in the U.S., 159.0cm in West Germany. 130.0cm in the U.K., 150.0cm in the Soviet Union, 181.9cm in Japan and 299.7cm in India. Most collieries utilize mechanical loading devices.

However, when in 1974 the price of oil skyrocketed the entire world went jittery. The after-math of this situtation was the beginning of the World coal study (WOCOL) meetings. In these meetings the strategic role of coal as the bridge to the future of more reliable energy source diversification was appreciated. The cleaner, more easily transportable and higher-heat-capacity other fossil fuel, petroleum, was calculated to be on the fast depletion slope [20,21]. Unlike oil, the reserve base for coal is sufficiently great to support large increase in production for a long time into the future. Moreover, the technology for its safe and environmentally acceptable, production, transport, and use is proved and already widely applied in most cases.

1.5. WORLD ENERGY PROSPECTS

There has been a worsening outlook for oil. During the past two decades world oil consumption has grown twice as fast as that of all other energy sources combined. Two-thirds of the growth in energy consumption in the member countries of the Organization for Economic Cooperation and Development (OECD) came from oil during this time.

In Western Europe and Japan, oil has provided over 80' percent of this increase in energy use. In all but a few of the developing countries oil has also supplied virtually all of the increase in energy consumption. It is a matter of considerable concern therefore that there now appears to be no realistic prospect of oil meeting any substantial part of future increases in the world's energy needs.

The present international oil trade amounts to about 35 million barrels a day (mbd), which is about 55 percent of the world's total oil consumption. Of that amount about 80 percent is exported by the Organization of Oil Exporting Countries (OPEC). There are no secure grounds for assuming that OPEC production of oil will increase in the future, and there are good reasons to fear it may be less. Moreover, key producers like Saudi Arabia and Kuwait have adopted policies that will conserve oil in the ground as a better security for their future than income they cannot spend or usefully invest. It is now the declared policy of OPEC member countries to limit oil production to amounts that will total nor more than the present level of about 30 million barrels a day. This policy is directed toward optimizing rather than maximizing revenues.

The goal is to generate only as much revenue as their economies can handle without causing too rapid social change and high inflation. Increasing prices while holding production stable, or even reducing it, meets this financial objective and at the same time conserves oil resources.

Prospects for increased oil supplies from non-OPEC areas are not sufficiently encouraging to change this picture. Production from fields in the North sea and Alaska's Prudhoe Bay in USA are projected to peak and level off in the next few years. It appears that Mexico and Norway will continue to limit oil exports to meet revenue needs. Increases in the production of oil in the People's Republic of China and the developing countries will be largely used to meet growing domestic demand. Furthermore, some experts have suggested that production in the Soviet Union, which now exports 1 million barrels a day to countries outside Eastern Europe, is levelling off, and that the Soviet Union/Eastern Europe area is therefore unlikely to remain self-sufficient in oil. For all these reasons, it is believed that oil will provide, at most, a small fraction of any future increase in world energy needs.

The world must find ways other than oil to provide the additional energy required for its future economic growth.

Natural gas and liquefied natural gas (NLG) have found wide use in many industrialised countries, such as the United States and Western Europe. Inports of gas by pipeline as liquefied natural gas (LNG) are significant in some countries and may increase. The rate of increase is however, likely to be slow because of long lead times and the large capital investment required for under-sea pipelines, liquefaction, transport, and regasification facilities. Furthermore, since it is OPEC member countries that are in possession of natural gas for export, there is high probability that ceiling production levels might be instructed to raise prices like is now the case for oil.

A vast resource base exists in several countries for unconventional oil in the form of oil shale, tar sands and heavy oil, like for example, Canada produces about 7 percent of its oil from Alberta tar sand deposits. Coal beds, shales, tight sands, and geo-presurrized formations may also contain huge amounts of unconventional gas. A substantial expansion of unconventional oil or gas production is now being planned for some countries.

However, all these unconventional sources are characterized by high capital costs and long lead times for development, all which slow down rate of development.

Nuclear power, which already plays a substantial role in electricity generation in some countries, like in some regions of the United States and Europe where it provides about 30 percent of the total electricity, has several uncertainties. Within very recent times the nuclear explosion and/or leakage in Long Island and Chenobil in the United States and the Soviet Union respectively have heightened fears of insecurity in the use and storage of nuclear power. Until public confidence is restored, it is difficult to make reliable predictions about the future contribution that nuclear power will make.

Aggressive conservation programs now occupy a central place in the energy strategies of many nations. The achievement of substantial savings, however, tends to be gradual, even with government intervention in the form of tax and financial incentives, as well as regulations governing energy performance standards of buildings and equipments. Relatively short term resources like good housekeeping, better insulation and improved heating conditions achieve 10-15 percent of energy savings. To achieve energy savings in the long term huge capital investments and long lead times are involved. Replacement of a country's industrial equipment stock may take from 20 to 30 years, for an example.

Hydroelectricity is presently the only renewable energy source that makes a substantial contribution to global energy needs. It can be expected to continue to provide about 5 percent of the world's energy need.

Geothermal sources provide useful quantities of energy in some areas today and additional sources are being discovered and developed. The total contribution from geothermal sources will nevertheless inevitably remain geographically limited and relatively small in relation to world energy requirements.

Conversion of Biomass to fuel alcohol is also under way in some countries, and it is already making a useful contribution to transport fuel requirements in Brazil.

Solar energy has probably the greatest long-term potential of the renewable resources but its rate of expnasion will be limited by the time required for new technologies to become economic and then penetrate energy markets.

Solar collectors for water heating, and in some cases for space heating, are already commercially available. About 2 percent or a little more, of the world's primary energy consumption could be supplied by the year 2000. Solar generation of electricity, whether by thermal methods or by photo-voltaics, is yet in its developmental infancy and so may require two to three more decades before it would make a strong impact in the world energy scene.

From the foregoing, it is to be noted that there is bright prospects for coal. The dwindling prospects for any substantial increase in the supply of oil at acceptable prices constitute the main reason for the increased importance of coal.

Even with the most optimistic forecasts for the expansion of nuclear power and the aggressive development of all other energy sources, as well as vigorous conservation, it is clear that coal has a vitally important part to play in the world's energy future. The world's coal production in 1977 was about 3,400 million metric tons of raw coal. This was a contribution of about 2,500 million metric tons of coal equivalent (mtce) or 33 mbdoe, already greater than any energy source except oil. Coal use has been projected to increase substantially from 1977 to 2000 A.D with almost a doubling or tripling increase.

1.6. WORLD COAL TRADE

In 1984 the world coal market was characterized by strongly expanding demand. The trend of growing expansion in world coal trade [25] as a consequence of the two oil crises of 1973 - 74 and 1979 - 80 is something which is unmistable. Economic recovery in many countries led to a growing demand for hard coal in the major coal consuming sectors. World-wide crude steel production grew by about 7 percent to 710 million tons in 1984. The more favourable market situation also had a positive effect on electricity generations and on the energy demand of all other industrial sectors.

The biggest demand on the world coal market was in Japan and in the European Community, accounting for three quarters of 1984 true world coal trade. Japan continued to be the largest purchaser with 86 mt imported coal and a market share of 27% in the total world coal market. This is largely attributable to the increase crude steel production there (coking coal having a 70 percent share in all Japanese imports).

On the supply side, the main coal exporting countries Australia, USA, Poland and South Africa predominate, accounting for almost three quarters of world exports (Table 13). For these countries the world coal market plays a crucial role as earner of foreign currency.

Table 13: Trend in hard coal exports(+) of the seven main exporting countries (in million tons).

Country	1973	1974	1975	1976	1977	1978
Australia	27	28	32	32	34	37
US	50	56	62	57	51	37
Poland	40	44	42	42	43	43
South Africa	2	2	2	6	11	15
USSR	31	32	32	32	34	31
Canada	11	11	12	12	13	14
FRG	27	35	25	22	22	31
Others	24	23	21	20	18	22
World coal) trade total)	212	231	228	223	226	230

Table 13 continued

Country	1979	1980	1981	1982	1983	1984(++)
Australia	40	42	51	49	61	76
US	61	86	101	98	71	75
Poland	44	33	17	31	37	45
South Africa	23	29	30	27	28	36
USSR	29	28	21	23	26	25
Canada	14	15	16	16	17	23
FRG	31	23	20	15	16	19
Others	25	26	31	27	23	19
World Coal) trade total)	267	282	287	286	279	318

+including coke converted to coal equivalent

++ Provisional

Table 14: Trend in World hard Coal (coke converted to coal) Imports

Import Countries Regions	1973		1983		1984	
	mill.t.	%	mill t	%	mill.t.	%
Japan	57	27	74	27	86	27
Third Country imports EEC	32	15	63	23	82	26
Rest World Trade	38	18	65	23	69	22
True World Trade	127	60	202	72*	237	75
Internal Trade EEC	30	14	22	8	23	7
Internal Trade Comecon	38	18	40	14	41	13
Exchange UK Canada	17	8	15	5	17	5
Internal Trade, Tonne	85	40	77	28*	81	25

In 1984 world coal trade reached a record figure of 317 mt (including coke converted to coal). This meant that the world trade volume in coal had risen 50 percent since the first oil crises (212)mt in 1973) by 105mt. The growth between 1973 and 1984 was exclusively in "true" world coal trade (Table 14).

---

*difference due to rounding up

**Provisional.

1.7. Coal Resources of Nigeria

The Mineral Survey of Southern Nigeria in 1909 for the first time discovered coal in Udi where mining commenced in 1915. Socio-economic activities which rejuvenated immediately after the second world war encouraged more active exploratory survey of the coal resources of Nigeria. Subsequently, the decade 1946 to 1956 saw the delimitation, quantification and characterization of sedimentary coal deposits. The areas of Udi, Enugu, Ezimo, Orukpa, Okaba, Ogboyoga, Ayangba, Aloko and Dekina indicated occurrence of coal deposits through their outcrops (Figure 6). The coal in these areas are referred to as Lower Coal Measures. It is more deep down and older in age than the coals of the Upper Coal Measures. The location of the Upper Coal Measures cover Abiriba, Ovim, Okigwi, Nsukka, Adoru, Akpanya, Ayangba, and Allomo. State-wise these are Cross River, Imo, Anambra and Benue States that house the coal deposits.

Lignites occur in Nigeria in Umu Duru, Okwele, Orlu, Umuchu, Nnewi, Onitsha, Asaba, Illah, and Ebu.

With 1 metre in thickness as the mineable cut-off the total coal reserve was quantified as 350 million tons [29].

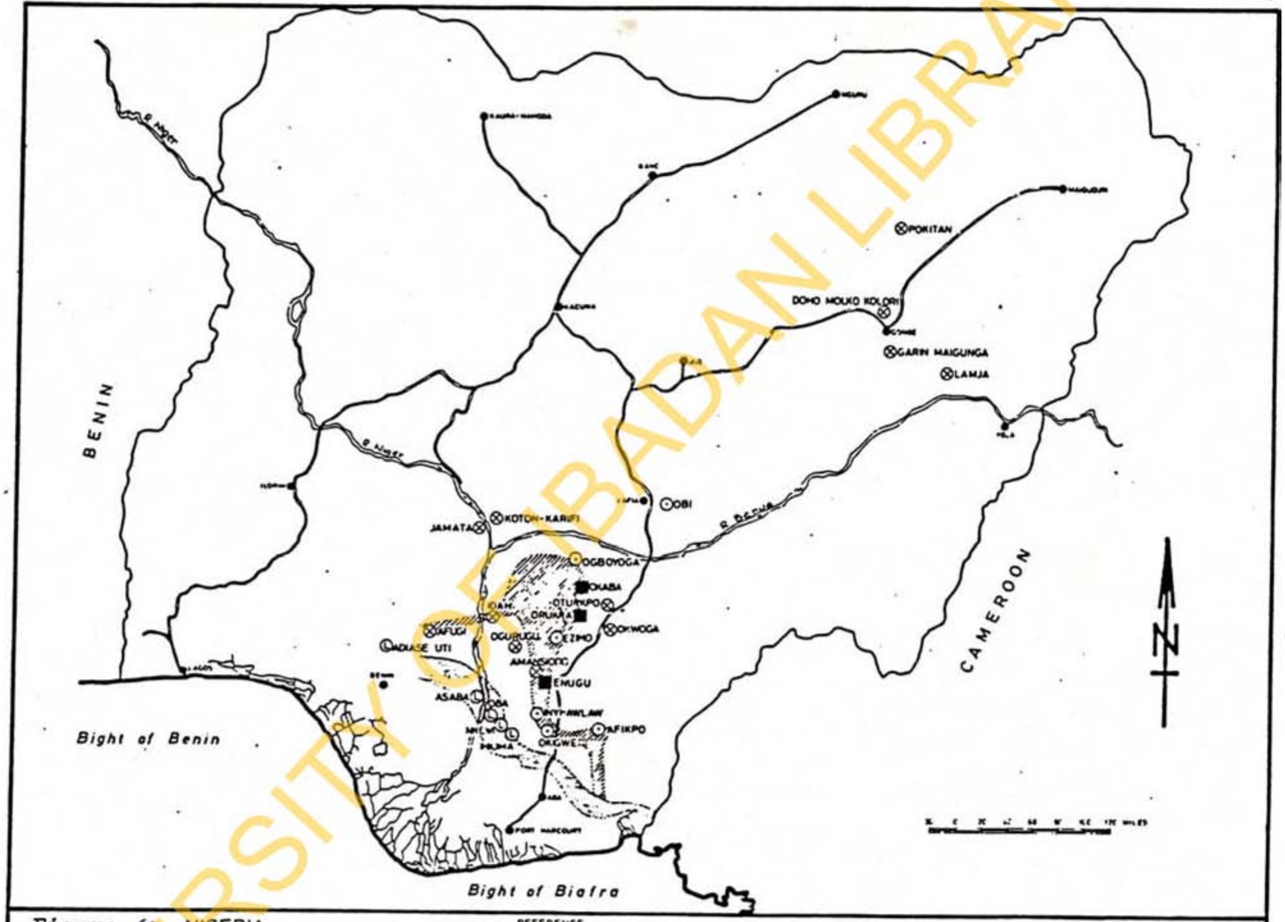


Figure 6: NIGERIA
COAL AND LIGNITE OCCURRENCES

Source. Geol. Surv. Nigeria & Nigerian Coal Corp.

The reserve was measured by the drilling of 123 boreholes different depths. Two boreholes reached (330m) deep while the others were each less than that.

Powell Duffryn Technical Services Limited [29] at that time carried out tests and analyses to determine the chemical and physical characteristics of the drilled core samples. Proximate and ultimate coal analysis were carried out together with microscopy and technological property examinations.

Nigerian coals are of the geologically young Upper Cretaceous and Tertiary age. Enugu, Onyeama, Obwettie, Orukpa, Okaba, and Ogboyoga coals were formed in their swamps about 70 to 76 million years ago in the period called Maestrictian/Campanian epoch of the Upper Cretaceous.

The Upper Coal Measures were formed about 65 million years ago in the Palaeocene epoch of the Tertiary period. The youngest of it all is the lignite sediment which were deposited only about 26 million years ago at the miocene epoch, also of the Tertiary period.

1.7.1. Geomorphology

Three main physiographic units can be recognized in the coal bearing areas of Nigeria. These are the Benue and Cross River plains, the Enugu escarpment and the Udi and Igala plateaus.

The sedimentary cycle in south-eastern Nigeria began with the deposition of a thick series of marine shales, with subordinate sandstones and limestones, ranging in age from Middle Albian to Uppermost Albian, and in places to Cenomanian. Deposition eventually became very slow or ceased altogether as a result of regression of the sea and the sediments were strongly folded. In Turonian time the sea again invaded large areas, and marine sedimentation continued well into the Senonian. This was followed by uplift and renewed folding, and erosion in certain areas.

In Campanian-Maestrichtian times another transgression of the sea took place and marine sediments were laid down. In some places these rest unconformably on the underlying beds while in others there appears to have been no break in sedimentation.

The sea became gradually shallower, this led to the formation of an extensive, low lying coastal area with lagoons and swamps, on which a thick succession of coal measures and fresh-water sandstones were deposited, transgressing to the North and West on to the Pre-Cambrain basement. Sedimentation was entirely non-marine until the deposition of the Upper Coal Measures, in which marine intercalations indicate a gradual encroachment of the sea. Sedimentation was again marine during the Paleocene and Lower Eocene, but during Miocene times there was widespread emergence of the coastline with the re-establishment of conditions favourable for the deposition of fresh-water sediments, carbonaceous beds and lignites.

1.7.2. The Lower and Upper Coal Measures of Nigeria

The Lower and Upper Coal Measures are the deeper and shallower coal seams respectively. The deeper or lower seam was formed first before the shallower or upper coal layer.

(a) The Lower Coal Measures

The Lower Coal Measures comprise alternating sandstone, shales, sandy shales and mudstones, with coal seams or carbonaceous shales at various horizons (Figure 8). West of Enugu there is a characteristic pattern in the sequence of rock types, marked by units which are repeated at least five times, which suggests repeated uplift and subsidence of the land during deposition. A typical vertical section from lower down, say, is

5. Shale or sandy shale, etc.
4. Coal, sometimes shaly at top.
3. Carbonaceous shale, passing downwards into shale.
2. Sandstone, with a few shaly layers or alternating sandstone and shales.
1. Shale or sandy shale.

The sandstones in each unit vary considerably in thickness and sometimes rest directly on the coal. The coals and shales may die out laterally in any direction and re-appear further on. The lithology of the Lower Coal Measures remains remarkably constant from Enugu to Ogboyoga and from there west-wards to the Dekina area.

South of Enugu, however, towards the Abakaliki - Okigwi anticlinorium, shales and carbonaceous sediments gradually decrease in importance and where it crosses the structure the formation is indistinguishable from the underlying Awgu sandstone, and contains only a few thin carbonaceous horizons and no coal seams.

(b) The Upper Coal Measures

The Upper Coal Measures rest comformably on the False-bedded sandstone. North of Okigwi in Imo State and in Udi and Nsukka Divisions in Anambra State, the lithology of the formation is similar to that of the Lower Coal Measures. The main rock types are sandstone, sandy shale and carbonaceous shale. Gritty and pebbly bands are locally present in the sandstones, and the shales, which are on the average much darker than those of the Lower Coal Measures, contain many concretions, bands, and stringes of marcasite and siderite. In Nsukka Division there is a persistent basal sandstone horizon

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averaging about 16.6m in thickness. This is followed by about 10m of shales, which contain most of the known coal seams.

The ferruginized equivalent of these shales forms the capping of the many flat-topped hills on the Udi plateau. The shales are overlain by another sandstone horizon, from 10 to 23 metres thick, and this is succeeded by a great thickness of dark shales with many sandstone beds. In Udi and Awgu Divisions, the two lowest units become indistinguishable, and are represented by about 20 metres of alternating sandstone and shale.

In a few areas minor folds have been superimposed on the limbs of the major structures. An example of this occurs at Ogboyoga, where the drilling has proved a small, low anticline near the axis of the Anambra syncline. In the Enugu colliery workings, No 3 seam shows a series of broad undulations. These are probably the result of differential setting during the consolidation of the beds and not the effect of subsequent deformation.

Faults with displacements ranging from a new centimetres to over 61 metres are present in the coal-bearing formations. Several have been recognized at Enugu and Inyi, and there are also indications of small scale faulting at Orukpa.

The coal deposits of Nigeria will now be looked at closely in order to obtain information that would help in exploitation and utilization of this resource.

1.7.3. COAL SEAMS IN THE LOWER COAL MEASURES

Enugu, Ezimo, Orukpa, Okaba and Ogboyoga are typical examples of coal in this category and some coal exposures in streams near Enugu occur in Ofam, Nveni, Nyaba, Olawba, Asata, Ogololo, Atoto-Ebedde, Obwetti, Obokwa, Discovery Creek, Iva, Kraal Creek, Prison Creek, tributaries of Prison Creek and Ekulu streams, Obuya stream, Eva or Afa stream and stream 213.4m north of beacon CP12 [29]. These streams drain the Enugu escarpment which rises about 233.3m above sea-level in some places. From borehole drilled cores it was understood that there are five persistent coal seams in Enugu area which are known as No's 1 to 5 from the bottom to the top. No.3 seam is the thickest seam in Enugu area, and it is the only coal at present being worked.

West and South of Enugu No.3 seam consists of two coals of roughly the same average thickness separated by from 50mm to 300mm of shale or sandy shale, the so-called "dirt band". The seam is 1.4metres in thickness, and passes downwards into carbonaceous shale, sandy shale or sandstone and the junction with coal is usually sharp. The dirt band grades into the upper coal but these lower junction is always well defined. The band is absent in certain parts of the workings. Its position is usually taken by a thin band of durain, sharply differentiated from the coal below, but where this is absent the two coals become indistinguishable.

The annual production of coal from Enugu collieries from 1916 to 1980 is shown in Table 15. Mining in Enugu collieries is by the pillar and stall method. The seam is worked "on board" with the face parallel to the cleat. This facilitates the breaking off of large slabs.

TABLE 15: ENUGU COAL PRODUCTION (1916-1980)

Year	Production (long tons)	Year	Production (long tons)
1916	24,511	1946-47	633,852
1917	83,405	1947-48	551,706
1918	145,407	1948-49	610,283
1919	137,844	1949-50	526,613
1920	180,122	1950-51	583,487
1921-22	194,073	1951-52	566,393
1922-23	112,818	1952-53	613,374
1923-24	175,137	1953-54	679,437
1924-25	220,161	1954-55	675,918
1925-26	242,582	1955-56	750,058
1926-27	353,274	1956-57	790,030
1927-28	345,303	1957-58	846,526
1928-29	363,743	1958-59	905,397
1929-30	347,115	1959-60	684,800
1930-31	317,681	1960-61	565,681
1931-32	263,548	1961-62	596,502
1932-33	259,860	1962-63	615,681
1933-34	234,296	1963-64	600,230
1934-35	258,893	1964-65	698,502
1935-36	257,289	1965-66	730,183
1936-37	310,308	1967-69	Civil War
		1970	11,703
1937-38	391,159	1970-71	24,000
1938-39	323,266	1971-72	176,000
1939-40	300,090	1972-73	334,940
1940-41	328,594	1973-74	340,972
1941-42	402,640	1974-75	277,753
1942-43	463,978	1975-76	270,935
1943-44	528,421	1976-77	282,727
1944-45	668,158	1977-78	267,158
1945-46	505,565	1978-79	204,837
		1979-80	140,487

Source : Geol. Survey Nigeria & Nigerian Coal Corporation

The roof of the seam is strong and stands up well for long periods without support. Most of the coal is hand-hewn and the high extraction rate of 98 percent is obtained. Efforts, however, are on to mechanize the coal mining.

Ezimo Coals

Ezimo lies at the foot of the Enugu escarpment and it is approximately 64km north of Enugu. Coal outcrops in the Ezimo area could be seen in streams like Iyinzu, Iyiocha, Iyiozuma, Oshenye, Ogele, Ikpeke, Aboine and Enerve. The main seam at Ezimo outcrops for a distance of about 25km along the escarpment. In the sourthern and central part of the area the coal is from 1.2 to 1.5metres thick, but north of the Oshenye stream a parting develops in the seam. In the Enerve the seam has thinned to 0.3metre. The coal is friable and partially weathered and is overlain by ferruginous sediments.

Orukpa Coals

Orukpa village is about 96 Kilometres north of Enugu by road. The Orukpa seam outcrops west of the town, near the base of the Enugu escarpment. Coal outcrops in Orukpa area show along streams like Addu, Iyari, Ojeche, Achimodo and Akpawku. At outcrops the seam is from 0.19 to 2.4metres thick.

Okaba Coals

At a point 7° 28', latitude North of the Equator and 7° 45' longitude East of Greewich in Nigeria situates the town of Okaba located in Ankpa local Government area of Benue State[29]. Here the Igala plateau which is about 500m high has a steep slope through a range of about 100m and then slopes gently for about 3km Eastward to a height of 300m where Okaba situates 16km from Ankpa town. The area is Igala speaking. Enugu escarpment rises from here. An outcrop of Okaba coal was discovered in 1930 in the Otokpa stream. Pitting revealed the deposit to be 2.3m in thickness. Between September 1954 and February 1955 seventeen boreholes were drilled and all intersected the seam.

The depths of the boreholes range from 12.8m to 131.8m. The coals are overlain sharply at the top and bordered at the base by progressive gradation and carbonaceous shale, shale or sandy shale. The reserve is estimated at a total of 74.2million tons.

Ogboyoga Coals

Ogboyoga is situated in hilly country where the trend of the Enugu escarpment and the Lower Coal Measures outcrop changes from north-south to east-west. The escarpment is much indented by steep-sided valleys. It was in 1930 that coal was first discovered in this area and later in 1950 many more outcrops were located near Ogboyoga and further south around Odokpono. Streams like Ocakuca, Ede, Edili, Ajuna, Ikelu, Ucomo, Agbajitodo, Umutayaji, Orikpi, Odokpono, Ajafo, Ebene and Adawa revealed the outcrops at different locations. In most of the area investigated the coal is over lmetre thick. There is no evidence of large scale faulting, but small irregularities in the seam contours in the north-east and near Odokpo stream may be the expression of minor faults.

Nigerian Coal reserves (in million tonnes) in some of the areas described in the foregoing is indicated in Table 16.

Table 16: Nigerian Coal reserves (in million tonnes)

Nigerian Coal Corporation Mines	Indicated reserves	Inferred reserves
Enugu	42.7	120.2
Ezimo	29.5	17.3
Orukpa	50.9	70.1
Okaba	54.9	190.3
Ogboyoga	83.3	310.5
Inyi	10.3	- -
TOTAL	271.6	707.4

1.7.4 Coal Seams In the Upper Coal Measures

The coals of the Upper Coal Measures show considerable variation in thickness and quality along the strike, especially in the Awlaw-Owerri area. A pit on the bank of the Iyiofia stream near Owerri penetrated a seam with a very irregular **roof.** In the small section exposed the coal is from 1.2 to 1.5metres thick and the presence of irregular coal fragments in the overlying sandstone suggests a local uncomformity. In the Omuma stream three exposures of the same seam, a small distance apart are respectively 1.7, 0.6, and 0.8 metres thick. Impersistent bands of shaly coal and shale are also common in the seams.

Inyi Coals

The main coal seam in Inyi is extremely variable both in thickness and in quality. Sandstone usually rests directly on the coal, but a thin layer of shale sometimes intervenes. The floor of the seam nearly always has shales.

1.7.5. Other Coal Formations

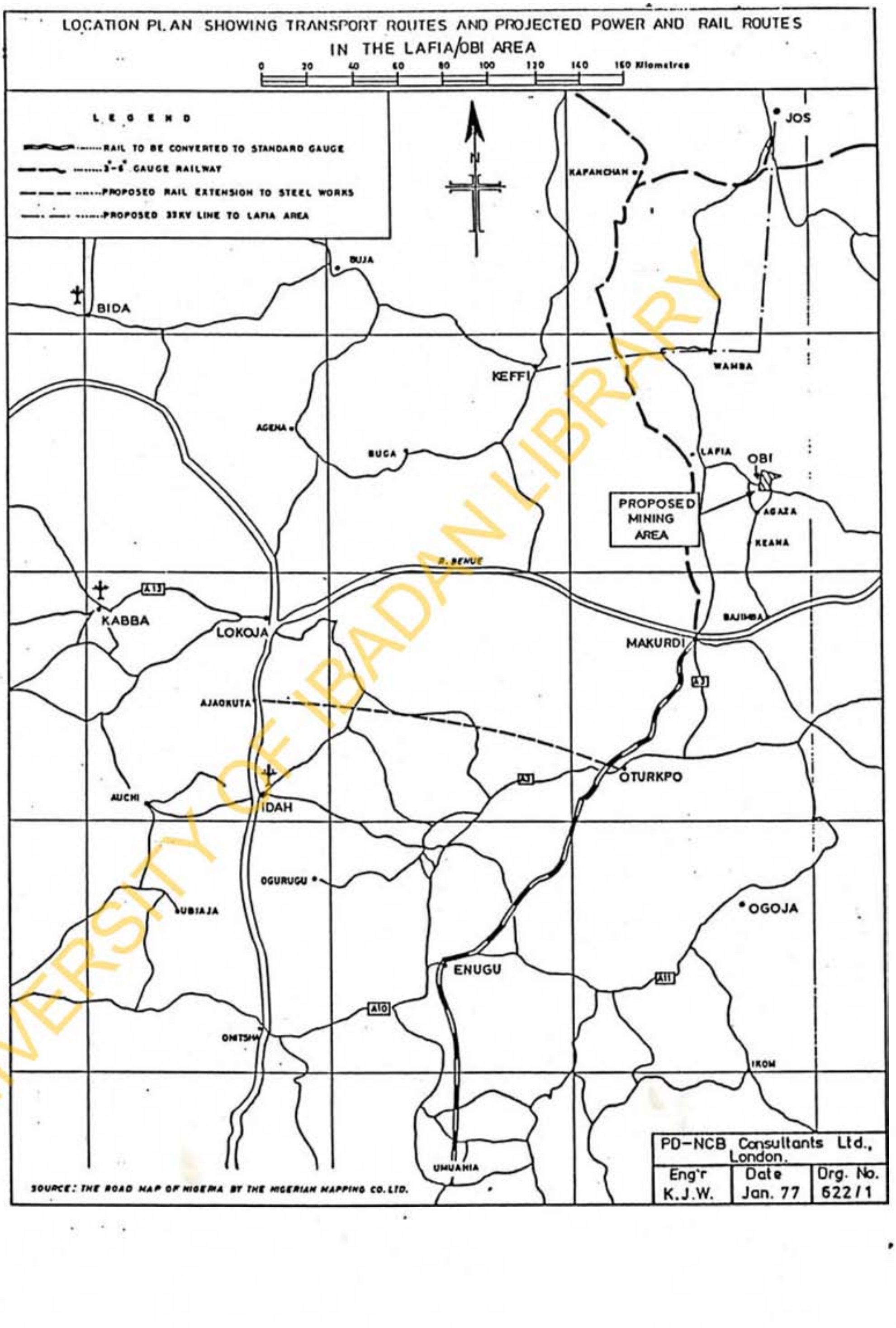
As far back as the early part of this century it was known that coal outcrops and subsequent pitting revealed occurrences of coal in Okwoga (Benue State), Lafia (Plateau State), Jamata, Agbaja Plateau, Koton Karfi (Kwara State), Doho, Kurnu-Pindiga - Putuk (Bauchi State), Lamja, Lediye, Chikila, (Gongola State) and Gindi akwati (Plateau State). They were observed however, to be tertiary coals or of the uppermost cretaceous age-like those of the Lower Coal Measures.

Afuji (Bendel State) and Ute (Ondo State) have shown coal-outcrops that indicate good qualities of the coal.

1.8. Geology of Lafia Coal Deposit

Lafia town lies 08.30N latitude and 08.34E longitude which is some 85km North of Makurdi and about 150km South-southwest of Jos, the State capital. Lafia coal deposit extends in a broad belt 30 kilometres wide to the south-east of Lafia town (Figure 7). The deposit may overall cover an area of approximately 2,500 square kilometres.

Figure 7



LOCATION PLAN SHOWING TRANSPORT ROUTES AND PROJECTED POWER AND RAIL ROUTES IN THE LAFIA/OBI AREA

SOURCE: THE ROAD MAP OF NIGERIA BY THE NIGERIAN MAPPING CO. LTD.

Detailed investigations have been restricted to an area covering 50km^2 around the village of Obi, which itself lies in the south-eastern part of the deposit (Figure 8).

The deposit is part of a vast synclinorium with the coal-bearing measures contained within this structural basin, along with other rocks of the Cretaceous age. The whole succession is folded into numerous synclines and anticlines which have their axes orientated in a general NE - SW direction. To the north-west of the basin, the Cretaceous rocks out-crop against the Precambrian Basement, and to the south-east, the structure changes to an anticlinorium composed of rocks lower down in the Cretaceous succession than the coal-bearing measures.

The Obi area is located on one of these groups of folds within the basin. The Giza anticline represents the Central structural feature, and it was on the crest of this that drilling for core-samples was intensified (Figure 8).

The area covering the deposit is relatively flat, varying in evaluation from 120m around the numerous seasonal streams that intersect the area to 250m above sea-level on the areas of higher ground associated with outcrops of sandstone which lie within the sequence. A well defined, very low escarpment created by sandstones older than the coal-bearing measures forms the south-eastern limit of the deposit.

In the western half of the deposit, the main drainage directions is to the south, with both seasonal and persistent water courses draining into the River Guma, which itself joins the River Benue east of Makurdi. To the East, the River Biu, supplied by numerous seasonal streams, flows in an easternly direction to join the River Dep. The area is prone to poor drainage during the rainy season, particularly away from the limited sandstone ridges, (Figure 8) near the seasonal streams.

1.9. Stratigraphic Seccession of Lafia Coal

The coal-bearing measures are represented by predominantly areanaceous and argillaceous sediments of the Upper Cretaceous age. The coal measures are about 550m thick and contain at least 36 coal seams of variable thickness, quality, and lateral extent. The coal bearing succession is over-lain by a marine argillaceous series of 1000m which, in turn, is overlain by a thick areanaceous sequence, the Lafia sandstone. Beneath the coal measures the sequence has not been proved by drilling but it is though to consist of a thick section of Keana sandstone.

Arufu limestone and the Uoma formation of arenaceous/ argillaceous measures. These are assumed to rest uncomformably on the Pre-cambrian basement (Figure 9). The coal seams are concentrated much deeper down within the measures. A zone of maximum coal occurrence of 60m has been identified in the lower part of the section and it is this location that contains the coal seams which are though to be of commercial value. It is primarily in this zone, around Obi area that detailed exploration was intensified.

Seams 12, 13, 18, 19, 20 and 24 were considered in this Ph.D work as deserving further attention and examination because of their thickness, persistence and macroscopic visual quality appearance. The choice of Lafia/Obi Coal otherwise referred to as Lafia Coal in this study is explained later on in this thesis.

1.10. Exploration of Lafia Coal Deposit

The search for coking coal started in 1973 in and around Lafia and Obi towns. It started with a general search for outcrops of coal in the locality. Before long, a major group was found in the valley of the River Dep in the extreme east of the area (Figure 8).

Figure 8: GEOLOGICAL PLAN OF LAFIA AREA WITH BOREHOLE LOCATIONS OUTSIDE THE OBI AREA

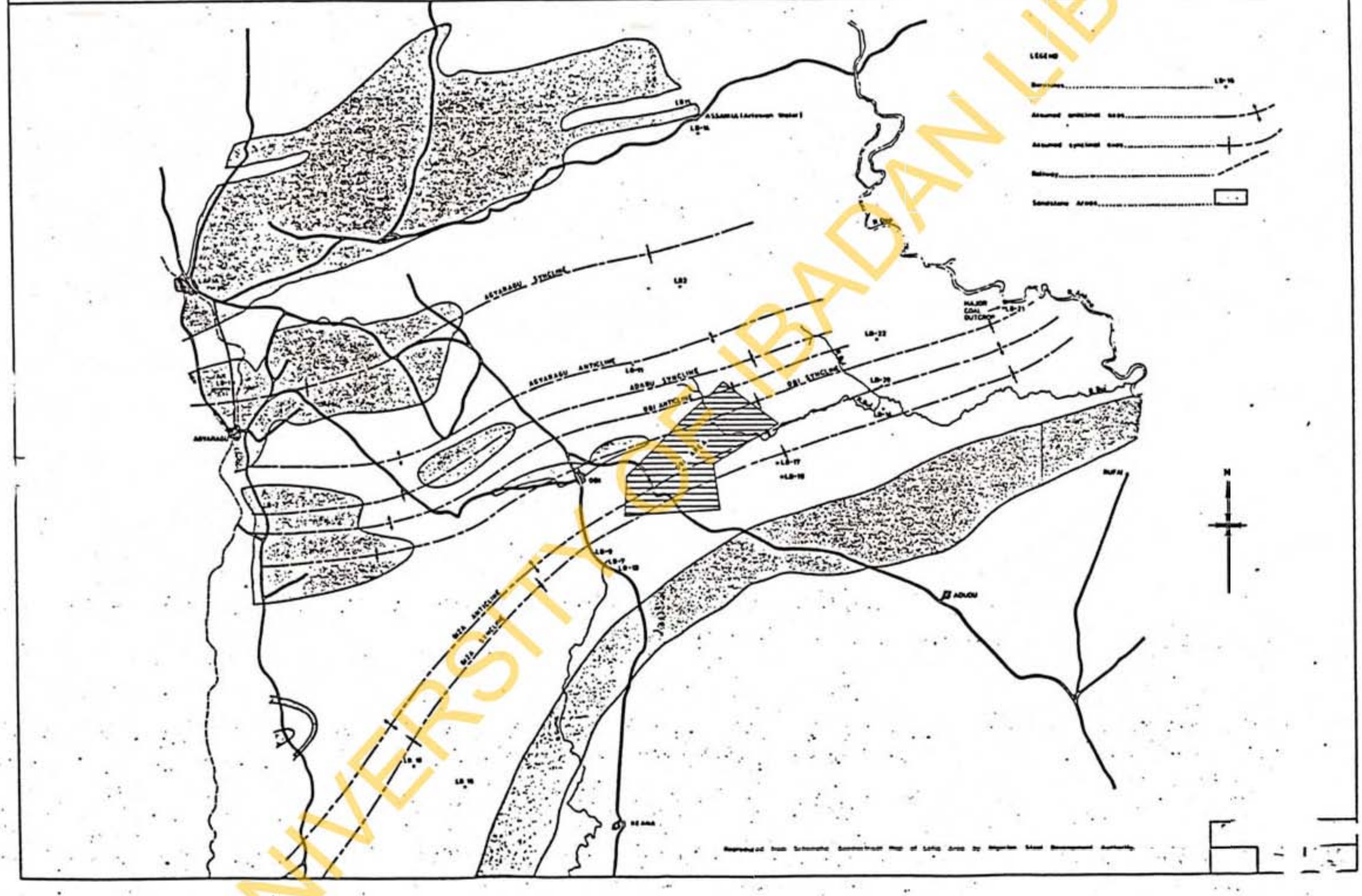
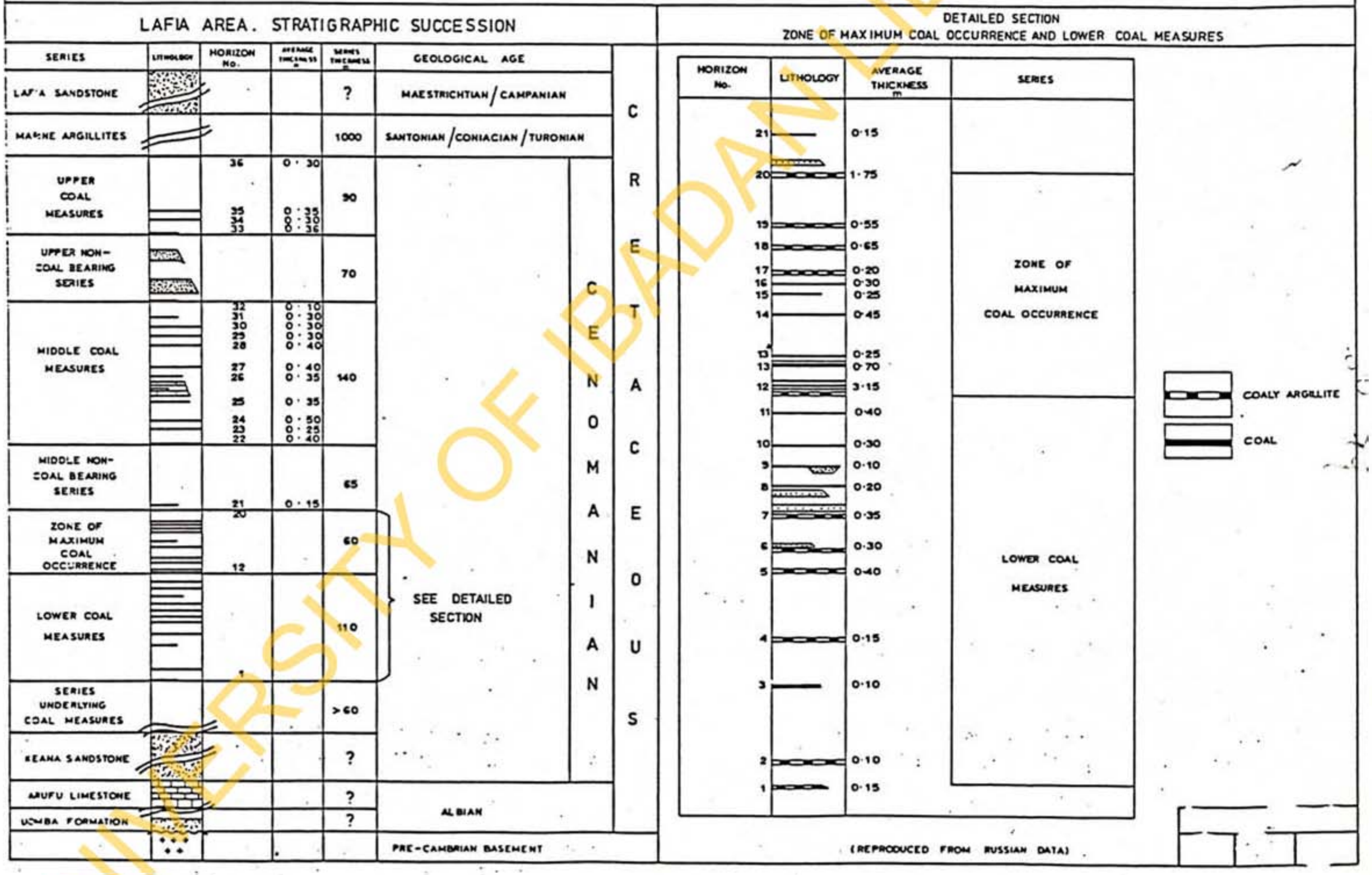


Figure 9: LAFIA COALFIELD STRATIGRAPHICAL SUCCESSION



LAFIA AREA. STRATIGRAPHIC SUCCESSION

SERIES	LITHOLOGY	HORIZON No.	AVERAGE THICKNESS m	SERIES THICKNESS m	GEOLOGICAL AGE		
LAFIA SANDSTONE				?	MAESTRICHTIAN / CAMPANIAN		CRETACEOUS
MARINE ARGILLITES				1000	SANTONIAN / CONIACIAN / TURONIAN		
UPPER COAL MEASURES		36 35 34 33	0·30 0·35 0·30 0·36	90		CENOMANIAN	
UPPER NON-COAL BEARING SERIES				70			
MIDDLE COAL MEASURES		32 31 30 29 28 27 26 25 24 23 22	0·10 0·30 0·30 0·30 0·40 0·40 0·35 0·35 0·50 0·25 0·40	140			
MIDDLE NON-COAL BEARING SERIES		21	0·15	65			
ZONE OF MAXIMUM COAL OCCURRENCE		20 12		60	SEE DETAILED SECTION		
LOWER COAL MEASURES		1		110			
SERIES UNDERLYING COAL MEASURES				>60			
KEANA SANDSTONE				?			
ARUFU LIMESTONE				?	ALBIAN		
UOMBA FORMATION				?			
					PRE-CAMBRIAN BASEMENT		

DETAILED SECTION
ZONE OF MAXIMUM COAL OCCURRENCE AND LOWER COAL MEASURES

HORIZON No.	LITHOLOGY	AVERAGE THICKNESS m	SERIES
21		0·15	
20		1·75	ZONE OF MAXIMUM COAL OCCURRENCE
19		0·55	
18		0·65	
17		0·20	
16		0·30	
15		0·25	
14		0·45	
13'		0·25	
13		0·70	
12		3·15	
11		0·40	LOWER COAL MEASURES
10		0·30	
9		0·10	
8		0·20	
7		0·35	
6		0·30	
5		0·40	
4		0·15	
3		0·10	
2		0·10	
1		0·15	

(REPRODUCED FROM RUSSIAN DATA)

The whole of Lafia area was then surveyed using vertical sounding techniques to delineate potential areas represented by argillaceous successions. Earlier drilling in the western portion of the area which had arenaceous sequence gave no useful result other than that the search should continue and be re-located. Obi area gave encouraging coal-bearing drilled cores and hence efforts were concentrated here. About 100 boreholes were drilled in Obi area while about 17 were scattered around Lafia. Two shafts were sunk to depths of 28m and 32m respectively for the purposes of obtaining large samples for pilot scale tests. The deepest borehole reached 520m while others are much shallower. The spacing of boreholes external to the obi area was generally less than 1 per 50km^2, but within the area it rose to a maximum of about 8 per km^2 on the crest of the Giza anticline (Figure 8).

The boreholes sunk in Lafia area reached only about 350m depth because of the limited capacity of the drilling rigs available at that time (Figures 10 and 11). Boreholes LB-2, LB-3, LB-9, LB-10, LB-11, LB-15, LB-18, LB-20 and LB-22 showed no coal. Only traces of coal were shown by boreholes LB-14 and LB-17. In borehole LB-1, two thin coal horizons were intersected, but it was not possible to correlate them in any way to the 36 seams discovered later.

Only in boreholes LB-7, LB-13, LB-16, LB-19 and LB-21 were recognizable seams intersected. Borehole LB-3, which was located about 8 km south-south-east of Obi Village, on the road to Agaza, went down to 225m depth, intersecting seams 32, 31, 26, 22 and 21. None of these seams were of any commercial value.

In borehole LB-13, located close to LB-7, being less than 1km to the south-east, the position of the borehole on the anticline was more fortunate. The hole intersected the "zone of maximum coal occurrence", but unfortunately, the section of seam 12 which later proved of commercial value in Obi area was disturbed by faulting and was therefore difficult to evaluate at that time.

Borehole LB-16 located approximately 12km west of the village of Keana, once again intersected seams 13 and 12 in the zone of maximum coal occurrence. Here the section of the seam 13 was very thin and that of seam 12 thick at 2.85m, but very dirty, and along with the remaining seams, can be considered of little commercial value. This also applied in general terms to borehole LB-19, located 14km east of Obi village, where once again no horizons of commercial value were recorded.

Borehole Lb-21 was drilled at the extreme north-eastern part of the deposit near the River Dep where outcrops of coal had been noted. Here seams 13 and 12 appear to be of the same workable thickness, at 0.50m to 0.55m

1.11. Contours of Coal-seam floor level for reserve evaluation

In the majority of boreholes drilled in the area, accurate checks were carried out on coal-horizon floor levels using down-hole logging equipment. Contouring of these levels was done on 50m intervals, based on the assumption that the throw and position of the major faults are proven. In areas of low borehole density, (i.e. less than 1 hole per 10km^2) where, because of depth, drilling did not reach the main coal horizons, contouring was obviously very approximate.

Based on the limitations of approximate fault position orientations and lateral extents, a tentative structure delineating structurally bounded blocks of coal has been prepared for use in calculating geological and workable reserves in seams 12 and 13 (Figures 10 and 11 and Tables 17 and 18). With regard to faulting, a number of assumptions have been made which provide the most optimistic picture and the maximum reserve figures. The estimates of geological reserves are given in Table 17 whilst Table 18 gives the workable and recoverable reserves.

Figure 10: - ISOPACHYTES OF SEAM 12

COAL IN WORKING SECTION

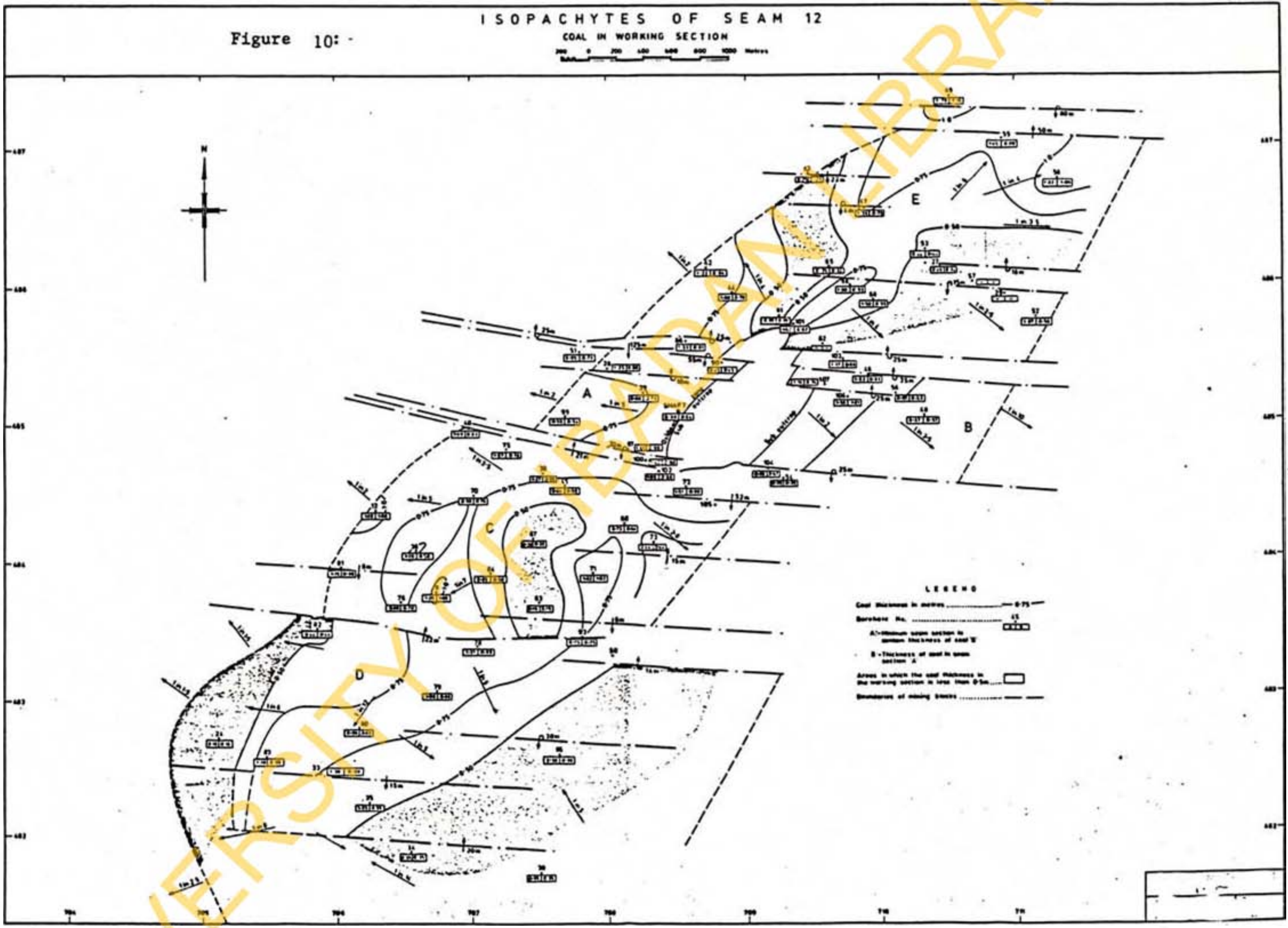


Figure 11: ISOPACHYTES OF SEAM 13

COAL IN WORKING SECTION

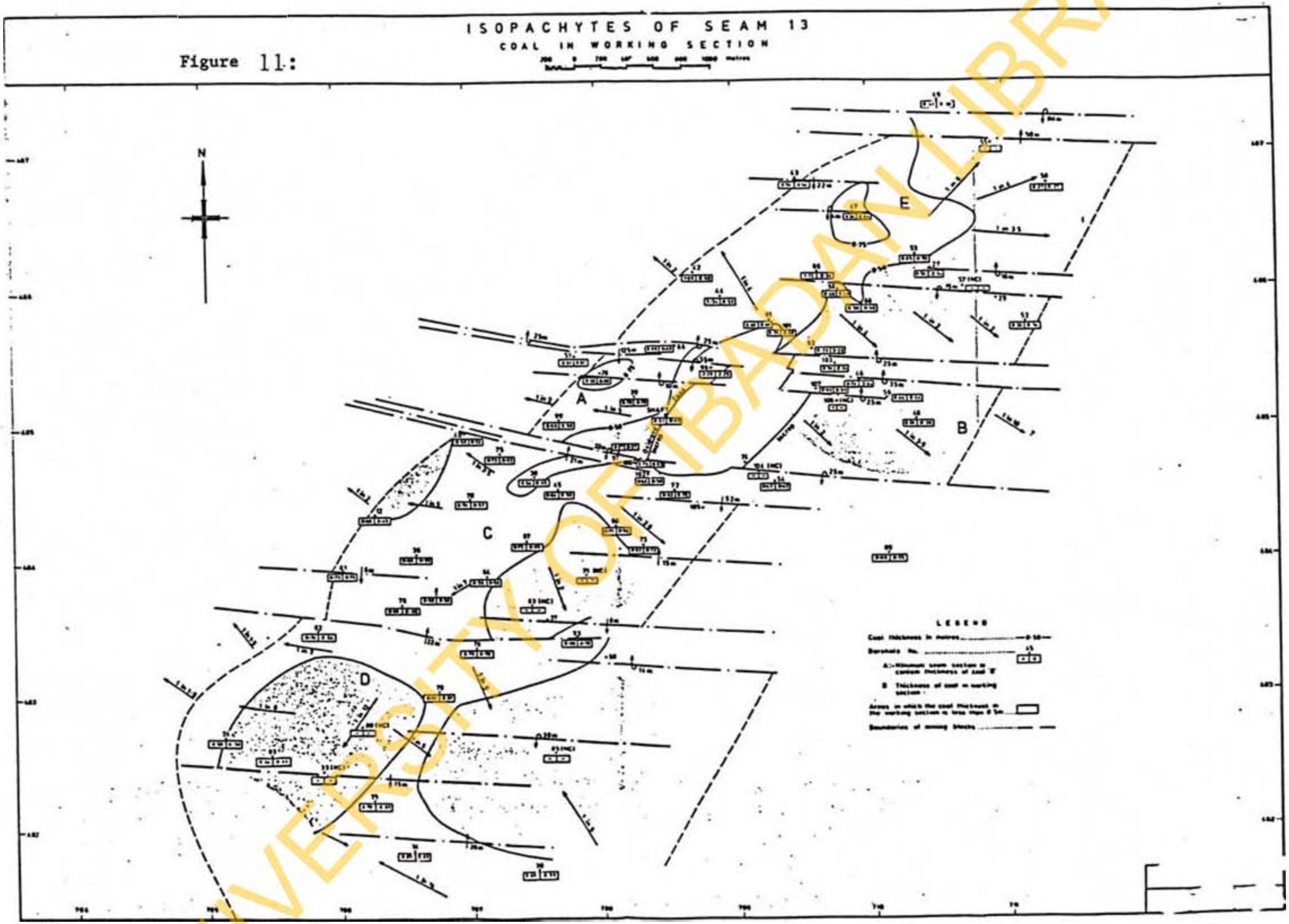


Table 17: GEOLOGICAL RESERVES OF SEAMS 12 AND 13

(tonnes)

Block	Total Coal Thickness				Total
	<0.5 m	0.5 - 1.0 m	1.0 - 1.5 m	1.5 - 2.0 m	
Seam 12					
A	-	81,600	828,000	288,000	1,198,000
B	-	752,400	-	-	752,400
C	80,600	920,400	3,854,000	811,500	5,666,500
D	699,500	3,694,800	5,686,000	-	10,080,300
E	99,200	2,257,200	3,200,000	-	5,556.400
Total	879,300	7,706,400	13,568,000	1,099,900	23,253,600
Seam 13					
A	-	593,000	-	-	593,000
B	407,700	-	-	-	407,700
C	286,100	2,061,000	-	-	2,347,100
D	758,400	2,394,000	-	-	3,152,400
E	1,335,700	1,487,000	-	-	2,822,700
Total	2,787,900	6,535,000	-	-	9,322,900

Note: Total geological reserves = 32.6 million tonnes, which ar present can only be classified as "inferred". This classification is the same as that given in v/o Technoexport's report on the Lafia Coal-field,November 1976, ie. Class C.

Table 18: WORKABLE AND RECOVERABLE RESERVES OF SEAMS 12 AND 13
(tonnes)

Block	Total Coal Thickness			Total	
	0.5 - 0.75 m	0.75 - 1.0 m	>1.0 m	Workable Reserves	Recoverable Reserves*
Seam 12					
A	235,500	518,800	-	754,300	490,300
B	774,700	-	-	774,700	503,600
C	1,065,000	1,908,700	-	2,973,700	1,932,900
D	1,567,000	1,401,200	-	2,968,200	1,929,300
E	1,100,000	1,057,300	57,600	2,214,900	1,134,800x
Total	4,742,200	4,886,000	57,600	9,685,800	5,990,900
Seam 13					
A	379,000	73,200	-	452,200	-
B	-	-	-	-	-
C	1,618,600	-	-	1,618,600	-
D	1,481,300	-	-	1,481,300	-
E	1,688,400	141,800	-	1,830,200	428,900x
Total	5,167,300	215,000	-	5,382,300	428,900

x Recoverable reserves in part of Block only.

* Recoverable reserves based on 65% recovery factor.

Note: Total recoverable reserves = 6.42 million tonnes, which at present can only be classified as "inferred".

1.12. HISTORY OF GOVERNMENT EFFORTS ON THE DEVELOPMENT OF NIGERIAN COAL INDUSTRY

From the very early part of this century the successive governments of Nigeria has given attention to coal, having recognized its role as an important energy source.

1.12.1. Geological Survey of Nigeria (GSN)

The Geological Survey of Nigeria (GSN) has been an active and result-oriented organ of the Government on mineral exploration and survey of the natural resources of the country from the beginning of this century. In 1903 and 1905 respectively the GSN recorded exposures of lignite near Okpanam and Ibusa in the Asaba Division of the former Western region of Nigeria and near Moroko, some 16 kilometres south of Abeokuta, in the same region [30]. Lignite was discovered also in Nnewi and Oba area about 18 kilometres South East of Onitsha. The Mineral Survey of Southern Nigeria formed in 1903 discovered bituminous coal in 1909 in the Udi district of former Eastern region of Nigeria while proceeding on a route from Onitsha to Abakaliki and thereafter interest in the lignite diminished; virtually ceasing after coal production commenced in Enugu in 1915.

Field indication of coal type and rank was obtained from the ratio of fixed carbon(c) to the volatile matter content (v) of the coal.

The interpretation [30] given to the c/v ratio was such that for anthracites $c/v > 20$ and for lignites $c/v < 0.5$. On this scale, the values for Enugu coal are 1.1 to 1.0, the upper coal measures have 1.0 to 0.8, and lignites, generally, 0.75 or less. The results of these earlier efforts to explore and exploit the coal resources of Nigeria have been amply documented by the publications of the Federal Ministry of Mines, Power and Steel including the Geological Survey of Nigeria bulletin No.28.

1.12.2. Nigerian Steel Development Authority (NSDA) And Tiajpromexport (TPE) of the Soviet Union

The physico-chemical characteristics of the metallurgical coke that would be used in Ajaokuta Steel Company Limited had been defined in the project design prepared by Tiajpromexport who are the technical partners [31, 32]. NSDA/TPE therefore started to investigate the possibility of using Nigerian coals in Ajaokuta Steel Plant. Shaft sample of Lafia coal, run-of-mine of Enugu coal and foreign coals were carbonized as a multicomponent blend at the pilot scale level.

The coke produced was satisfactory in many respects except that it was too high in ash and sulphur for the designed steel process. Furthermore, the exploratory work done on Lafia coal deposit by the drilling of many boreholes revealed that the field had both normal and reverse faults. As such, the mineability question arose for mining engineers to answer and consequently an in-seam mine-ability test has been recommended for the deposit.

1.12.3. British Carbonization Research Association (BCRA) Tests

In December 1976, a United Kingdom based Company, BCRA (Chesterfield, U.K.) tested Nigerian coals [33]. Enugu Onyeama seam 3 was valued non-coking but could be a 10% or less component of a coking blend especially judging from its low ash. A float-and-sink test on Lafia seam 12 coal at specific gravity (S.G) of 1.4 gave 61% yield with 17.6% ash content.

1.12.4. Eisenbau Essen GmBH Test

Industrial scale Experiment of coking coal for the steel Industry was contracted to Eisenbau Essen GmBH of West Germany [31] by the Steel Development Department in 1980. The target of the study was to produce a metallurgical coke out of a coking blend with a maximum content of Nigerian coals and coal products. The results of the test was expected to produce sufficient qualitative data on coking coal blends that would suit the coke oven plant being erected in Ajaokuta for the manufacture of metallurgical coke of the characterisitcs stipulated by the Tiajpromexport designers. The study was done by carrying out laboratory analysis, pilot and industrial scale carbonizations tests and computer calculations based on the theory of Prof.Dr. rer.nat. W. Simon's, Technical University of Berlin.

The results of laboratory analysis, pilot scale tests, industrial scale tests and various computer calculations gave an optimised blend containing maximum content of 30% of Nigerian coal and coal products in the ratio: Enugu washed fine coal 20%, coke breeze 3%, carbo pitch 7%, Imported coal E type 20%, Imported coals A,B,C, and D 50%.

The report also considered the use of preheating of the coking blend in which case up to 38.5% of Nigerian coal and coal products would be accommodated in the coking process.

The composition of the blend would then be: Enugu washed fine coal 25%, Imported coal E type 20%, Imported coals, A,B,C, and D-types 41.5%, coke breeze 7.5%, and carbon pitch 6.0%.

According to the report imported coals, A,B,C, D and E-types are available in the world market on a long term contract. Coke breeze can be recycled from the high temperature coking plant. A part of the required pitch can be produced by distillation of the high temperature tar. The rest of the carbon pitch can be imported or otherwise produced from low temperature carbonization of Okaba run-of-mine coal.

The result also proposed an enlargement of the coke oven chamber width at Ajaokuta plant from 410mm to 450mm. Such a measure is expected to increase the throughput by about 10%. If preheating technology is combined with expansion of the oven chamber, then an over-all throughput increase of 34% was projected. These measures were costed and substantial savings in foreign currencies were forecast if the chamber is widened and if preheating was done. In the former a savings of US$80million and in the latter US$102million savings were estimated.

Several defects were observed and pointed out at joint discussions with Eisenbau Essen (EBE) testers. The petrographic analysis done was not satisfactory. Vitrinite maceral count was too low and inertinite too high, compared with previous examinations. There was no differentiation of the inertinite maceral group into its various maceral types of micrinite, semifusinite, fusinite and sclerotinite.

Only average reflectance value(Ro) was given instead of providing a complete reflectogram. Analysis of the reflectogram gives useful information of the homogeneity and rank of the petrographic constituents. This is very important for evaluation of the coals used in carbonizing multicomponent blend. Prof. Simon's derived a formula for calculating cokeability of Ruhrcoals. The formula is $G = \frac{E+V}{2}, \frac{k + d}{KV+dE}$ where E = softening temperature, V = solidification temperature, K = contraction, d = dilatation.

G-value calculated from the Ruhr dilatometer readings applying Prof.W. Simon's was used to determine coking blend possibilities of the coals. There has been known cases of coals from both the High and Low silesian coal basins in Poland that exhibited distinct disagreement with Simon's Theory.

As such the formula requires to be first of all tested on Nigerian coals and the imported coal types before its adaptation.

Coal E-type cannot be considered a typical coal because the exinite is very high (17%), also the hydrogen content in this coal is unusually high ($H^{d.a.f} = 7.42\%$) and carbon content ($C^{daf} = 83.11\%$) too low. A medium rank coal with such high content of exinite would not be easily available in the coal market.

In the coals A,B,C, and D-types proposed for coking blend there is no prime coking Coal of International classification code Number 434 or 435 which could act as a binder for these coals and coal E during carbonization. Coals A,B,C, and D fall into the International Classification Code Number (ICCN) 333 and 334 while coal E is 634.

Coke "breeze" (i.e. coke fines) needs special requirements for crushing and hence cost of blend preparation will be considerably higher. Considering that in briquetting hard coals, 8 to 12% itch is normally used, the proposed amount of 7% in the EBE report is on the high side. There is no preheating facility installed in Ajaokuta coke oven plant. So, the blend proposed to include preheating is going to be impossible to implement.

Furthermore, preheating system would have adverse effect on the pitch binder proposed because the softening point of pitch is 70% and it belongs to the group of substances of low thermal stability.

It would be observed that the coking plant in Ajaokuta Steel plant does not provide for grinding and blending of the composition of coals and coal products proferred by the Eisenbau Essen (EBE) testers.

When E.B.E was told of the short-comings of their test results they went back to re-consider the blend composition in view of the limitations in Ajaokuta to their earlier proposals. EBE then came back having done some computer calculations to limit the portion of Enugu coal in the blend to 3% and to be ground 100% through 0.1mm [31].

An amount of 3% of Enugu coal in a coking blend of 1.3 million tons of coal annually would imply the need to import too much of the required coals from foreign sources and the use of too little from local resources. This situation calls for more investigation and exploration of technical means of bringing in more Nigerian coals. Very many tests have been done on Nigerian coals but a thorough and comprehensive record is yet to emerge.

What Nigerian coals are really worth in the process of metallurgical coke preparation has not been convincingly evaluated. Furthermore, Coal types are the product of the vegetation and climate from which the former were developed and since the tropical vegetation has always differed from that of the temperate, therefore, there is the fundamental need to carry out local research on the suitability of Nigerian coals for steel making with an open mind that allows for new methods hypothesis, and interpretation in order to achieve satisfying research results bordering on new knowledge area and to obtain the optimum of energy, and chemicals contained in coal.

Answers must exist to these questions and there are the compelling reasons of economy and existing new frontiers of knowledge to be gained for setting out on this project.

1.13. ENVIRONMENTAL HEALTH & SAFETY IN COAL USE

The principal areas of public concern include land reclamation after surface mining; from underground mining; acid drainage from the refuse from coal mines and coal preparation plants; emissions from combustion such as SO_2, NOx, and particulates; safe disposal of ashes, and the possible effects of CO_2 on climate. The applicability of these concerns vary from country to country depending on a number of circumstances.

Except for the CO_2 question, however, technology is available to meet these concerns and to comply with the most stringent of the current environmental standards in many countries at costs that leave coal competitive with oil at mid-1979 prices in most areas. There is no practical method of controlling CO_2 emission from the combustion of fossil fuels and from other sources, and further research is needed on the possible effects of increased CO_2 emission on global climate. Control of long-range transport of gaseous and particulate emissions may also require new forms of international cooperation.

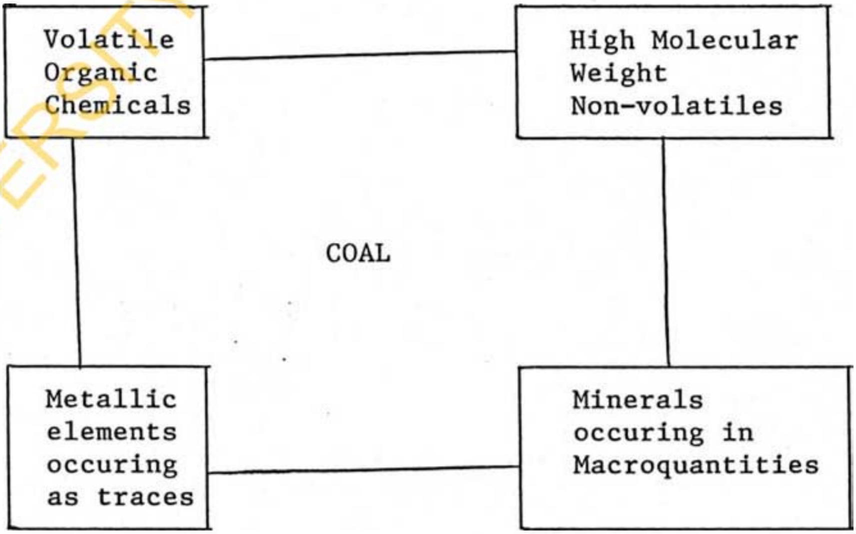
Occupational health and safety are important concerns in coal mining. The major occupational health problems associated with underground mining has been the lung (pneumocosis or black lung) disease caused by breathing coal dust. The greatest safety hazards in underground mining have been from gas explosion and flooding. Reduction of dust and gas levels by much improved ventilation and filtration systems; dust suppression by water-spraying, or laying of powdered limestone; continous monitoring of air quality and the application of strict work rules and practices have done much to reduce the risk of lung disease and to improve safety by reducing the risk of explosion. Moreover, increasing mechanization and better equipment have reduced the exposure of the work force per unit of coal produced.

In mines where the best practices are observed, the accident and illness rates are now comparable with construction work and many sectors of heavy industry. As coal production expands, extending the application of these health and safety practices and continuing workers' training should ensure acceptable occupational health and safety conditions.

1.13.1 ENVIRONMENT HEALTH & SAFETY

COAL COMBUSTION

A schematic look below at the mineralogical and chemical nature of coal will serve as a good guide to the understanding of possible pollution species that do arise from coal utilization.



(2) Low molecular weight hydrocarbons like methane, ethane, etc, carboxylic acids, and thioethers, are examples of volatile organic chemicals contained in coal primary nature. Below and up to 600°C they volatilize, combust, ignite both completely and incompletely depending on the fuel/air ratio. Where the combustion is complete the gaseous products in high oxidation states evolve and could constitute pollution hazards. Thioethers and pyridine do for example, give rise to sulphur dioxide, nitric oxide, and carbon monoxide which are poisonous gases and hence require that their level in the air be controlled.

(3) High molecular weight organic chemicals found in coal are the various alicyclics, polycyclics and polynuclear aromatic hydrocarbons like phenanthrene, anthracene, naphtalene, cumanne, etc. Many of these are non-volatile but do get broken down to smaller fragments and radicals due to high heat effect. Such fragments then become air-borne and are carcenogenous pollutants. Some of these due to their high C/H ratio are often incompletely burnt and evolve as solid micro-particles which constitute soot. They may size-wise be submicron and thus escape arrestment by antipollution devices.

(4) Inorganic macrocomponents of coal end up as their oxides usually, after complete combustion. These comprise alumina, silica, lime, magnesia, titania, oxides of sodium, potassium and barium with a few others.

They remain in the ash or slag and where thewaste management or disposal system is not seriously takcled these ashy oxides would contaminate the agricultural soils and pollute and poison natural water system and the fauna and flora.

(5) Thus Coal combustion releases a number of different substances into the atmosphere. Table 34 gives typical pollutants from an integrated steel plant. Greatly reducing the quantities of such products emitted into the atmosphere requires high costs for emission control. Cleaning up some of the emissions especially sulphur, creates new waste disposal problems such as limestone sludge from the flue gas desulphurization.

(6) Copper, lead, strontium, bismuth, nickel, zinc, mercury, germanium, selenium, arsenic are among the elements that occur as traces in the inorganic mineralogical admixed composition of coal. They occur as various compounds and complexes in coal. Sizeable proportions of each element or its compound volatilize during coal and coke combustion, coke quenching and along with coke oven gases. As submicron particulates they escape dust collection gadgets and precipitators and pollute the atmosphere. Due to their high surface to volume ratio they are good concentrators of other flue gas air-pollutants. Research has further shown that these trace metal pollutants favour brochial and pulmonary tracts for settling in high concentrations.

(7) Coal desulphurization arises from the understanding that in coal utilization sulphur in coal is a nuisance and notorious environmental pollutant. Legislation in many countries has set a maximum level of tolerance of SO_2 that various industries, utilities, and hence coal producers must comply with.

(8) Emissions from coal utilization including combustion may also affect crops, fisheries, and materials. A wide variety of field, vegetable, fruit and nut, forage and forest crops are sensitive to sulphur and nitrogen oxides under controlled exposures. Limited field studies to date indicate potential reductions in crop yield for some species, but increases in yields have been found in soils deficient in sulphates.

(9) Lakes in several parts of the world appear to have recently become acidic and in a number of cases, the fist population has declined considerably. Sulphur and nitrogen oxide emissions contribute to acid rain. It is still to be established what mitigation strategy would be most cost-effective. As a remedy, some researchers have suggested the addition of lime to affected areas to buffer off acidity.

(10) Damage to non-living materials from emissions is of concern in a number of countries.

Deterioration of building materials and works of art, fading of dues, weathering of textiles, and the corrosion of metals under long-term exposure to acidic deposition are outstanding problems resulting from coal combustion.

(11) Sulphur dioxide is converted in the atmosphere to sulphate that scatter light and may reduce visibiltiy. The problem of reduced visibility in cities has, however, been greatly diminished by the use of smokeless fuels or the virtual disappearance of small-scale residential and commercial uses of coal.

(12) Some countries control the potential adverse impacts of emissions on the environment by establishing national air quality standards that speciry the maximum concentrations of certain chemicals permitted in the air. The major emissions that are regulated include sulphur dioxide (SO_2), particulate matter (total suspended particulates or TSP), and nitrogen dioxide (NO_2). Some countries control SO_2 concentration by limiting the sulphur content of the coal burned. Others, such as the United Kingdom, rely on mechanical dispersion of emissions by tall stacks and prevailing winds. Yet some countries, such as the United States and Japan, rely on combined chemical and mechanical systems as well as low-sulphur fuels to reduce emissions.

1.14. AIM OF THIS STUDY

The main raw materials used in the Blast-furnace method of iron and steel production are iron ore, coking coal, limestone, dolomite and refractory clays. Coal is, by far, the single most expensive of all the raw materials.

Before it can be used in the blast-furnace, coking coal is carbonized to give metallurgical coke which provides the thermal energy that smelts the blast furnace charge, produces the reducing gases that convert iron ore to iron, and acts as support material for the blast furnace burden. In addition to these functions, metallurgical coke also provides the permeability required to maintain uniform flow of gases, and hence, high efficiency of the blast furnace operation. In order to prevent contamination of the iron, metallurgical cokes must contain minimum amounts of ash and sulphur.

Nigerian coals are of the stock of upper cretaceous and early tertiary geological age. As such palaeobotanically their level of coalification is that of high volatile bituminous A,B, and C ranks from a good amount of lignite which fringes the coal area towards the souther boundary. Apart from being young coals Nigerian coal resources present sapropelic structural composition microscopically, particularly the Okaba Coal deposit.

There is therefore a high amount of tar evolved on low temperature carbonization. The residue is coke fines of about 50% 0.5mm diameter and the rest is much finer. Lafia Coal is coking but has high ash (mean 25%) and sulphur (mean 2.5%) contents.

Ajaokuta Steel Company Limited which will start producing 1.3million tons of crude steel in its first stage, 2.6 and 5.2million tons respectively in its second and third expanded stages will require the corresponding amount of 1.3, 2.6, and 5.2 million tons of coking coal. In the present Nigerian situation with inadequate transport infrastructure for carrying coal from the sea-port to the plant a ton of imported coking coal will cost, including insurance and freight (CIF) 150 US dollars per ton (US1$ = 4.25₦) to deliver to the stock-yard in Ajaokuta [32]. If all the coals required in Ajaokuta would be imported then corresponding to the expanded stages of pig iron production outlined above the cost of coal import would be 195,390 and 780 million US dollars (at 150 dollars per ton). At todays's Foreign Exchange Market rate of 4.25 naria to the dollar the cost of coal will amount to 828.75, 1,657.5 and 3,315 million naira respectively at the first, second and third stages of expansion of Ajaokuta Steel production. It therefore calles for the development of our local coal resources to make same usable in the metallurgical coke preparation.

CHAPTER TWO

2.0. REVIEW OF COAL DESULPHURIZATION METHODS

2.1. Introduction

The deleterious effect of sulphur in coal and subsequently in coke when the latter is used in the blastfurnace process has been recognized [34]. It thus becomes desirable to remove the sulphur from the coal before carbonization to produce coke containing the minimum of sulphur.

A literature survey has been undertaken here on coal desulphurization methods. The survey covered identification of sulphur forms in coal, methods of determining the sulphur forms, washability tests, and coal desulphurization processes.

2.2. Occurrence and forms of Sulphur in Coal

Sulphur is present in coal [2] in three main forms (or varieties):

- as metallic sulphides (pyrites and marcasites).
- as metallic sulphates and
- as organic sulphur

2.2.1. Pyritic Sulphur

Pyritic sulphur in coal is present as the minerals but differ in crystalline structure. The two forms are usually simply designated as pyrites or iron pyrites.

Pyrites are distributed in coal in many ways. Pyrites occur in lenses and bands, balls or nodules, joints, or cleats, and as finely dispersed particles.

The particle size and distribution affect the amount that can be removed by conventional coal preparation methods. Coarse crushing may release much of the pyrites in the lenses, bands, cleats, and joints, for subsequent removal by mechanical cleaning. In order to free the fine particles from the coal, it is necessary to grind the coal very fine prior to cleaning, but this fine ground coal presents an adverse effect on process efficiency and severely restricts the methods of cleaning that can be applied.

2.2.2. Sulphate Sulphur

Sulphate sulphur in coal [2] occurs as the iron sulphate or calcium sulphate. It is generally of the order of 0.05 per cent but it can gradually increase due to oxidation of the pyrites. Relatively high amounts (0.05 to 1.0%) of sulphate sulphur in coal is an indication that the coal has weathered. As the amount of sulphate sulphur in commercial coals is usually low the sulphate variety of sulphur does not constitute a problem in coal utilization. In coal beneficiation by wet methods the solubility of the sulphate variety results in its dissolution and eventual removal in the aqueous washery bath.

2.2.3. Organic Sulphur in Coal

Organic sulphur in coal is distributed through the coal matrix as an integral part of the coal molecular structure and accordingly it cannot be removed by conventional cleaning or preparation processes, but requires chemical treatment.

In general, organic sulphur is the predominant form of low-sulphur coals; however, as the total sulphur increases, both the organic and pyritic forms increase.

There is no fixed direct relationship between the amounts of each form. However, even though there is no exact knowledge of the forms in which organic sulphur is present in the coal matrix, some groups have been suggested as follows [2].

(i) Bhatnagar and Duti [35] reported mercaptans and disulphides in the extracts. However, the bulk of the sulphur compounds remained in the residue.

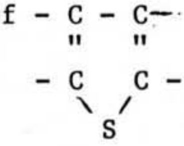
(ii) Roy [36] obtained extracts from an Indian coal using benezene and ethylenediamine, in which he was able to identify mercaptans, sulphide, disulphides, and thiophenes. He also treated the Indian coal with $KMnO_4$ and found that 70% of the sulphur remained in the oxidized residue, so he concluded that 70% of the sulphur is in ring structure.

(iii) Kavcic [37] similarly concluded that 70% of the organic sulphur present in a high sulphur Yugoslavian coal is a part of the heterocyclic aromatic ring structure.

(iv) Gusev [2] demonstrated the presence of both thioether and biothioether groups in Russian coals.

(v) Postowski [38] demonstrated the presence of organically fixed thioether sulphur in coals.

(vi) Lissner and Nemes [39] studying the distribution of sulphur in various fractions obtained when coal is carbonised in an atmoshpere of nitrogen and steam or with steam along at temperature from 200°C to 560°C, found that the organic sulphur occurs in four forms. Between 100°c to 300°C sulphur is evolved from phenolic sulphur groups and probably from ≡ c - SH or ≡ CSSC ≡ groups containing nitrogen. Superheated steam hydrolyzes sulphidic sulphur at 380°C, and retards the formation of new organic sulphur compounds with the evolution of inorganic sulphur.

(vii) Hill and Lyon [2] have represented coal molecule with structures built up of  R-S-R and R-SH

where R and R' are in general cyclic compounds.

(viii) Wiser [2] has written structures for coal molecule containing R-S-SR' (R & R' are cyclic compounds).

2.3. THE SIGNIFICANCE OF SULPHUR IN COKE

The economies of the production of iron are influenced by the sulphur content of the charge [27] especially of the coke used in the blast furnace and hence by the sulphur content of the coal blend carbonized for the production of coke.

Kennedy and Thornton [28] concluded after tests that "if uniformly low sulphur is expected, any increase in sulphur entering the blast furnace required a corresponding increase in slag volume and/or basicity. Increased basicity is to be preferred to increased slag volume, providing the proper slag fluidity can be maintained. Up to an optimum level of somewhere between 5 and 8 percent magnesia in the slag, its substitution for lime improves the desulphurization of the iron. Increased magnesia beuond this range results in a reduction in sulphur-absorbing-power [27]. They also gave theoretical calculation showing that 53.5 additional kilogrammes of coke and 133.8 additional kilogrammes of slag would be required for a furnace producing 0.03% (S) in iron and using 771kg of coke of 1.016 tonnes of iron when the sulphur in the coke is increased from 1.00% to 1.25%. Based on these figures and with coke at US$15 for 1.1016 tonnes and limestone at US$3 for 1.016 tonnes, the increased cost for each 0.10% (S) increase equals 52 cents for 1.016 tonnes of pig iron.

To this must be added the cost due to decreased iron production because of the increased slag-volume.

Secondly, even before the advent of the use of coke in the furnace it was realised [40] that sulphur tended to make iron "hot short", or brittle when hot, and it would not withstand the hammering required in the manufacture of bar or wrought iron. Today the presence of excess sulphur in the pig iron necessitates the introduction of costly pretreatment process for its removal before steel production. Hence a maximum permitted level of sulphur in the pig iron is generally imposed at this stage.

The removal of sulphur in the furnace depends largely on the interaction with lime in the presence of carbon in the general form: $C + FeS + CaO \rightleftharpoons Fe + CaS + CO\uparrow$

The reaction is endothermic and the elimination of sulphur is thus a function of temperature [41]. A high concentration of lime also enhance the removal of sulphur [42], but a slag with higher basicity is associated with higher hearth temperature and the coke rate is increased. Multi-variable equations have been produced with coke rate as the dependent variable and the important independent variables explicitly include the sulphur content of the coke.

Practical limits on the sulphur content of coke are common. In the blast furnace coke, a limiting value of something less than 1.0 percent is appropriate, whilst coke for foundry or cupola use may be subject to a maximum sulphur content of about 0.8 percent. Such limits have a profound influence on the carbonization process. The relationship between the sulphur in coke and that in the parent coal has been the subject of much study. A number of relationships have been produced between the total sulphur in the coke (Sk) and the total sulphur in the coal (Sc) and in general are similar to that proposed by Mott [43]: $S_k = 0.82\ S_c$

It is thus incumbent on a coke-maker to either select low sulphur coals or to ensure that a comprehensive programme of coal cleaning produces a coking coal with the appropriate level of total sulphur.

2.4. THE OBJECTIVE OF COAL DESULPHURIZATION

Sulphur is regarded as an undesirable but economically important constituent of all coals. A survey of world coal deposits indicates presence of sulphur from traces to as high as 10 or 12%. The presence of sulphur in coal is objectionable whether coal is used in the raw state or being carbonized or gasified. Sulphur in coal remains to the extent of 50% of its total value in coke after carbonization and when used in metallurgical iron and steel production the sulphur in the coke may pass into the metal under treatment. Therefore, coal is desulphurized to produce low sulphur (and low ash) cokes for metallurgical and foundry use.

On combustion, some of the sulphur in coal may be evolved by strong heating and be absorbed by the grate bars. If the sulphur is burned to sulphur dioxide serious corrosion of parts of a boiler plant may occur. Desulphurization helps considerably in mitigating corrosion in combustion processes.

During gasification, part of the sulphur originally present in coal is removed as hydrogen sulphide, which causes atmospheric pollution. For each 1% of sulphur in the coal gasified, the gas made contains about 100 grains of sulphur per cubit foot.

The greater part of sulphur is contained in the gas as hydrogen sulphide but some sulphur dioxide is also present and may react with hydrogen sulphide, depositing solid sulphur [40]. In most nations, the permissible limits of sulphur dioxide, hydrogen sulphide, and other sulphur compounds have been laid down by statutory authority. Toxic gases like hydrogen sulphide, sulphur dioxide, and other sulphur compounds can be reduced by desulphurization and atmospheric pollution including acid rain problems can be avoided.

Low-sulphur coals of the world are being gradually depleted. With faster exploitation of good coals and dearth and soaring price of petroleum fuel the energy world has to cope with usage of coals of average (or even high) sulphur content. Desulphurization is the only course open to mitigate the deleterious effects of using high sulphur coals and to upgrade local coal reserves.

In chemical processes, e.g., production of liquid fuels from coal, the gas processed from the Fisher-Tropsch synthesis, ammonia, methanol, or other related synthesis should be almost completely free of sulphur for protection of the catalyst. Therefore, reduction in coal sulphur is practised in preparation of raw materials for coal gasification and liquefaction and other chemical preparations.

Economic circumstances presently do not encourage desulphurization on the simple grounds of the recovery of the sulphur, but it is a possibility. Thiessen (1945) has estimated that in a year of good activity 500 million tons of coal were mined in the U.S. Assuming an average of sulphur content of 1%, 5 million tons of sulphur would be carried by the coal. The greater part of this sulphur eventually escapes to the atmosphere as sulphur dioxide. The recovery of this sulphur would eliminate atmospheric pollution and at the same time add to the mineral resources of the country. The sulphur may be recovered as pyrite during the processes of coal mining and preparation, as sulphur or sulphur compounds in the manufacture of coal gas, producer gas, or water gas, or by scrubbing the gaseous products of combustion of coal.

Coke-oven gas and benzole emanating from high sulphur coking blend would be high in sulphur content and thus require more extensive purification and lead to costly byproducts [41]

With the ultimate objective being to produce metallurgical coke the process of coal desulphurization is essentially coal cleaning and de-ashing which carries with it several advantages and benefits.

High ash content in coal decreases the yield of coke in terms of carbon content or thermal value. High ash in coal also decreases the yield of by-products. When carbonized, coal with high ash produces weak coke and consequently more breeze and fines. Use of high-ash coke in the blast-furnace causes an increase in consumption of coke. More limestone for flux will be required if high ash coke is used in the blast furnace. The furnace capacity is reduced and the control of temperature becomes difficult with high ash coke.

Just like the case of the ash in coke the sulphur in coke produces undersirable effects on the blast-furnace products. Sulphur in coke results in added slag volume, increased coke consumption, greater difficulty of furnace control and longer open-hearth treatment.

Being a soluble alkali metal salt sodium chloride in the coal is reduced subsequent to desulphurization beneficiation of coal. If the amount of sodium chloride in coal is greater than 0.05 per cent when coked the refractory lining of the ovens get corroded except when the lining is of silica, and, in the latter case it would have to be relined after a few months. Silica bricks are more resistant than fireclay bricks to such attack.

One case is known where a premature failure of the silica walls of a coke oven was attributed to the penetration of iron from the coal into the oven walls in the presence of high oven-face temperature. Such iron was probably a constituent of pyrite or marcasite and should otherwise be removed by the process of desulphurization prior to combustion or carbonization. A high sulphur content of coal used for coke-making will, in addition to yielding a high sulphur content of the coke, give crude gas and benzole high sulphur contents and make them require more extensive flue gas desulphurization (FGD) [44].

2.4.1. ADVANTAGES OF COAL CLEANING

Pre-combustion and pre-carbonization cleaning of coal has many advantages in the areas of animal and plant lives, environment, corrosion protection and general economy. Among the many advantages of coal cleaning, the following may be catalogued, [29, 31, 32].

1. A high quality coal that can frequently be obtained by washing at less expense than is provided by "selective mining" is obtained.

2. A coal that would otherwise be too dirty to sell is made saleable.
3. Greater flexibility in looking for buyers is assumed subsequent to coal cleaning.
4. Ability to offer "compliance coal" designed to meet air quality standards is assumed.
5. Although coal cleaning alone cannot enable a utility to meet emission standards for new boilers, it can remove enough sulphur to meet standards in some older boilers.
6. Cleaning can yield a fuel which, used in combination with scrubbers, can insure that utilities that want to use high sulphur coal can consistently meet the most stringent sulphur dioxide emission standards.
7. Scrubber operations from burning cleaned coal derive one or more of such benefits as
    1) improved efficiency and availability;
    2) less consumption of energy;
    3) reduced waste disposal; and
    4) lower operation, maintenance and capital costs.

8. Boiler operations by using cleaned coal derive the advantages of
    1) improved efficiency;
    2) reduced maintenance requirements;
    3) greater availability; and
    4) increased generating capacity.
9. Mixed effects are possible on ancillary power plant processes. Use of cleaned coal reduces pulverizer operation and waste handling requirements. In some cases use of cleaned coal leads to some difficulty in particulate removal as well as difficulties in handling due to coal dust and increased moisture.
10. Cleaning results in lower shipping and general transportation costs because it has been noted that the quantity of coal that gives out a given number of energy units cleaned coal weighs 23 percent less than the same coal raw.
11. Coal cleaning plants can remove an average of 25% sulphur.
12. Coal cleaning plants can remove on the average of 51% ash.
13. Cleaning increases the Btu or heating value of the raw coal by 5 to 44%.

14. Cleaning coal is a way of making the sulphur dioxide concentration of the coal burnt less variable.

15. A case study found the typical fuel cost of cleaning to be an average of $4.8 per ton ($1 = ₦4.25). The same study found that a typical utility would save approximately $7.20 per ton of fuel if cleaned coal was used. Cleaning can thus yield a net gain of $2.35 per ton. It is to be noted however, that generalization on the cost of cleaning coal cannot be allowed by the variability in the factors that govern local cleaning costs, viz, the capacity of the preparation plant and its mode of operation; the type of coal being washed; the degree of cleaning; the local cost of resources such as water, electricity and labour; and the profit the preparation plant owner takes. Similarly, the savings to utilities have to consider the various aspects of generating electricity with coal, including flue gas desulphurization (FGD) systems, boilers, ancillary processes, and transportation.

16. Along with its reduced ash and sulphur content, cleaned coal's reduced mineral content (iron, sodium and calcium) decreased boiler fouling and slagging.

    A study by Hoffman-Muntner (1980) found that each per cent of ash removed reduces the cost of burning a ton of coal by \$0.05 to \$0.10 in 1978 dollars while a percent of sulphur removed reduced the cost by \$0.30 to \$0.50 per ton.

17. In handling and storage cost savings from using cleaned coal were found to be of the order of \$0.01 per ton of cleaned coal.

18. Holmes found a significant correlation between the cost of maintaining pulverizers and the ease with which the coal feed can be ground. Pulverizers maintenance costs increased linearly as the coal's sulphur and ash reached 17.5%, pulverizer maintenance costs increased at a higher rate.

    Hoffman-Muntner (1976) found pulverizer savings to be between \$0.01 and \$0.04 per ton of cleaned coal, with an average saving of \$0.03.

19. Coal cleaning reduces ash handling and maintenance costs slightly by an average of \$0.05 per ton of cleaned coal.

20. Savings achieved on particulate collection on capital cost of electrostatic precipitator when cleaned coal is used has been estimated to be of the order of an average of \$0.45 per ton of cleaned coal. In the design of the electrostatic precipitator a 5 to 25 per cent size reduction is cut in for using cleaned coal from what it would have been for uncleaned coal.

21. Capital savings of the order of the average \$0.11 and \$0.20 are achieved for high and low sulphur respectively cleaned for 0.10 lb $SO_2/10^6$ Btu emission standard.

22. The reduced tonnage involved in shipping cleaned coal saved utilities money on transportation costs. This savings in some cases worked out to an average of \$0.20 per ton or up to \$0.18 per net Kwh depending on the distance shipped and the increase in Btu's per ton of coal.

2.5. Washability Test

2.5.1. Introduction

The washability characteristic of a coal is the most important tool available to determine the extent to which a coal may be cleaned. Examination of washability data for a particular coal or for a particular size of coal will reveal the quality of coal which may be obtained by mechanical cleaning as well as the quantity of coal of a particular quality. Further examination of the data will indicate the ease or difficulty with which the coal may be cleaned.

A washability study of a coal is done by testing the coal sample at pre-selected, carefully controlled specific gravities This is commonly termed "float-and-sink" analysis and/or specific gravity fractionation [45,46,47,48,27].

2.5.1.| Methods of Testing Washability

In descending order of specific gravities a weighed and characterized coal sample is introduced into liquid media. The fraction of the coal sample that floats on the first medium is skimmed off, allowed to drain off and then introduced into the bath of the next lower specific gravity and the skimming off and further testing is continued till the bath of the lowest density is served. The bath of the lowest density has both sunk and float fractions. The portion of coal that sank (or/and floated as in the medium of lowest density) in each of the baths is filtered, drained, and allowed to dry. Each of these fractions is now marked with the specific gravity of the medium in which it sank (or floated).

After drying, the specific gravity fractions are weighed and analysed generally for ash. A table is then compiled showing weight percent of each specific gravity fraction, together with the analysis of each fraction. The data are mathematically combined on a weighted basis into "cumulative float" and "cumulative sink" and used to develop the washability curves that are characteristics for the coal.

Heavy organic liquids and solutions of zinc or calcium chlorides in water are commonly used to obtain the desired specific gravities. However, the latter, although less expensive than organic liquids, cannot be practically concentrated to specific gravities greater than 1.92. Furthermore, these solutions, especially at the higher gravities become viscous and are neither accurate nor satisfactory for separating fine sizes of coal. Also, the solution tends to be absorbed into the pores of the coal, thereby changing its apparent specific gravity, and the viscous solution interferes with the separating process. When either zinc or calcium chlorides are used, it is imperative that the coal be thoroughly washed with warm water to ensure that all traces of the chloride compound is removed, since these compunds distort both the weight of recovered coal and the subsequent ash analysis.

Previously, carbon tetrachloride and benzene were used for float and sink testing. However, because of their high toxicity and cumulative physical effects on human body, their use is now discouraged. Instead, the following organic liquids with the characteristics shown are used in float and sink testing (Table 19). A typical coal washability data is given in Table 20.

Table 19: Organic liquids commonly used in Float Sink Testing of Coal

Name of Org. Liquid	S.G.
Toluene	Sp.gr. 0.866
1,1,1, - Trichloroethane	Sp.gr. 1.349
Tetrachloroethylene	Sp.gr. 1.631
Ethylene dioxide	Sp.gr. 2.172
Methyl Iodide	Sp.gr. 2.280
Perchloroethylene	Sp.gr. 1.62
Bromoform	Sp.gr. 2.89
Heavy naphtha	Sp.gr. 0.7-0.8

Table 20: Typical Coal Washability Data

1 S/No	2 Sp.Gr.	3 % wt diff.flt	4 % ash of diff flt	5 % wt cum flt	6 % Ash cum flt	7 % wt cum sink	8 % Ash cum sink	9 Midpt of sort	10 Sp.Gr. dist (±0.05)
A	-1.35	49.5	8.3	49.5	8.3	50.5	50.1	24.75	-
B	+1.35	8.6	16.2	58.1	9.5	41.9	57.0	53.80	
	-1.40								13.3
C	+1.40	4.7	19.4	62.8	10.2	37.2	61.8	60.45	
	-1.45								8.9
D	+1.45	4.2	24.7	67.0	11.1	33.0	66.5	64.90	
	-1.50								8.4
E	+1.50	4.2	35.7	71.2	12.6	28.8	71.0	69.10	
	-1.55								8.1
F	+1.55	3.9	52.0	75.1	14.6	24.9	74.0	73.15	
	-1.60								6.5
G	+1.60	2.6	65.0	77.7	16.3	22.3	75.0	76.40	
	-1.65								5.8
H	+1.65	3.2	70.4	80.8	18.4	19.1	76.1	79.30	
	-1.70								
I	+1.70	19.1	76.1	100.0	29.4	0	x	90.45	

Legend

(-) = float at or lighter than
(+) = sink at or heavier than
(x) = reference point
wt = weight
diff = difference
flt = float
cum = cumulative
Sp.Gr.dist = specific gravity distribution
midpt of sort = mid-point of sort.

2.5.1.2 Calculations in Coal Washability Test

Calculations:

The mathematical rigours are excluded as use is made of only the simplified percent weight of widely-acclaimed answer-producing arithmetic.

(cumulative float;
(Column (5) = Sum of column (3) up to the required point

e.g. 5(D) = 3(A + B + C + D)
= 49.5 + 8.6 + 4.7 + 4.2
= 67.0

(Percent ash of
(cumulative float,
(Column (6) = Sum of products of column (3) and (4) up to required points divided by column (5) at that point.

$$\text{e.g. } 6D = \frac{(3A\times4A) + (3B \times 4B) + (3C \times4C) + (3D\times4D)}{5(D)}$$

$$= \frac{(49.5 \times 8.3) + (8.6 \times 16.2) + (4.7\times19.4)+(4.2\times24.7)}{67.0}$$

(Percent Weight = 11.1
(of cumulative sink
(Column (7) = 100.0 - column (5) at any point.

e.g. 7(D) = 100 - 5(D)
= 100 - 67
= 33 .

(Percent ash in
(cumulative sink
(
(Column (8) = Sum of products of column (3) and (4) up to required point less one starting from bottom divided by column (7) at required point.

e.g. 8(G) $= \dfrac{(3I \times 4I) + (3H \times 4H)}{7(G)}$

$= \dfrac{(19.1 \times 76.1) + (3.2 \times 70.4)}{22.3}$

(Mid-point of sort,
(Column (9) = Sum of column (3) up to required point less one + half of column(3) at the required point.

e.g. 9(D) $= \underline{3(A + B + C) + \tfrac{1}{2}\,3(D)}$

$= 49.5 + 8.6 + 4.7 + \tfrac{1}{2}\,(4.2)$

$= 62.8 + 2.1$

$= 64.9.$

12

Typical Coal washability curves are shown in **Figure 14**.

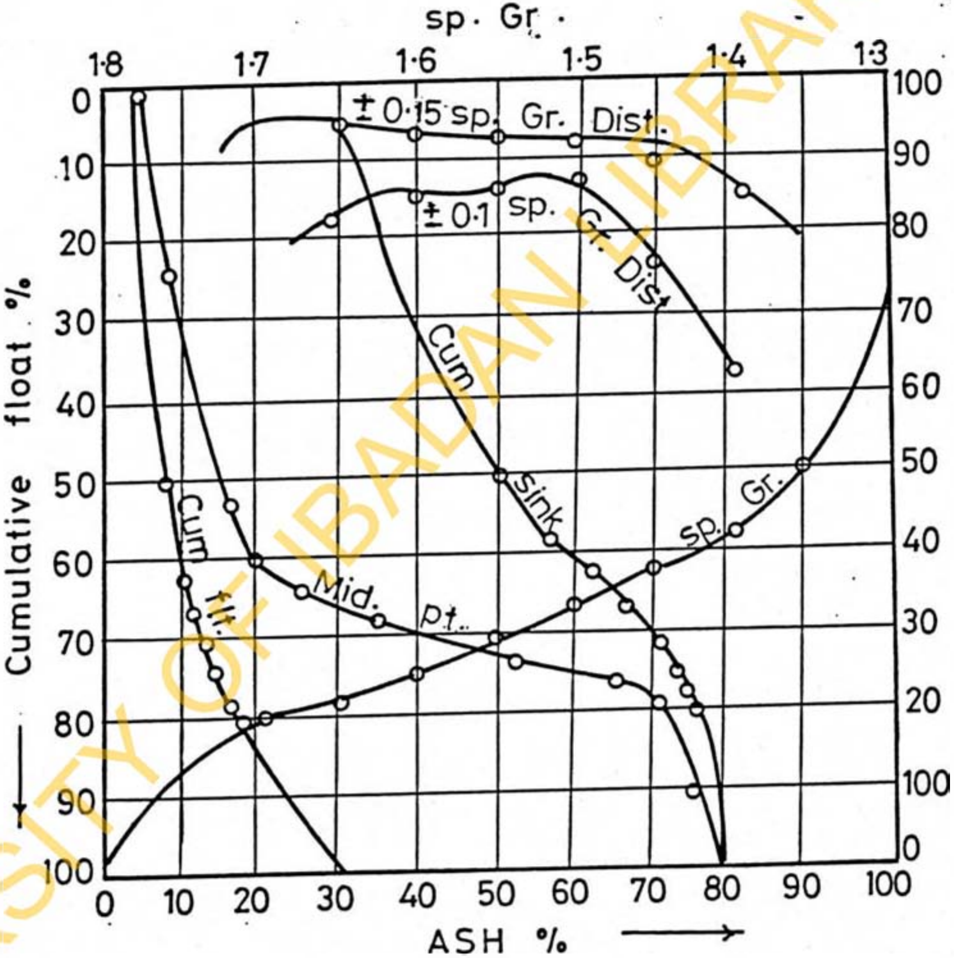


Fig. 12 : Typical Coal washability Curves.

S.G. Distribution
(± 0.1)
Column (10). It is to be noted that the data of of this column are placed in between the rows. They are calculated from differences of data in column (5) as per following examples.

$$10(B/C) = 5(C) - (5A) = 62.8 - 49.5 = 13.3$$

$$10(F/G) = 5(G) - 5(E) = 77.7 - 71.2 = 6.5$$

etc.

2.5.13. Plotting of Washability Curves

There are five standard curves as follows:

1. Cumulative float - columns (5) vs (6)
2. Cumulative sink - columns (7) vs (8)
3. Mid point of sort or ordinate D or characteristic curve or increment curve or elementary ash curve or instantaneous ash curve - column (9) vs (4).
4. Specific gravity curve - columns (2) vs (5)
5. ± 0.05 Sp.gr. distribution - columns (2) vs (10) This curve can also be conveniently plotted by direct readings from curve (4), e.g. for ±0.1 Sp.gr. distribution the following readings are obtained as given in Table 32 below.

Table 21 ±0.05 Specific Gravity Distribiton Curve

Sp.gr	Sp.gr.range - from	±0.1dist. to	Cum float (%wt) readings from Sp.gr. curve
1.40	1.30	1.50	67.0 - 29.0 = 38.0
1.45	1.35	1.55	71.2 - 49.5 = 21.7
1.50	1.40	1.60	75.1 - 58.1 = 17.0
1.55	1.45	1.65	77.7 - 62.8 = 14.9
1.60	1.50	1.70	80.9 - 67.0 = 13.9
1.65	1.55	1.75	87.5 - 71.2 = 16.3

2.5.1.4. Interpretation of Washability Curves

The following is an outline of the interpretation derivable from washability curves.

1) Say, we want to limit ash content of washed coal to 13%
2) Starting from that 13% point on the bottom abscissa and going vertically upwards up to the cumulative float curve, we get, from the corresponding ordinate (left ordinate) the expected yield of about 71%.
3) Moving horizontally to the specific gravity curve at this ordinate and then moving vertically upwards

from the point of intersection, we get, from the top abscissa, the required sp.gr. of separation to be 1.55.

4) As in (2) above, the 29% sink at this sp.gr. will assay about 71% ash, as obtained from the cumulative sink curve.

5) Moving horizontally at the same ordinate (i.e. 71% cum.flt or 29% cum sink) at the point of intersection with the elementary ash curve (mid point of sort curve, characteristic curve, etc) moving vertically downwards, we get from the ash-scale (bottom abscissa) the % ash assay of the most impure fraction of the floated material to be 41.5%. However, the greater significance of the elementary ash curve is that the flatter the curve near the specific gravity of separation, the easier the coal is for washing and vice versa.

6) The specific gravity distribution curve also indicates the relative ease or difficulty of washing a coal at different densities of separation.

Referring to the ±0.1 sp.gr. distribution curve we find that, at 1.4 sp.gr. the % distribution is 38, whereas at 1.55 sp. gr. the % distribution is 14.9 - indicating that washing is considerably easier at the latter sp.gr. Bird's table (Table 22) offers an encyclopedic guide on coal washability.

Table 22: Bird's Table Summarizing ease or difficulty in Coal Washability

S/NO	±0.1 sp.gr. distribution (% wt)	Degrees of difficulty	Preparation
1.	0 - 7	simple	Almost any process process with high tonnages
2.	7 - 10	moderately difficult	Efficient Process high tonnages
3.	10 - 15	Difficult	Efficient process, medium tonnage, good operation.
4.	15 - 20	Very Difficult	Efficient Process, low tonnage, expert operation
5.	20 - 25	Exceedingly difficult	Very difficult Process, low tonnage, expert operation
6.	over 25	formidable	Limited to a few exceptionally efficient, processes, expert operation

2.5.2 Dense Medium Separation

Dense Medium Separation(DMS) otherwise called Heavy Media Separation (HMS) include those coal preparation processes which clean raw coal by immersing it in a fluid having a density intermediate between clean coal (i.e. S.G. 1.3 - 1.6) and reject (S.G. 1.6 - 1.8 inclusive). As there is a general correlation between ash content and specific gravity, it is possible to achieve the required degree of removal of ash-forming impurities from a raw coal by regulating the specific gravity of the separating fluid.

The heavy media separation (HMS) process is a practical application of the familiar laboratory float-and-sink test, which is used as a standard gravimetric separation with almost 100% efficiency. Theoretically, any size particle can be treated by dense medium processes. In practice sizes ranging up to 350mm are treated. Commonly, however, sizes treated are between 0.5mm (32-mesh) and 150mm. Furthermore, sizes coarser than 6mm are treated in static dense medium separator (DMS). Sizes between 0.5mm and 6mm are normally washed in apparatus employing centrifugal force, for example, the dense medium cyclone washer.

Ideally, the separating medium would be a true liquid low in cost, miscible with water, capable of adjustment over a wide range of specific gravities, stable, non-toxic, non-corrosive, and low in viscosity. In the absence of an ideal medium and from a practical stand-point any medium should be cheap at the point of use, physically stable so that it does not decompose or degrade in the process, chemically inert so that it does not react with the coal, easily removable from the coal product, easily recoverable for re-use, low in viscosity at the desired specific gravity of separation and a stability capable of being maintained over the range of specific gravity likely to be required. Commercially used types of separating media are organic aerated solids and suspensions consisting of fine solids in water.

Organic liquids used for coal separations ranges in specific gravity from 0.86 to 2.96. The use of organic liquids for coal preparation is limited to quality control application involving a small number of continous float-and-sink separators. Organic liquids have not been used for preparing salable clean coal commercially. For experimental laboratory float-and-sink, however, organic liquids used are gasoline, benzene, perchlorethylene, carbon tetrachloride, bromoform, acetylene tetrabromide and pentachlorethane.

Chloride salt solutions of iron, manganese, barium and calcium could be used as media in HMS process of coal preparation. However, it is calcium chloride solution which has found commercial application in this wise. The actual density of the solution of separation is 1.14 to 1.25 specific gravity, depending upon the difficulty of separation of the product; the differential between the effective specific gravity of separation ranging from 1.40 to 1.60 and the actual specific gravity of the washer solution is effected by the controlled circulation of the solution. No attempt is made to completely remove the calcium chloride on the coal, because this makes it dustless and non-freezing. The washed coal is rinsed with clear water and the resulting solution containing suspended solids (slimes) is returned to the washer where it is used to replace a large portion of the calcium chloride otherwise necessary to maintain a high density solution in the washer tub[49]

Aerated solid of 30 by 80-mesh and behaves as a heavy fluid having a specific gravity intermediate between clean coal and reject has been effective to achieve a dry, specific gravity separation of coal from refuse. Coarse bituminous dry coal could be cleaned by this device but moist coals cannot be effectively handled.

Suspensions of solids in water are used for coal beneficiation. A suspension may be defined as any liquid in which insoluble solids are dispersed and kept in a state of fluid energy. The specific gravities of separation for coals range from about 1.35 to 1.90. To achieve this range of specific gravity while keeping the volumetric concentration at a reasonable level, it is necessary either to select high specific gravity solids or to introduce upward currents in the separating vessel. As the usually accepted volumetric concentration is between 25 and 45 percent, a size and specific gravity of the suspended solids must be selected that will provide for the desired separating specific gravity while at the same time have the required medium stability. The coarser the solids, the faster the settling rate, the lower the viscosity, and the easier it is to recover the medium; the finer the solids, the slower the settling rate, the higher the viscosity, and the more difficult it is to recover the medium. Additionally, the higher the specific gravity of the suspended solids, the lower the volumetric concentrations for a given specific gravity. Hence, it is possible to select the specific gravity, size range and volumetric concentration of the suspended solids to achieve medium characteristics that provide over-all optimum performance and economy.

It has been shown mathematcially that the density of the suspension is a function of the specific gravity of the medium solids and it naturally follows from this, that media composed of higher specific gravity solids will have a higher density at the limiting volume concentration that suspensions composed of lower specific gravity solids.

High density solids, such as magnetite (5.0 S.G.) and barytes (4.2 S.G) give suspensions which generally are unstable but which permit suspension density up to 2.0. Fortunately, however, the stability of such media can be considerably modified by control of the size grading and by the amount of coal and shale slimes which are present in the suspension. It is found that by fine grinding in the presence of 30 to 40 percent slimes, semistable suspensions can be achieved at densities as low as 1.30 to 1.35.

Viscosity is the "resistance to flow" of a true Newtonian liquid; however, the definition generally is accepted to include suspensions. The viscosity of a medium in the coal cleaning process has little effect on the light coal particles or the heavy reject particles, but becomes critical in the seperation of material of a density equal to or near that of the medium.

As the falling velocity of a particle in a fluid is proportional to the density difference between, the particle and the fluid, a low viscosity medium must be maintained to separate near-density material at a high rate of feed.

A variety of materail that has been used as the solids suspended in water for dense media include sand, magnetite, pyrite, shales, barytes, loess, clay, and mill scale.

2.5.2.1 SEPARATORY VESSELS FOR DENSE MEDIUM SEPARATION

Vessels for washing coals may be categorized as cones, drums, and troughs and include solid cylindrical drums, perforated and solid cylindircal durms rotating in a tank of dense medium, conical tanks and other shaped vessels.

In some vessels, all the medium enters near the top of the tank; in others, a portion enters the bottom of the tank as an upward current; in still others, a portion of the medium enters at various levels to form horizontal currents or maintain homogeneity of the medium throughout the tank.

The feed generally is introduced at the top of the medium bath. In some vessels, however, the feed is forcibly submerged before being released into the medium bath.

In most washers the clean coal is removed with a portion of the medium over-flowing a weir: in a few washers, the floating layer of coal is skimmed off the top of the bath. In cone-typed washers, the reject is removed either through an internal or external airlift or through a lock-hopper arrangement; in drum-type washers, the reject usually is removed from the medium btah by lifters mounted on the inside of the revolving drum; in trough-type washers, the sink usually is carried out of the vessel by a chain conveyor which, in some cases, is the same conveyor that is used to skim off the float coal.

Dense medium processes have many advantages over other coal cleaning processes. some of these advantages of the dense medium processes include

1) ability to make sharp separations at any specific gravity within the range normally required even in the presence of high percentage of the feed in the range of plus or minus a tenth of specific gravity units;
2) ability to maintain a separating gravity that can be controlled within plus or minus five thousandths of specific gravity units;

3) ability to handle a wide range of sizes, from near zero up to 350mm;
4) relatively low capital and operating costs when considered in terms of high capacity and small space requirements;
5) ability to change specific gravity of separation to meet varying market requirements;
6) ability to handle fluctuations in feed in terms both of quantity and quality.

2.5.3. Magnetite Process For Beneficiation of Coal

Magnetite (Fe_3O_4) is used to set operative medium density [50], and operationally, the magnetite process involves:

1) raw coal penetreatment,
2) washing,
3) products recovery and
4) medium recovery, which are now described briefly:

2.5.3.1 Raw Coal Pretreatment

This involves crushing and screening to creat a size-range which the washer can handle. A rotary crusher is often used to break the large size coal to desirable smaller size specification especially where the firability of the coal and impurity differ significantly.

The most important raw coal pretreatment function is the removal of those sizes too fine for washing by dense medium processes. If the finer sizes are to be marketed without further cleaning or if to be cleaned by dry methods, multideck vibrating screens using heated screen surfaces are used extensively. Where the fine sizes are to be washed wet, screening ususally is accomplished on wet multideck vibrating screens. It is imperative that this function is done at high screen efficiency to prevent a build-up of fines in the medium circuit which increases the viscosity of the medium and significantly increases the loss of media. After presizing, the raw coal must be wetted before washing. This is accomplished automatically if wet sizing is used or can be accomplished by spraying the coal with water or dilute media. One of the reasons for wetting the coal is to prevent "rafting" of particles in the separator.

2.5.3.2. Washing of Coal

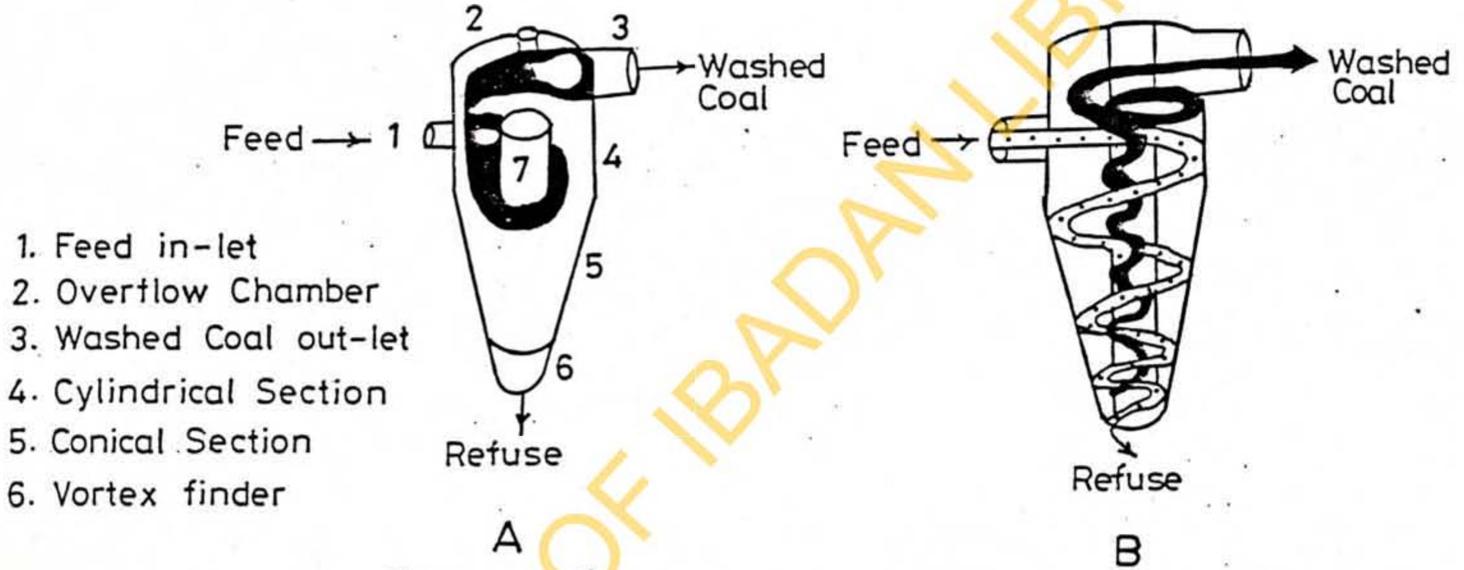
The function of the washer is to effect a separation of the raw feed into a clean coal product and a refuse. Some washers are designed to produce a middling product in addition to a clean coal product. Washers vary widely in design, performance, capacity, and operation to the extent that there is a washer of the type and capacity available for any need.

The products from the washer must be separated from the medium and the medium subsequently recovered. In most cases, the products flow over a short stationary screen where the bulk of the medium is removed without dilution and returned to the medium circulating system. The products then flow onto a vibrating draining screen where sprays of water wash the remaining magnetite from the products. The screens are made sufficiently long to allow most of the water to drain from the products. The dilute medium from this operation is sent to the medium conditioning and recovery system.

2.5.3.3. Medium Recovery System

It is the function of the medium recovery system to recover the magnetite that is rinsed from the products on the rinse screen and to remove the non-magnetite material from a portion of the main medium circulation system for viscosity control.

In the Heavy Media Separation (HMS) process, the float coal left the cone-type washer by over-flowing a weir; the sink product left by an air-lift. These products flowed over a rinsing vibrating screen where most of the medium was recovered and returned to the separator.



Dense medium Cyclone Structure

Dense medium Cyclone idealized flow pattern of medium in Cyclone

Fig. 13. Set up of Dense medium Cyclone (Dmc).

The products then passed onto a washing screen where water sprays removed practically all the remaining magnetite. The rinsing screen underflows are sent to magnetic separators. The magnetite separator tailings are bled from the circuit or used as spray water; the concentrate is sent to a medium sump where it is thickened prior to its re-use in the dense medium circuit.

2.5.4. Dense Medium Cyclone (DMC) Method of Coal Beneficiation

The dense medium cuclone is constructed as in the structure illustrated in Figure 13. The mixture of medium and raw coal enters tangentially near the top of the cyclindrical section, thus forming a strong vertical flow. The refuse moves along the wall of the cyclone and is discharged through the underflow orifice. The washed coal moves towards the longitudinal axis of the cyclone and passes through the vortex finder to the overflow chamber. The washed coal is discharged from this chamber, usually through a tengential outlet, as in diagram B. of Figure 13.

Static bath dense-medium processes effect a specific gravity separation between coal and impurity in a medium of intermediate specific gravity.

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Static bath dense-medium processes effect a specific gravity separation between coal and impurity in a medium of intermediate specific gravity.

The buoyant forces acting on the coal particles causes them to rise to the surface but the impurity particles, being heavier than the liquid they displace, sink to the bottom. The magnitude of the gravitational and buoyant forces that separate the particles is a primary consideration because it governs the velocity with which the particles separate, which in turn determines the capacity of the separating vessel.

Furthermore, the medium currents in the cyclone, also influence the disposition of the particles. The medium flowing through the overflow and underflow orifices act as vehicles for removing the washed coal and refuse. At the in-let a descending vortex starts off and progresses through the cyclone to the underflow outlet providing exit for the refuse. An ascending vortex takes off toward the center of the cyclone and surrounds a cylindrical air core that surrounds the entire longitudinal axis of the cyclone and which promotes the up-lift of coal concentrate through the overflow opening.

Coal size cleaned by the cyclone include top size of minus 25, 9, 6mm by plus 0.5mm at the lower end. Also the most common dense medium solid is magnetite. Grade A magnetite is about 65% minus 65 mesh and grade B is 90% minus 325 mesh with the latter being preferred for its higher efficiency in medium density control.

Finer medium solid presents problems of recovery. On the whole the superiority of the cyclone over other coal cleaning devices is prominent in cleaning low density of "near gravity" material like 1.30 float and 1.30 to 1.40 specific gravity.

2.5.5. Jigging Method of Coal Desulphurization

Jigging is a process of particle stratification in which the particle rearrangement results from an alternate expansion and compaction of a bed of particles by a pulsating fluid flow. The particle rearrangement results in layers of particles which are arranged by increasing density from top to bottom of the bed. This response, developed from the many and continously varying forces acting upon the particles, is a solid-fluid separation more related to particle density and less to partilce size than any other "hydraulic-type" process. The stratification is usually carried out in a rectangular, open-top container, called a jig, in which the mass of particles (termed a "bed") is supported on a perforated base through which the water flows in alternating directions.

Following the particle stratification, the particle bed is physically "cut" at any desired particle density plane thus creating the desired gravity products.

The jig includes means for continously introducing the raw coal, for moving the water through the coal bed in a controllable manner as well as for separating and removing the stratified particles from the system as two or more product streams.

In coal preparation, this highly versatile unit operation is more preferrably applied to a wide size-range of particles with top sizes up to 200mm than to a closely-sized fraction. In gravity ranges, the process has been applied to gold particles with a density of 19.3 and to coal particles having densities less than 1.30. Single jig washers have capacities from five to greater than 700 tons per hour of feed coal. The separation results attainable by jigging have favoured this unit operation for creating a clean coal product as required by steam-coal specification.

Although the jig is used in preparing coals which are difficult to separate, its limitations to achieve both quality products and high recovery are being recognized in comparison with heavy media-based processes which make sharper separations from feed having high "near-gravity" contents. Thus the accuracy of the densimetric stratification in the upper portions of the jig bed are less precise.

In jigging, as in most mineral preparation unit operations, high recovery and product quality are interdependent and inverse process characteristics.

Initially in jigging, a basket was moved up and down in a tank filled with water. The fixed-screen type, in which water is forced through the screen by a plunger was introduced later in jigging practice. Air pressure from an adjacent sealed chamber as the force moving the water back and forth through the fixed screen was much later developed by Mr. Fritz Baum. Alternatively, the water motivation could be achieved by a piston-like plunger [49].

2.5.6. Hydrocyclone Usage in Coal Desulphurization

The Hydrocyclone does not employ heavy medium, rather it employs water movement for particle separation. Its principal design feature includes a greater cone angle and a longer vortex finder than the conventional Dense Medium Cyclone, (DMC).

The separating mechanism is attributable to the ascending vortex. The high and low specific gravity particles moving upward in this current are subjected to centrifugal forces.

The mass of particles, its acceleration and tangential velocity in a vortex bear the relationship

$$a = \frac{(Mp - Mf)}{Mp}\frac{V^2}{r}$$

Where a = acceleration of particle

Mp = Mass of particle

Mf = Mass of fluid displaced by particle

V = tangential velocity

r = radius of cyclone.

For particles in the same position in a given cyclone, this relationship symplifies to

$$a = C(1 - \frac{1)}{SG}$$

Where C = constant, which is equal for all particles

SG = specific gravity of particles.

We would consider a 2.4 specific gravity rock particle and a 1.3 specific gravity coal particle rotating in the ascending vortex. It would be seen from the equation above that the initial acceleration of the rock particle in an outward direction will be about 2.5 times that of the coal particle. The rock particle is therefore much more likely to migrate to the descending vortex and thus report to the underflow product.

At the same time, the coal particle is more likely to be retained in the ascending vortex and to report to the overflow. This acceleration theory applies to the coarser particles but it is not true for small particles. The very fine particles, regardless of their specific gravity, tend to report to the washed-coal product, probably due to the resistance of the fluid and strong secondary currents in addition to ascending and descending vortices. The washed coal ash content can be reduced by decreasing the diameter of the vortex finder or increasing the diameter of the underflow orifice or decreasing the length of the vortex finder.

2.5.6.1. Practical Application of the Hydrocyclone

It is generally observed that much of the low specific gravity particles report to the refuse product. Thus it follows that the hydrocyclone is not suitable for cleaning coals that require cutting at low spcific gravity. This situation necessitates two stage treatment in which the refuse product of the first run is re-treated and the overflow added to the first-treatment-overflow.

The two stage treatment is widely applied to the minus 9mm down to minus 6mm top-size coals. Its application to minus 28 mesh coal is less efficient than to the coarser sizes but Hydrocyclone is especially applicable to minus 28 mesh coal that do not respond well to floatation and yet requiring reduction of finely disseminated pyrite in the washed coal.

2.5.7. Wet Concentrating Tables

Wet concentrating tables are inclined tables (Figure 34) with riffled surface used to separate material particles based on the latter's specific gravity difference [51]. The wet concentrating tables were of two basic types, viz, the bumping tables and the differential motion tables.

2.5.7.1. Bumping Table Type

Bumping tables as the name connotes, utilized a bumping action for conveying the heavy impurity and discharging it over the refuse end. The tables were hung from rods attached to overhead supporting beams. A shaft and cam arrangement at one end of the table served as the actuating mechanism and imparted an endwise, back-and-forth, swinging motion to the table.

A bumping block was fixed in the proper position at the shaft end, so that the table would strike against it at the end of each backward swing. The hanger rods or stirups toward the refuse end; that is, the end next to the drilling mechanism.

2.5.7.2. Differential Motion Table Type

The differential motion feature has been substituted for the camshaft and the bumping-block combination for actuating the table, and the riffled deck with cross-flowing water now replaces the former box-like deck with its longitudinal flow of water and end discharge of the washed coal.

The differential motion imparts a conveying action to the table without resorting to bumping. The fundamental effect of this motion is to make the table deck approach its reversing point and recede from it with greater speed at one end of its travel than at the other. It must be remembered that the table deck in its back-and-forth motion comes to a stop at each end of its travel before its direction of movement is reversed. This means that if it approaches one end of its travel with greater speed than the other, its rate of retardation must be relatively greater at that end.

Hence, both the retardation and acceleration are greater at one end than at the other. This imparts to the table a conveying action as fully effective as that obtained by bumping, with much greater smoothness.

2.5.7.3. Principle of the Tabling Process

The most generally accepted explanation of the action of a concentrating table is that as the material to be treated is fanned out over the table deck by the differential motion and gravitational flow, the particles become stratified in layers behind the riffles. This stratification is followed by the removal of successive layers from the top downward by cross-flowing water as the stratified bed travels toward the outer end of the table. The cross-flowing water is made up partly of water fed separately through troughs along the upper side of the table as shown in Figure 34. The progressive removal of material from the top toward the bottom of the bed is the result of the taper of the table riffles toward their outer end, which allows a successively deeper layer of material to be carried away by the cross-flowing water as the outer end of the table is approached. By the time the end of the table is reached only a thin layer, probably not thicker than on or two particles, remain on the surface of the deck, this layer being finally discharged over the end of the table.

Gavdin's [52] mathematical analysis of flowing film concentration are quite useful to an understanding of basic principles but they do not explain in full the excellent separations that tables are capable of making. Bird and Davis [52] conducted experiments aimed at bringing out the fact that stratification, contrary to the common belief, will not account for the separation effected by the coal washing table, and that cross-flowing water in addition to removing the top strata found on the table, must also have an important selection action in completing the separation according to specific gravity, both in the upper and in the lower strata found between riffles.

The theory of Bird and Davis [52] as to the selective action of the crossflowing water is that only a part of the water flows over the top of the bed between riffles; the remainder flows through interstices in the bed. These interstices are comparatively large near the top of the bed but become progressively smaller toward the bottom thus forming, in effect, V-shaped troughs. In this way the water currents are relatively swift near the top of the bed and become progressively slower toward the bottom. According to them, with paths for the water such that the top strata are subjected to relatively swift currents, and the lower strata are subjected to progressively slower currents, the separation actually occurring on the table can be explained.

As the coarse particles at the top receive swift currents and each successively finer size at the lower levels received slower currents, the velocity of the water matches the size of the materials comprising the different strata. Under these conditions a separation occurs in the lower strata similar to that in the top strata, only it takes place more slowly. The slow currents of water within the bed carry the finer - coal particles along from riffle to riffle, at a more rapid rate than they do the fine bone and shale particles.

2.5.7.4. Effect of Size and Shape of Particles on tabling efficiency:

Shape is not an important factor in the coal tabling process. Yancey [53] made a study of the effect of shape of particles, and for the coal used in his study, decided that "shape of particle is a factor of minor importance in tabling unsized coal, in so far as the over-all efficiency of the process was concerned. Size, and, of course, specific gravity difference are the major factors".

On size, an illustration is a 10-mesh mixture of two materials one of which is low in specific gravity and the other high.

According to the principles of stratification and hindered settling, the separation of the mixture by either process into the two gravity layers will be simple and fast. If, however, there are added two more sizes, say 8-mesh and 14-mesh of heavy particles, the separation into two specific gravity layers will now become more difficult than the uniform 10-mesh size range. That is to say, that, if a table feed contains too wide a range of sizes, some of the sizes will be cleaned inefficiently.

In actual practice there is no objection to a considerable variety of sizes in the feed; in fact, if all particles were of the same size there might be some disadvantages, because the bed would be less mobile and less fluid and conditions within the bed would be less favourable for efficient separation than when there is some variety of sizes. For efficient table operation, however, it is important to guard against having too wide a range of sizes in the feed. A normal feed size consist is about 9mm by 0, but with widespread acceptance of flotation, it is becoming quite common to remove the minus 48- or 100- mesh prior to tabling.

2.5.8. FROTH FLOTATION METHOD OF COAL DESULPHURIZATION

Froth flotation is a physico-chemical method of concentrating finely ground ores [54,55,56]. The process involves chemical treatment of an ore pulp to create conditions favourable for the attachment of certain mineral particles to air bubbles. The air bubbles carry the selected minerals to the surface of the pulp and form a stabilised froth which is skimmed off while the other minerals remain submerged in the pulp.

Flotation is a selective process and can be used to achieve specific separations from complex ores such as lead-zinc, copper-zinc, etc. Initially developed to treat the sulphide of copper, lead zinc, the field of flotation has now expanded to include the Oxidized minerals and non-metallics, including fine coal.

2.5.8.1. Principles of Froth Flotation

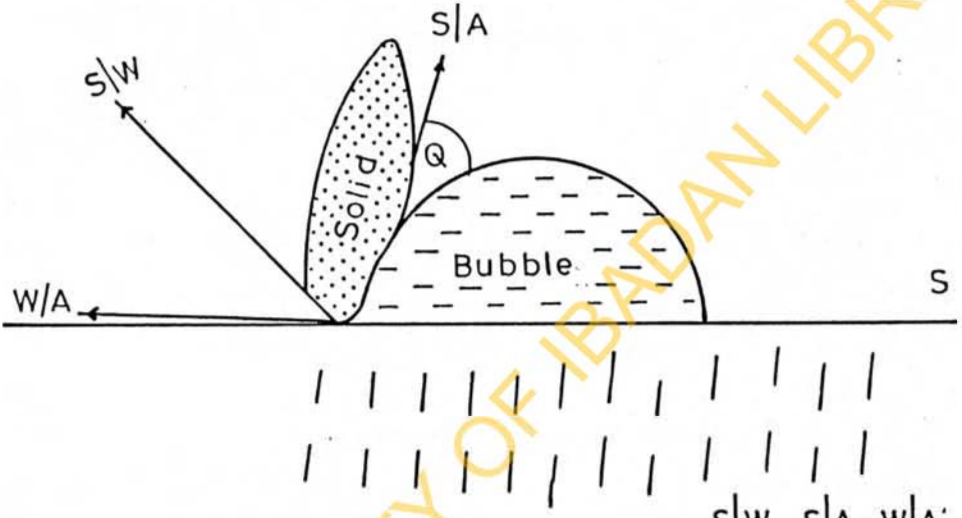
Froth flotation utilizes the differences in physico-chemical surface properties of particles of various minerals [57,58,59,60, 61]. After treatment with reagents, such differences in surface properties between the minerals within the flotation pulp become apparent and, for flotation to take place, and air bubble must be able to attach itself to a particle, and lift to the water surface.

The process can only be applied to relatively fine particles. If they are too large the adhesion between the particle and the bubble will be less than the particle weight and the bubble will therefore drop its load.

In flotation concentration the mineral is usually transferred to the froth, or float fraction, leaving the gangue in the pulp or tailing. This is DIRECT FLOTATION as opposed to REVERSE FLOTATION, in which the gangue is separated into the float fraction.

The air bubbles can only stick to the mineral particles if they can displace water from the mineral surface, which can only happen if the mineral is to some extent water repellent, or Hydrophobic. Having reached the surface, the air bubbles can only continue to support the mineral particles if they can form a stable froth, otherwise they will burst and drop the mineral particles. To achieve these conditions it is necessary to use the numerous chemical reagents known as flotation reagents.

The activity of a mineral surface in relation to flotation reagents in water depends on forces which operate on that surface. The forces tending to separate a particle and a bubble are shown in Figure 14 below.



S|W, S|A, W|A' are surface energies between solid-water, solid-liquid, and water-air respectively Q is contact angle between mineral surface and bubble.

Fig. 14: Forces tending to seperate a particle and a bubble in froth floatation.

$$S/A = S/W + W/A \cos\ Q \text{-eqn}\ (2)$$

where S/A, S/W and W/A are the surface energies between solid-liquid, solid-water, and water-air, respectively, and Q is the CONTACT ANGLE between the mineral surface and the bubble.

The force required to break the particle-bubble interface is called the WORK OF ADHESION, W s/A, and is equal to the work required to separate the solid-air interface and produce separate air-water and solid-water interfaces, i.e.

$$Ws/A = W/A + s/W - s/A \_\_\_\_\_ \text{ eqn } (3)$$

combining equations (1) and (3)

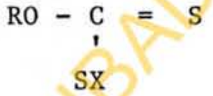
$$s/a = W/A\ (1-\cos\ Q) \_\_\_\_\_ (4)$$

It can thus be seen that the greater the contact angle the greater is the work of adhesion between particle and bubble and the more resilient the system is to disruptive forces. The flotation of a mineral therefore increases with the contact angle: minerals with a high contact angle are said to be AEROPHILIC, i.e. they have a higher affinity for air than for water. Most minerals are not water repellent in their natural state and flotation reagents must be added to the pulp. The most important reagents are the COLLECTORS, which adsorb on mineral surfaces, rendering them hydrophobic (aerophilic) and facilitating bubble attachment. The frothers help maintain a reasonably stable froth.

Regulators are used to control the flotation process; these either activate or depress mineral attachment to air bubbles and are also used to control the pH of the system. Xanthates, for example, are collection regulators, and being an important class of such flotation agents some expantiation is given to the class below.

Xanthates are completely water-soluble, stable flotation agents and are selective.

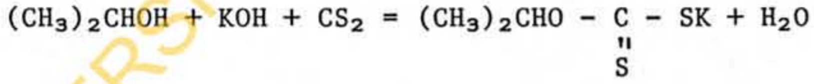
Xanthates chemically are dithiocarbamates corresponding to the structural formula -



$$RO - \underset{SX}{\underset{|}{C}} = S$$

in which R represents an alkyl radical and X is an alkali metal.

Synthesis of xanthates is done using an aliphatic alcohol, carbon disulphides and a caustic alkali, e.g.



$$(CH_3)_2CHOH + KOH + CS_2 = (CH_3)_2CHO - \underset{S}{\underset{\|}{C}} - SK + H_2O$$

Xanthates find wide application in flotation operations of sulphide minerals. Potassium isoprophy xanthate denoted by Z-9 commercially has good collecting power. It is used chiefly for the flotation of complex lead-zinc and copper-iron ores in which the chief minerals are chalcopyrite, chalcocite, enargite, galena, sphalerite, marmatite, pyrite and pyrrhotite.

It is also used to concentrate native copper, gold, silver and cobalt or nickel-bearing iron pyrite. It finds use for concentration of iron pyrites for acid manufacture.

Xanthates are non-frothers and this makes it possible to use them together with a non-collecting frothing agent. Xanthates can only be used in alkaline media although in some cases weakly acidic pulp may be used. pH much lower than 6 would bring about decomposition of xanthates. One, five and ten percent aqueous solution concentrations are common strengths of usage. Because of their rapid, almost instantaneous, action on sulphide minerals no extended xanthate conditioning time is required and it could be added in the flotation processes at any point.

Xanthates should be stored in a cool and dry condition and away from open fires and flames. Contact with water in storage would cause decomposition. Xanthates and its decomposition products are inflammable.

2.5.8.2. Unique Two Stage Froth Flotation

A unique two-stage froth flotation process was developed and patented by the U.S.A. Bureau of Mines, to remove pyritic sulphur from fine-size coal [62,63,64]. The process consists of a first stage standard coal flotation step in which high-ash refuse and the coarse or shale associated pyritic sulphur are removed as tailings. The first stage coal-froth concentrate is then repulped in fresh water, pH is maintained below 7, and coal depressant, a pyrite collector, and a frother are added to float any pyritic material carried over in the first-stage froth; the second-stage under-flow is left as the final clean coal product.

Laboratory and pilot plant flotation tests with coals from various coalbeds throughout the Appalachian region showed that pyritic sulphur reductions of up to 80% could be achieved using this technique.

The coal depressant used is a carbohydrate colloid (a starchy substance) that is recommended for depression of carboneceous gangue in the flotation of minerals. Examples of these bydrophilic carbohydrate substance include cornstarch and quebracho extract.

The pyrite collector most frequently used is potassium amyl xanthate, one of the more powerful xanthates available, and one that is particularly useful in operations where a strong non-selective sulphide collector is needed.

For most of the two-stage flotation work, methyl isobutyl carbinol (MIBC) was used as the frother, but one of the other commonly used aliphatic alcohols or water-soluble polyglycol-type frothers would probably be satisfactory. Any pH adjustment necessary was made with either hydrochloric or sulphuric acid. Sulphuric acid is preferred for large-scale industrial application because it is less expensive.

The pilot plant work was done in a specially designed two-stage flotation facility. Two tanks of flotation cells were used. Both the first- and second-stage flotation machines shown (Plate 21) consist of four 0.6 cubic-foot-capacity Wemco Fagergren flotation cells.

Table 23 below shows first- and second-stage coal-pyrite flotation test results from the pilot plant study done by the U.S. Bureau of Mines, treating a Lower Freeport bed coal from Cambria Country, Pa. The analyses show a pyritic sulphur reduction from 2.19 to 0.87% in the first stage,with a further reduction to as low as 0.43% in the second stage. Second stage recoveries for all tests shown were over 90%.

Table 23: Two-stage Pilot Plant Flotation Results
From Treating Lower Freeport Bed Coal .

Product	Weight percent	Ash	Analysis w/w % Pyritic Sulphur	Total Sulphur
		First Stage		
		2 tests (average)	0.1 lb MIBC/ton	
Clean coal	62.3	7.8	0.87	1.40
Reject	37.7	71.4	4.36	4.50
Feed	100.0	31.8	2.19	2.57
		Second Stage		
	Test 1:	0.6 lb.	Aero Depressant	633/ton
		0.7 lb.	Potassium amyl	xanthate/ton
		0.1 lb.	MIBC/ton	
Clean coal	95.1	7.5	0.55	1.12
Reject	4.9	13.3	6.85	7.41
Feed	100.0	7.8	0.86	1.43
	Test 2:	0.6 lb	Aero Depressant	633/ton
		0.7 lb	Potassium amyl	xanthate/ton
		0.1 lb	MIBC/ton	
Clean coal	93.3	7.5	0.49	1.02
Reject	6.7	12.4	6.28	6.42
Feed	100.0	7.8	0.88	1.38
	Test 3:	0.6 lb	Aero Depressant	633/ton
		0.8 lb	Potassium amyl	xanthate/ton
		0.1 lb	MIBC/ton	
Clean coal	90.5	7.4	0.42	0.97
Reject	9.5	10.7	5.07	5.53
Feed	100.0	7.7	0.86	1.40

2.5.9. Magnetic Separation Method of Desulphurization

Yurovsky and Remesnikov [65,66] reported on research demonstrations that a magnetic separation can result in the removal of pyritic sulphur from coal. The coal was treated with superheated steam and air for a short period of time and then put through a magnetic separator with a field intensity of about 10,000 oersted. During the steam treatment, the reactions on the surface of the pyrite grains produced magnetite, heamatite, and possibly pyrrhotite, ferrous sulphate, and ferric sulphate. All of these minerals are ferromagnetic, whereas pyrite is not. It was reported that a reduction of -32 percent in total sulphur was achieved for a 1 milimeter by zero fraction of Donets coal after the coal had been treated at 320°C for 15 seconds before being put through the magnetic separator. The yield was approximately 84 percent. The total sulphur content of a commercially dried coal was reduced by five to six percent by magnetic separation.

Kester [65] recently devised a similar set of experiments and also found that a significant reduction to total sulphur could be obtained by thermal treatment followed by magnetic separation.

He heated Upper Freeport coal for a period of either 5 to 10 minutes at temperatures from 176°C to 281°C. At temperatures above 190°C, this resulted in reductions in the sulphur contents of the coal ranging from 14.66 percent to 78.79 percent. He then attempted to magnetically separate untreated Freeport Coal fines and found an average reduction of 51.66 percent sulphur in the 100- by 150- mesh size fraction. The material recovery ranged from 83.78 to 91.38 percent. He did not postulate the mechanism whereby the pyrite had been made susceptible to magnetic separation in the untreated coals. Yurovsky and Remesnikov found little reduction in sulphur content by magnetic separation of colas that were not steam treated [67].

Without thermal or other treatment Kester, Leonard, and Wilson [66] reported reduction of pyritic and sulphate sulphur as well as ash by treating certain sized raw coals. The trials were strictly bench-scale runs in the laboratory on 48 to 200-mesh coals particles from the Upper Freeport, Sewickley, Redstone and Pittsburgh beds. The closely sized fine coal was passed through the field of a Frantz isodynamic separator. With the Upper Freeport coal pyritic sulphur was reduced from 1.44 percent to 0.28 percent and total sulphur from 2.58 percent to 1.09 percent.

At the same time ash content dropped from 16.4 percent in the feed to 11.3 percent in the product from the magnetic separator. With coal from the other beds sulphur reduction were: Redstone total sulphur of 2.45 percent in the feed dropped to 1.86 in the product while pyritic sulphur of 1.07 percent in the feed dropped to 0.53 percent in the product; in Sewickly total sulphur of 2.73 percent in the feed dropped to 1.81 in the product while pyritic sulphur of 1.27 percent in the feed dropped to 0.52 percent in the product; in Pittsburgh coal total sulphur of 4.38 percent in the feed dropped to 1.94 percent in the product and feedstock pyritic sulphur of 2.34 percent dropped to 0.37 percent in the product.

2.5.10. Screening Method of Desulphurization

Removal of fine pyrite by screening has been described. In a paper primarily devoted to the reduction of sulphur by froth flotation, Miller [68], found that with an unnamed coal, rougher flotation tends to remove sulphur and ash more effectively in plus 100-mesh material than in the minus 100-mesh sizes. Accordingly, he tried treatment of a classifying cyclone underflow by screening on a sieve bend at 0.25mm.

At the same time ash content dropped from 16.4 percent in the feed to 11.3 percent in the product from the magnetic separator. With coal from the other beds sulphur reduction were: Redstone total sulphur of 2.45 percent in the feed dropped to 1.86 in the product while pyritic sulphur of 1.07 percent in the feed dropped to 0.53 percent in the product; in Sewickly total sulphur of 2.73 percent in the feed dropped to 1.81 in the product while pyritic sulphur of 1.27 percent in the feed dropped to 0.52 percent in the product; in Pittsburgh coal total sulphur of 4.38 percent in the feed dropped to 1.94 percent in the product and feedstock pyritic sulphur of 2.34 percent dropped to 0.37 percent in the product.

2.5.10. Screening Method of Desulphurization

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The undersize of the sieve bend contained 3.05 percent sulphur and the oversize only 1.21 percent sulphur.

Research at the bethlehem Mines Corporation, Johnstown, Pennsylvania, by Benson and Burgess [69] has shown that it is industrially feasible to screen out and reject exceedingly fine pyrite, 150 mesh and even finer, using the Dutch State Mines sieve bend with 0.1mm appertures provided the screen is equipped with a special rocker arm rapping device to strike the surface of the screen. With the rapping device, which Bethlehem designed, both efficiency of undersize removal and capacity are greatly increased. The required specification of 1.0 percent sulphur in the washed coal could not be met by treatment of the 28-mesh to 0 size of this particular raw coal either by hydrocycloning or froth flotation until rapped sieve bends were developed and installed in the washery circuit.

2.5.11. Desulphurization Using the Methods of Oil Agglomeration

Atlantic Richfield Company [70] reduced pyritic sulphur content of coal by the following outlined multistage process:

1) preparing a coal slurry;
2) mixing the slurry with 0.5 - .15 wt% of an inorganic acid-forming agent (e.g. calcium aluminate);
3) blending the mixture with 1 - 60% (w/w) of a hydrocarbon oil (e.g. kerosine)
4) separating the agglomerated coal formed in stage 3;
5) diluting the agglomerated coal (containing 20 wt% oil) to give slurry containing 25 - 75 wt% oil and residual impurities (ash and pyritic sulphur);
6) mixing this slurry with 0.5 - 15 wt% (based on the impurities) water to form clusters (containing water and the impurities);
7) separating the clusters; and recovering coal from the clusters-free coal-oil slurry. By this process, 30 wt% of the sulphur was removed from the coal.

Burk [70,71] applied oil agglomeration to sulphur removal from coal. To reduce the sulphur and ash content of coal, coal particles are treated in an aqueous slurry with a hydrocarbon oil to form coal-oil aggregates which are separated from ash and mineral matter in the slurry by dissolved gas flotation.

The coal particleas are sometimes treated, prior to aggregation, with a conditioning agent which modifies the surface characteristics of the ash and pyritic sulphur mineral matter.

Burk's reduced the pyritic sulphur and ash content of coal particles by treating an aqueous slurry with hydrocarbon oil to form coal-oil aggrègates, incroporating a gas into or on the aggregates to modify their apparent density and separating the aggregates from ash and mineral matter in the slurry gravitationally. Sun [70] studied oil agglomeration approach to coal desulphurization and de-ashing. A process for reducing the sulphur and ash content of coal comprises treatment of the coal particles in a hydrocarbon oil slurry with a conditioning agent and water. Agglomeration of ash and pyritic sulphur mineral matter are effected. The agglomerates are separated from the slurried coal. The coal is recovered from the oil.

2.5.12. Desulphurization using the methods of Solvent refining

Wolk, Steward, Alpert [72] worked on solvent refining for clean coal combustion. Solvent refining removes sulphur and particulates from coal by the use of hydrogen in a reaction system at elevated temperature and pressure. In this process, a substantial portion of the coal is dissolved by an internally generated solvent. Particulates are removed by filteration or other methods. About 70% of the sulphur is converted to hydrogen sulphide and filterable iron sulphides. The end product, a liquid under reaction process conditions, is cooled to an essentially ash-free solid of reduced sulphur content that is useful as a fuel for power generation, solvent-refined coal (SRC) is of particular interest to electric utilities as a way to extend coal-fired plant life, even whel meeting newly lowered limits 1.2 lb SO_2/MBTU for sulphur emissions. As development continues on process equipment and controls, two pilot plants are successfully refining several U.S. coals to mee EPA sulphur specifications (1.2lbs SO_2). Equally important is finding the best way for utilities to pulverize and burn SRC.

2.6. Chemical Methods of Coal Desulphurization

Chemical methods of coal desulphurization are those methods that employ the use of chemicals, including acids, bases, salts, etc, for sulphur removal in coal. Some specific cases of desulphurization by the application of one chemical reaction or the other will now be described because it is strongly believed that there is a case and a place for chemical coal cleaning (CCC) in the science and technology of coal utilization.

The case for chemical coal cleaning is primarily that physical methods of coal cleaning have been found inadequate to up-grade the quality of coals to meet several end-users - carbonization, combustion, gasification, liquefaction, coal chemicals processing, etc. Furthermore, environmental pollution problems invariably still persist after physical coal cleaning as manifest in combustion flue acid gas causing acid rains (e.g. in Scandinavian countries and some other parts of Western Europe). The inadequacy of physical methods of coal cleaning can be illustrated from a survey of sulphur of contaminant in some coals in the United States of America.

The average sulphur content of 455 U.S. coal beds representing principal coal producing regions occurs in the species of total sulphur 3.02%, pyritic sulphur (mineral) 1.91% and organic sulphyr (by difference) 1.11%. It has been experimentally observed that the most advanced physical coal cleaning techniques remove at most 75% of this pyrite sulphur (or 47% of the total sulphur). Thus, an "average" U.S. coal, physically cleaned by the best available practice, would still contain 1.4% total sulphur (and some Appalachian and Mid-Western U.S. coals would nearly double this "average" figure). Combustion of such coals in the utilities, say, would release SOx that would be higher in quantity than is allowed by the environmental protection regulation.

To obtain best results from chemical coal cleaning it has been observed that it is necessary to apply chemical coal cleaning on coal which has already been physically cleanned (and crushed to fine or middling size) as large amounts of coarse pyrite actually inhibit the chemical reactions which remove organic sulphur. Coal with pyrite and ash-forming minerals also tend to use up more reagent in CCC than does cleaned coal and thereby adding an unnecessary cost. As such, the economic estimates of CCC assume that the coal feed is physically washed.

This coal is mixed with water and chemical reagent solution to form a slurry. The slurry and any additional chemical reagent are fed into a reactor vessel, which is sealed, heated and/or pressurized (though there are exceptions using dry coal and operating at ambient temperatures). Under these conditions, much of the sulphur in the coal goes into solution with the liquid reagents which are subsequently separated from the cleaned coal.

It is to be observed from the literature survey that major chemical coal cleaning technologies utilize either "oxidative" or "caustic" leaching to remove sulphur. In "oxidative" leaching, individual organic sulphur atoms are oxidized to form sulphur oxides (SO_x) or sulphates, chemical groups that are easier to "cleave" or break from the rest of the coal molecular structure than unoxidized sulphur. "Caustic" leaching utilizes strongly basic substances such as lye, that actually displace organic sulphur atoms from the coal molecular matrix.

Even though chemical coal cleaning is expected to improve on the quality of coal prior to utilization it itself brews some problems which may now be enumerated.

Subsequent to chemical coal cleaning coal storage and handling present a dust and particulate problem similar to that of physical cleaning but one that is aggravated by the high volume of very fine sizes handled in chemical coal cleaning plants.

Gaseous SO_2 and other obnoxious gases are still produced at the end of CCC and such required scrubbing or flue gas desulphurization of some sort. Gypsum by-product is in excess of demand and inappropriate for some industries like the wallboard which utilizes 73% of all U.S. gypsum; but the brown CCC product would not be an acceptable substitute for white and inexpensive natural gypsum.

The central issue is to maximize the usage of coal and as such no efforts should be spared to improve on coal quality and develop it to meet man's need. As the technology of physical coal cleaning advances that of chemical coal cleaning ought side by side to grow.

2.6.1. Desulphurization by Oxidizing the Organic Sulphur

The basic problem of completely removing organic sulphur has not yet been solved. One proposal is to oxidize the organic sulphur and form soluble sulphates which can be subsequently removed from the cleaned coal. However, the reaction mechanisms are not fully understood and there are insufficient data on the nature of the chemical bonding of organic sulphur in the coal substance.

It is known that some organic sulphur can be converted into sulphuric acids. Accordingly, samples of bituminous coals from the Pittsburgh coalfield, were treated with aqueous oxidizing solutions and were finally desulphurized with the aid of known catalysts. The oxidizing solutions were made up with hydrogen peroxide, mixtures of hypochlorites of sodium (NaOCl) and calcium ($Ca(OCl)_2$. Direct oxidation with peroxide compounds removed little, if any, organic sulphur. Additional treatment with water or phosphoric acid promotes.a substantial reduction in sulphur content. Oxidation with hypochlorite converst some organic sulphur directly into soluble compounds, i.e. gives immediate sulphur removal. Sodium hypochlorite oxidant is more effective than calcium hypochlorite. All the experiments were carried out at moderate temperatures and normal pressures. It has been suggested that more "severe" conditions (increased temperature and pressures) could remove a higher proportion of organic sulphur, but they could also promote serious carbon losses by oxidation and major changes in coal structure. More data are required on the reactions and reaction mechanisms involved in organic sulphur removal before chemical desulphurization can be developed and novel high-efficiency techniques can be devised.

2.6.2. Use of a molten mixture of sodium hydroxide and potassium hydroxide to remove sulphur from coal

A recent study [73] used a molten mixture of equal parts of sodium hydroxide and potassium hydroxide to remove all the pyritic sulphur from coal. A mixture of four parts of molten caustic and one part of coal was used at temperatures of 250°C to 400°C.

2.6.3. Meyers Process of Desulphurization Using aqueous Ferric Sulphate

In the Meyers Process [15,74,75], aqueous ferric sulphate, $Fe_2(SO_4)_3$, is used at 90° - 130°C to oxidize selectively the pyritic sulphur content of coal to yield iron sulphate and free elemental sulphur as shown in the equation below:

$$4.6\ Fe_2(SO_4)_3 + 4.8H_2O + FeS_2 \rightarrow 10.2FeSO_4 + 4.8H_2SO_4 + 0.8S. \quad \_\_\_\_ \quad \text{eqn (2)}$$

The iron sulphate dissolves in solution while the free sulphur is removed from the coal matrix either by vapourization or solvent extraction. The leaching (oxidizing) agent is then regenerated at a similar temperature using oxygen or air and is recycled while product iron sulphate are removed by liming and/or crystallization.

About 100g of 100 - 150 microns top size coal was refluxed with 2 litres of IM $Fe_2(SO_4)_3$ solution. After 4 - 6 hours, the refluxed solution was filtered from the coal and the coal was refluxed with 2 litres of fresh IM $Fe_2(SO_4)_3$. After a total extraction time of 10 - 24 hours, the slurry was filtered and the coal cake was washed with 0.2 N H_2SO_4, then with water to remove residual sulphate. Toluene (400ml) extraction of the coal removed elemental sulphur. The coal cake was then dried in a vacuum oven at 100 - 120°C. This process has realised 83 - 99% removal of pyrite sulphur and total sulphur reductions of 25 - 80%.

It, further, has been found that the density of 1.2 to 1.4 aqueous ferric sulphate leach solution, was used in the Meyers Process, is ideal for accomplishing a practical gravity separation of coal for specific gravities between 1.2 and 1.4. For about 40% of Appalachian coal, the float coal (which is often 40 - 60% by weight of the total) averages 0.8 - 1.2 $lbs/10^6$ btu and needs no further processing to meet standards, while the sink coal slurry contains most of the coal pyrite. This coal slurry can be processed through the Meyers Process to give coal containing 1.2 - 1.5 lbs $SO_2/10^6$ btu. Gravity and chemical treatment are thus combined. This approach is termed Gravichem cleaning.

Gravichem allows production of coal which will meet air pollution control standards for both New and Existing stationary sources (in the USA), and reduces the cost of the Meyers Process through allowing 40 - 60% of the coal to bypass reactor, elemental sulphur extraction and dryer units. An 8-metric ton/day process test plant for chemical desulphurization of coal utilizing ferric sulphate leach solution has just been built TRW's Capistrano Test Site in California.

2.6.3.1. Process Chemistry, Kinetics and Scheme of Meyer's Process

The Meyer's Process is based on the oxidation of coal pyrite wuth ferric sulphate (equation 1). The leaching reaction is highly selective to pyrite with 60% of the pyritic sulphur converted tosulphate sulphur and 40% to elemental sulphur. The reduced ferric iron is regenerated by oxygen or air according to equations (2) or (3).

$$FeS_2+4.6Fe_2(SO_4)_3+4.8H_2O \longrightarrow 10.2FeSO_4+4.8H_2SO_4+0.8S \quad \_\_\_\_ \text{ eqn (3)}$$

$$2.4O_2+9.6FeSO_4+4.8H_2SO_4 \longrightarrow 4.8Fe_2(SO_4)_3+4.8H_2O \quad \_\_\_\_ \text{ eqn (4)}$$

$$2.3O_2+9.2FeSO_4+4.6H_2SO_4 \longrightarrow 4.6Fe_2(SO_4)_3+4.6H_2O \quad \_\_\_\_ \text{ eqn (5)}$$

Regeneration can be performed either concurrently with coal pyrite leaching in a single operation or separately. The net effect of the process is the oxidation of pyrite with oxygen to yield recoverable iron, sulphate sulphur, and elemental sulphur. Equations 3 and 5 yield ferrous sulphate, sulphuric acid, and elemental sulphur as indicated by equation 7.

$$FeS_2 + 2.4\ O_2 \longrightarrow 0.6FeSO_4 + 0.2Fe_2(SO_4)_3 + 0.8S \quad \text{\_\_\_\_\_\_\_ eqn.6}$$

$$FeS_2 + 2.3\ O_2 + 0.2H_2O \longrightarrow FeSO_4 + 0.2H_2SO_4 + 0.8S \quad \text{\_\_\_\_\_\_ eqn.7}$$

Several options exist in product recovery. Iron sulphates may be recovered as pure solids by stepwise evaporation of a spent reagent slipstream with ferrous sulphate being recovered first because of its lower solibility. Alternately, ferrous sulphate may be recovered by crystallization and ferric sulphate or sulphuric acid removed by liming spent reagent or spent wash water slipstreams. Iron sulphates may be stroed as solids for sale or may be converted easily to highly insoluble basic iron sulphates (by air oxidation) or clacium sulphate (by low temperature solid phase reaction) for disposal. Elemental sulphur may be recovered from coal by vapourization with steam or by vacuum, or it can be leached out with organic sulvents such as acetone depending on product marketability and product recovery economics.

Recovery economics may be influenced by quality and concentration of product in the process effluent streams which in turn are influenced by the pyrite concentration in the coal and the desired extent of desulphurization. The process scheme is depicted schematically below (in Figure 15). From the scheme it is apparent that the coal is

1) crushed to the desired size for processing,
2) contacted with hot recycled reagent in the mixer (90° - 100°C),
3) leached of pyrite in the reactor
4) with simultaneous or separate reagent regeneration,
5) washed with hot water and
6) stripped of elementalsulphurm dried and finally cooled. The iron and sulphate sulphur are recovered from spent reagent slipstreams prior to reagent recycle.

Engineering evaluation of available data shows that it is preferable to process fine coal (<2mm top-size) under simultaneous leaching-regeneration conditions at 110°C - 130°C until most of the pyrite is leached out, [75].

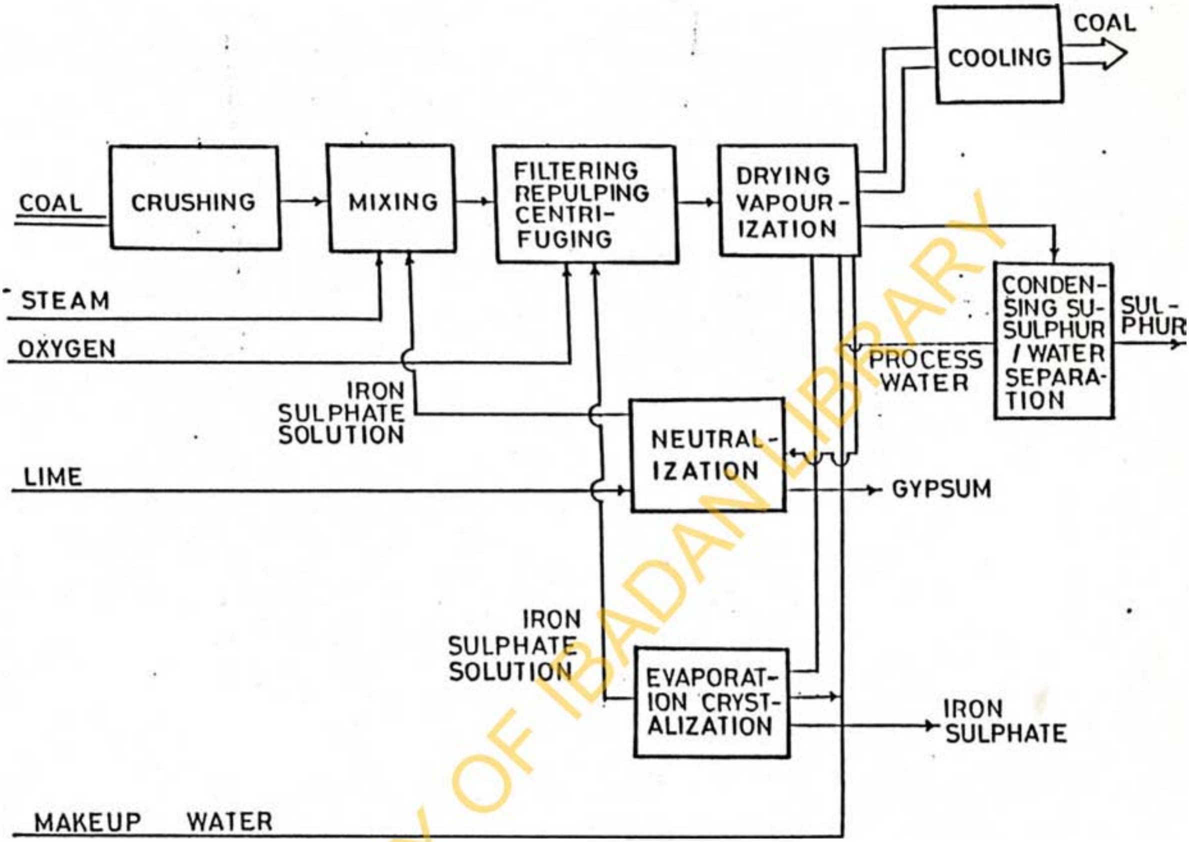


Fig .15. Meyers Process Flow Scheme.

Ambient pressure processing (approximately 100°C) is indicated to remove the last few tenths percent of pyrite since the low Wp value substantially reduces the rate of ferrice ion consumption and, therefore, the need for sumultaneous reagent regeneration. Ambient pressure processing appears to be indicated also for coarse coal (e.g. 10mm top-size) for several reasons. It is difficult to feed continously a non-slurryable coal into and remove it from a pressure vessel. It is much easier and less costly to drain leach solution from the coal and pump it into a small pressure vessel for regeneration. Also the slower reaction rate with coarse coal would require much longer residence times and unreasonably large total volume for pressure vessel.

It is of environmental interest to note that Meyer's Process of desulphurization shows effective removel of As, Cd, Cr, Ni, Pb, and Zn from product coal. These elements are otherwise concentrated (along with Se) in the fine particulates emitted from coal-fired power plants. This fine particulate matter passes through conventional fly-ash control devices.

2.6.4. Oxidative Desulphurization of Coal

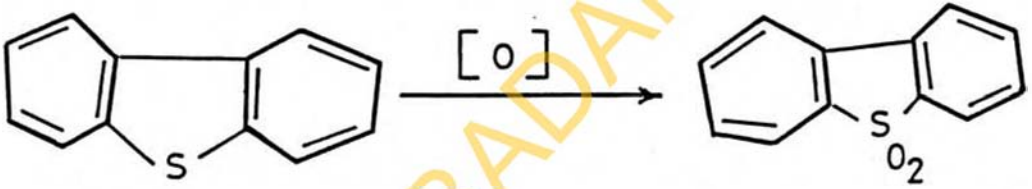
Removal of sulphur from coal prior to combustion by beneficiation techniques involving chemical oxidation is now described below:

A known-weight of minus 200 mesh coal and 100 ml water were placed in a liner (glass or teflon) in a 1-litre magnetically stirred, stainless steel autoclave. The autoclave was pressurized with cylinder air to the required gauge pressure (800 psi in the experiments in Tables 24, 24 and 26) and then was heated with stirring at 800 - 1000 rpm until the specific temperature was reached (approximately 1 hr heat-up time). Operating pressure for 800 psi initial experiments was 100 - 1200 psi, depending on temperature and extent of reaction [24]. After a specified time at reaction temperature, ranging from 5 minutes to 2 hours, the autoclave was cooled by an internal cooling coil. Final pressure at room temperature was between 650 and 750 psi. The contents were removed, filtered, washed until the pH of the filtrate was neutral, and then extracted in a soxhlet thimble with water until sulphate (present as $CaSO_4$) was no longer present in the fresh extract. The coal was then dried thoroughly in a vacuum.

The result of the experiments threw light on the chemistry of organic sulphur removal from coal. The chemistry under observation was based on two premises; The major portion of the organic sulphur in coal was of the dibenzothiophene (DBT), type, and, the reagents had to be inexpensive.

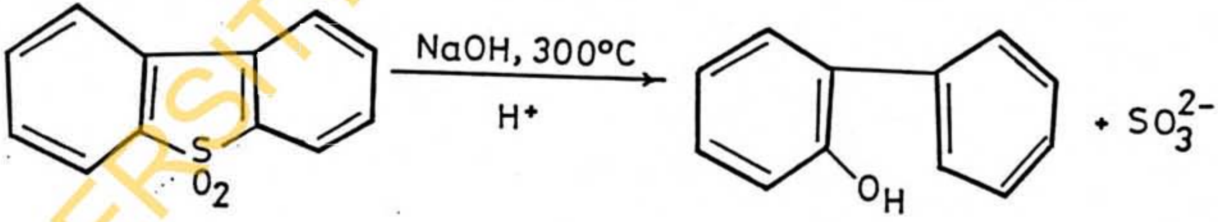
Analysis of the coal obtained showed that at least a sizeable fraction of the organic sulphur in coal is not dibenzothiophenic, but over 50% of it may be. These premises led to the following hypthetical two-step removal of organic sulphur from coal. Step 1: Oxidation of organic (or dibenzothiophenic) sulphur to sulphone and step 2 elimination of the SO_2 from sulphone by base as shown in equations 8 and 9 respectively.

Step. I: Oxidation of organic (or dibenzothiophenic) sulphur to sulphone.



— eqn. 8

Step II Elimination of the SO_2 from sulphone by base.



— eqn.

The first step in the reaction - oxidation to sulphone although extensively documented in the literature, presented more of a challenge. There are numerous oxidants reported which can effect the conversion of organosulphur compounds to sulphones, including $KMnO_4$, HNO_3, CrO_3, H_2O_2/HOAc and hydroperoxides [76]. These obviously do not fit the second premise that the reagents must be inexpensive. It was agreed that the only reagent which could be used as an oxidant was the oxygen in air. But, DBT, and pressumably the organic sulphur in coal, is inert to air at relatively high pressure and temperature. Transfer of oxygen to a carrier to form a hydro-peroxide, followed by reaction of the hydroperoxide with DBT, did give sulphone. It was found that with many hydrocarbons, such as tetralin, decalin, and cyclohexane, merely heating DBT with air under pressure in the presence of the hydrocarbon resulted in formation of sulphone [77], presumably as a result of in situ formation of hydroperoxides. Benzene, which does not form a hydroperoxide, affords no sulphone formation under comparable conditions.

Applying the above-mentioned two-step reaction of air oxidation followed by treatment with aqueous base to coal, LaCount [77] removed up to 50% of organic sulphur, and almost completely eliminated pyritic sulphur as a bonus.

Although this scheme appeared promising, it did require a suitable organic liquid and also NaOH.

LaCount [77] explored another oxidation system which used air as the ultimate source of oxygen. Nitrogendioxide (NO_2) is a good reagent for converting sulphides to sulphones, and it can be used in an easily regenerable system.

$$2NO_2 + -S- \longrightarrow 2NO + SO_2 \quad \text{\_\_\_\_\_ eqn. 10}$$

$$2NO + O_2 \longrightarrow 2NO_2 \quad \text{\_\_\_\_\_ eqn. 11}$$

It was found that DBT could be oxidized to its sulphone using NO_2 and air. When the reaction was extended to coal, however, a significant amount of concurrent reaction took place, including nitration of the coal, which consumed the nitrogen oxides. It thus would have necessitated a continous addition of NO_2 rather than the recycling shown in equations 10 and 11.

LaCount [77] next used H_2O in place of an organic liquid phase. The reaction of coal with water and compressed air almost quantitatively converted the pyritic sulphur in coal to H_2SO_4. In addition, 25% of the organic sulphur was removed. Here was evidence that there was some organosulphur in coal which was not DBT-like, since DBT failed to react with air and water under these conditions.

LaCount, Friedman and Sidney [78] performed their initial experiments on the air-water oxydesulphurization of coal using a batch, stirred autoclave system. In this apparatus in order to replace oxygen as it was used, it was necessary to cool the autoclave to near room temperature, vent the spent air, repressure, and reheat. Although this gave satisfactory desulphurization, it was an impractical approach for studying reaction parameters. The results of their work cited below in Tables 24, 25 and 26 are from 1 hour batch studies without repressurization and thus represent less than maximum desulphurization in some cases. All coals, except those noted, are bituminous.

Table 24: Pyrite Removal from Representative Coals by Oxydesulphurization

Seam	State	Temp (°C)	Pyritic Sulphur (wt%)	
			untreated	treated
Illinois No 5	Illinois	150	0.9	0.1
Minshall	Indiana	150	4.2	0.2
Lovilia No 4	Iowa	150	4.0	0.3
Pittsburgh	Ohio	160	2.8	0.2
Lower Freeport	Pennsylvania	160	2.4	0.1
Brookville	Pennsylvania	160	3.1	0.1

Table 25: Organic Sulphur Removal from Representative Coals by oxydesulphurization

(* subbituminous)

Seam	State	Temp (°C)	Organic Sulphur (wt%)	
			Untreated	Treated
Bevier	Kansas	150	2.0	1.6
Mammoth*	Montana	150	0.5	1.6
Wyoming No 9	Wyoming	150	1.1	0.8
Pittsburg	Ohio	180	1.5	0.8
Lower Freeport	Pennsylvania	180	1.0	0.8
Illinois No 6	Illinois	200	2.3	1.3
Minshall	Indiana	200	1.5	1.2

Table 26: Oxydesulphurization of Representative Coals (* subbituminous)

Seam	State	Temp (°C)	Total Sulphur(w%)		Sulphur(lb/10^6Btu)	
			Untreated	Treated	Untreated	Treated
Minshall	Indiana	150	5.7	2.0	4.99	1.81
Illinois No 5	Illinois	150	3.3	2.0	2.64	1.75
Lovilia No 4	Iowa	150	5.9	1.4	5.38	1.42
Mammoth*	Montana	150	1.1	0.6	0.91	0.52
Pittsburgh	Pennsylvania	150	1.3	0.8	0.92	0.60
Wyoming No 9	Wyoming	150	1.8	0.9	1.41	0.78
Pittsburgh	Ohio	160	3.0	1.4	2.34	1.15
Upper Freeport	Pennsylvania	160	2.1	0.9	1.89	0.80

From Table 24 it is observed that heating high-pyrite coals in aqueous slurry with compressed air at 1000 - 1200 psi and at 150° - 160°C decreased pyritic sulphur to near the lower limit of detection by standard analytical procedure. The sulphur which is removed is converted primarily to aqueous sulphuric acid, with small amounts of ferric and ferrous sulphate in solution. The oxidation of pyritic sulphur is temperature dependent, but at the conditions of experimentation described above reaction is sufficiently fast that above 150°C little improvement is noted.

From Table 25 it is to be observed that removal or organic sulphur from a series of coal varies from 20 to over 40%. As the temperature at which the oxidation is conducted is increased above 150°C an increasing amount of organic sulphur is removed from the coal. Although the percentage of organic sulphur removed parallels the temperature rise, so does the amount of coal which is oxidized. To prevent excessive loss of coal, a practical limit of 200°C has been chosen for carrying out the reaction on most coals.

Furthermore, the variability of the amount of organic sulphur removable by oxydesulphurization is caused by the functionality of the organic sulphur, and gives some rough measure of oxidation resistance or DBT type of sulphur. Obviously that sulphur which is removed by oxydesulphurization must be in some other structure which is readily oxidized such as thiol, sulphide, and/or disulphide. A similar amount of organic sulphur is removed from coal when it is heated at 300°C with aqueous alkali, a reagent which does not attack DBT [78,79].

Table 26 depicts that at 150 - 160°C many coals, including some with rather high sulphur contents, can be dramatically desulphurized.

In conclusion the following observation might be cited. Other than air, water, and coal the only other material needed for this process is the limestone used to neutralize the H_2SO_4. No sludge is formed, much of the water can be recycled, and the only waste product is $CaSO_4$. From a preliminary cost estimate of this process of US$8.00 - US$10.00 per ton the process would still be considerably less expensive than coal conversion to gas to liquid fuel. Over 95% of the pyritic sulphur and as much as 40% of its organic sulphur are removed. Much coal which otherwise would be an environmental problem are rendered envrionmentally acceptable as boiler fuel for industry and power plants through this oxydesulphurization process.

Summarizing, it is to be observed that both pyritic and organic sulphur in coal can be removed by a variety of oxidation techniques, including treatment with NOx, peroxygen compounds, air in the presence of specific organic media, or air and water at elevated temperature and pressure. The most promising method involves contacting an aqueous slurry of coal with air at pressures up to 1000 psi and temperatures of 140° - 200°C. Coal from different geographic areas of the U.S.A. have been treated with air and water in this way, resulting in removal of more than 90% of pyritic sulphur and up to 40% of organic sulphur as sulphuric acid, which is separated from the desulphurized coal by filtration. Fuel value losses are usually less than 10%. Costs of processing coal by this procedue will be somewhere between the less efficient, less thorough and less costly physical coal cleaning methods and the more thotough but much more costly coal conversion techniques. Oxidative desulphurization potentially can upgrade high-sulphur bituminous coals to environmentally acceptable boiler fuel with relatively little loss in total fuel value.

2.6.5. Coal Desulphurization
Ammonia-Oxygen System

This reaction system is carried out under contolled temperature and pressure in an autoclave [80,81,82,83,84].

2.6.5.1. Process Description

In this desulphurization scheme, run-of-mine coal is treated in a conventional preparation plant, where the coal is crushed and washed to remove rock and clay material. The coarsely crushed coal is then fed into closed-circuit wet ball mills where it is further reground to minus 100 mesh.

The ground slurry is pumped into oxygen-sparged leach reactors which operate at about 130°C and 300 psi oxygen pressure. All of the pyritic sulphur and up to 25% of the organic sulphur is removed in about 2 hours. The desulphurized slurry now goes through a solid/liquid separation operation where the coal and liquid are separated. Because of the formation of sulphates and the absorption of some of the CO_2 (from coal oxidation) into the ammonia solution, this mixed sulphate/carbonate stream must be regenerated to recycle the ammonia back into the process. The ammonia regeneration may be accomplished by calcining and/or steam stripping. Sulphur removal from coal is a function of ammonia concentration and retention time.

The chemical reaction for the oxidation of pyrite in an ammoniacal system is given by the equation 12:

$$FeS_2+4NH_3+7/2H_2O+15/4\ O_2 \quad 4NH_4^{+}+2SO_4^{=} + Fe(OH)_3 \quad \_\_\_\_\_\_\_\_ \quad \text{eqn. 12}$$

where all of the sulphide sulphur is oxidized to soluble sulphates. Care was taken to insure that the NH_3/FeS_2 molar ratio for the experimental study was always in excess of the 4 required stoichiometrically: a range between 6.5 and 65 was considered. The results obtained showed that approximately 90% w/w of the pyritic sulphur can be removed, and there appears to be no apparent effect of NH_3 concentration on pyrite removal.

There seemed to be a definite trend, however, in the organic sulphur removal as a function of NH_3 concentration. Measurements of total change in sulphur content of the coal, expressed as lbsSO_2/MBtu, showed a 50% change between the starging coal and the desulphurized coal. This compares against a 25% change after the desulphurization of Illinois No 6 coals when using the O_2/H_2O system [81] where only pyritic sulphur was removed. The desulphurized coals, after an NH_3/O_2 treatment, also showed no residual sulphate sulphur.

An important consideration in any chemical desulphurization process in which the coal sulphur is oxidized is the oxidant consumption. For this process oxygen consumption to oxidize the coal sulphur species and the coal itself may be listed as follows:

1) Reaction with pyrite
2) Oxidation of organic sulphur
3) Oxygen uptake by the coal
4) Oxidation of coal to form CO and CO_2 in the gaseous phase.
5) Formation of carbonates in solution.

The stoichiometric oxygen consumption for the pyrite reaction given by equation 12 calculates to be 1.0 lb O_2/lbFeS_2.

In the oxidation of pyrite sulphur, it is inescapable that oxygen will also react with the coal carbon. This oxidation of the coal usually results in the formation of CO and CO_2, together with soluble coal acids. There is a greater propensity for the formation of coal acids in basic systems than in acid systems. Furthermore, there is some pick-up of oxygen by the coal to form an intermediate oxygen-coal complex.

An important consideration for the viability of any desulphurization process is the over-all thermal efficiency of the system. The results showed that between 8% and 13% Btu loss may be expected after 2 hours of sulphur removal. The result showed that the carbon loss is a function of both time and concentration of the ammoniacal solution. In all cases the carbon loss is much greater than can be accounted for by CO_2 and carbonate formation. This suggested that the difference comes out in solution as coal acids. The formation of these coal acids is not surprising since the reaction of alkalis with coal to form humic acids is well known.

A comparison between the key parameters of the H_2O/O_2 system (only pyritic sulphur removal) and the NH_3/O_2 system (pyritic sulphur and some organic sulphur removal) is presented in Table 27..

Table 27 Comparison between H_2O/O_2 and NH_3/O_2 Systems (for Illinois No 6 coals, 20% slurry density, 2% pyritic sulphur)

Sulphur removal	H_2O/O_2	NH_3/O_2
Pyritic	90^+%	90^+%
Organic	0%	25%
Oxygen consumption ($lbs\ O_2/lb$ coal)		
O_2 for pyrite reaction	0.035	0.0375
O_2 uptake by coal	0.054	0.0340
O_2 to form CO_2	0.032	0.0350
Btu loss (m.a.f.)	8%	13%
Carbon loss (m.a.f.)	6.5%	10%

The higher Btu and Carbon losses for the NH_3/O_2 system are to be expected because organic sulphur removal necessarily implies some coal (and therefore Btu) losses.

Summary and Conclusions

1) Increasing reaction time and ammonia concentration improved the extent of organic sulphur removal.
2) Changing the ammonia concentration had no apparent effect on pyritic sulphur removal (0.5M. 1.0M, 2.0M, 5.0M strengths were used in the experiment with no visible difference in effect on pyrite removal).
3) The oxygen consumed to oxidize the coal carbon (reporting as CO_2 in the gas phase and carbonate in solution) was fairly insensitive to the NH_3 concentration.
4) Ammonia concentration had no apparent effect on oxygen up take by the coal.
5) Both the ammonia concentration and reaction time have an effect on the Btu and carbon values of coal. Increasing either one decreases the Btu and carbon value. As much as 13% Btu and 10% carbon loss may be realized with using a 3M NH_3 solution and reacting the coal for 2 hours.

The large carbon losses are from the formation

2.6.6. Desulphurizing Coal with Alkaline Solutions Containing Dissolved Oxygen

The extraction of pyritic sulphur from coal by leaching the comminuted material with hot aqueous solutions containing dissovled oxygen has been demonstrated in numerous laboratory experiments [85,86,87]. Although acidic solutions have generally been used for such experiments, basic solutions appear to offer several important advantages. Thus Majuma and Peters [89] showed that the rate of extraction of sulphur from relatively pure pyrite is much greater in basic solutions containing dissolved oxygen than in neutral solutions. Moreover it has been shown recently that basic solutions containing ammonium hydroxide and oxygen can extract a significant portion of the organic sulphur as well as most of the inorganic sulphur from coal at relatively moderate temperatures (e.g. 130°C) [88] whereas higher temperatures (150° - 200°C) seem to be required with acidic solutions to remove organic sulphur. Furthermore, some types of basic solutions are much less corrosive towards the common materials of construction than acidic solutions.

In a solution of some base such as sodium hydroxide, the stoichiometry of the reaction of pyrite with oxygen and the subsequent neutralization of the acid produced can probably be represented by the following equations.

$$FeS_2 + \frac{15}{4} O_2 + 2H_2O = \tfrac{1}{2}Fe_2O_3 + 2H_2SO_4 \quad \text{\_\_\_\_\_\_ eqn. 13}$$

$$2H_2SO_4 + 4NaOH = 2Na_2SO_4 + 4H_2O \quad \text{\_\_\_\_\_\_ eqn. 14}$$

Although these equations suggest that the main purpose of the alkali is to neutralize the acid and to drive the first reaction to completion, the actual reaction mechanism is probably more complex with the alkali playing a more subtle role as well as the obvious one.

An investigation by Mckay and Halpern [90] of the reaction of pyrite with oxygen in acidic solutions showed that the product distribution, and therefore the reaction mechanisms, is quite different from that noted above for basic solutions. Thus pyrite was converted to soulble ferrous and ferric sulphate, sulphuric acid, amd elemental sulphur when leached with oxygen-bearing acidic solutions at 100° - 130°C.

The conversion of all forms of sulphur in the coal particles to solube sulphate was determined by analysing the spent leachant. Essentially all forms of the sulphur was extracted from minus 200 plus 230 mesh pyritic particles in one hour by an oxygenated solution containing 1.5 wt% sodium carbonate. The results showed that of all the alkalis tests, which included sodium carbonate, sodium hydroxide, sodium phosphate, and ammonium carbonate, sodium carbonate was the most effective. The effectiveness of all the alkalis increased with initial concentration up to some optimum value and then decreased. For sodium hydroxide the optimum initial concentration was about 1 wt%, for sodium carbonate 1.5 wt%, for sodium phosphate 3 wt%, and for ammonium carbonate 4 wt%.

The results of leaching different sized fractions of pyrite with sodium carbonate solutions of different concentrations showed clearly that the conversion increased in an almost exponential manner as the size was reduced. Particles smaller than 60mm in diameter were completely desulphurized in 1.0 hour by solutions containing 2 - 5 wt% sodium carbonate for the conditions of 7 - 8 atmospheres of total system-pressure, and operative temperature of 150°C.

On the other hand particles larger than 210mm in diameter were only about half desulphurized under these conditions.

Several runs were made using longer reaction times to see how the conversion would change with time. The results indicated that the reaction rate decreased as the batch leaching operation proceeded. The declining rate could have been caused by the decreasing availability of unreacted pyrite, increasing resistance to diffusion of reactants within particles, increasing concentration of some rate - inhibiting reaction product in the leach solution, or a combination of these factors.

A series of further coal leaching experiments have been reported to determine the effects of various alkalis and alkali concentrations on the desulphurization of coal from the ISU Demonstration Mine No.1. The raw coal was pulverized so that 90% was finer than 200 mesh. For each experiment 50g of coal and 500ml of water or alkaline solutions were placed in the stirred autoclave. The chemical treatment was carried out for 2 hours at 150°C with oxygen dissolved under a partial pressure of 3.27 atmospheres and using a total system pressure of 7.83 atmospheres. The treated coal was recovered, dried, weighed and analysed to determine the effectiveness of the treatment.

The best over-all results were obtained with sodium carbonate among the following alkalis which were tested in various concentrations: sodium carbonate, sodium hydroxide, sodium phosphate, calcium carbonate, calcium hydroxide and ammonium carbonate. The only alkali which approached sodium carbonate in effectiveness was sodium phosphate. However, it required a 7 wt% initial concentration of sodium phosphate to produce coal with sulphur content comparable to that obtained with a 1.4 wt% initial concentration of soidum carbonate. Moreover, the Btu recovery was lower and the ash content of the product higher when sodium phosphate was used to obtain comparable desulphurization.

The best results were obtained with an initial concentration of 1.4 wt% and using either lower or higher concentrations resulted in less desulphurization. Under the mild leaching conditions used in this work under reveiw [85,86], mainly inorganic sulphur was extracted from the coal. Essentially all of the sulphate form of sulphur was extracted and a good share of the pyritic sulphur. It was not certain whether organic sulphur was removed or not.

The yield of dry coal was 93 - 94% and the Btu recovery was 88 - 92%. Since the heating value of the treated coal on a moisture-ash-free (m.a.f.) basis was nearly the same as that of the untreated coal, it did not appear that the coal was significantly degraded by the treatment. Also the oxygen content of the coal, as determined by ultimate chemical analysis, did not appear to be significantly different from that of the untreated coal.

2.6.7. Combined Physical and Chemical Cleaning of Coal

Combined physical and chemical cleaning of several coals have also been reported. Each coal was crushed to 6mm x 0 size with a double roll crusher and then cleaned by gravity separation in an organic liquid having a specific gravity of 1.60. The cleaned float fraction was recovered, dried, and pulverized with the pulverizer. The pulverized coal was wet-ground next in the ceramic jar mill for 20 hours and finally leached in the stirred autoclave for 2.0 hours with a 2.0 wt% sodium carbonate solution. The effects of different leaching temperatures and oxygen partial pressures were studied.

The results obtained showed that gravity separation removed a substantial part of the pyritic sulphur from each coal and that chemical leaching removed most of the remaining inorganic sulphur , both pyritic and sulphate. Thus through the combined treatment, the inorganic sulphur content of each of the three coals was reduced by 93 - 95%.

For the physically cleaned and finely ground coal, it did not seem to make any difference what leaching temperature in the range of 120 - 150°C or what oxygen partial pressure in the range of 2.4 - 5.7 atmospheres was used because essentially all of the inorganic sulphur was removed even under the mildest treatment conditions.

Thus alkaline solutions containing oxygen dissolved under pressure were very effective at elevated temperatures for extracting the inorganic sulphur from coal. Of various alkalis tested, sodium carbonate gave the best results. Sodium carbonate also had the advantage of being readily available, low in cost, and relatively non-corrosive in aqueous solution towards steel and other common materials of construction. Leaching coal fines with dilute sodium carbonate solutions at temperatures up to 150°C and with oxygen partial pressures up to 5 atmospheres for up to 2 hours does not seem to degrade high volatile bituminous coal.

However, the coal does absorb sodium from the leach solution which can be removed subsequently by washing with dilute acid. The rate of desulphurization can be increased by reducing the size of the coal and/or pyrite particles. Although sufficient alkali should be used to neutralize all of the acid which is produced through oxidation of the coal sulphur, an excessive concentration of alkali seems to slow the rate of desulphurization. Chemical leaching can be combined advantageously with physical cleaning since the latter is more adept at removing coarser particles of pyrite while the former is more adept at removing the microscopic particles.

2.6.8. Hydrothermal Coal Cleaning Process

Basically the Hydrothermal Coal Cleaning Process [91,92] comprises five major processing operations, namely, coal preparation, hydrothermal (desulphurization) treatment, lidquid/solid separation, fuel drying, and leachment regeneration. Coal preparation may entail a simple grinding operation to reduce the raw coal to the desired particle size of 70% minus 200 mesh or 100% minus 28 mesh.

On the other hand, this operation may involve two operations - grinding of the coal to the desired particle size followed by physical beneficiation to remove a portion of the mineral matter including a portion of the pyritic sulphur.

Hydrothermal treatment entails basically three processing steps:

(1) The ground coal is mixed with an aqueous alkaline leachment, for example, an aqueous solution/slurry of sodium hydroxide and lime, to produce a raw coal slurry.

(2) This raw slurry is heated in an autoclave at about 250° - 350°C (steam pressure of 600 - 2500 psig) to extract a significant portion of the sulphur and the mineral matter, depending on the leachment and processing conditions.

(3) The coal product slurry is cooled and pumped into a receiving tank.

The desulphurized coal is then separated from the spent leachant and washed with water. The final product is a solid fuel containing reduced concentrations of sulphur and, depending on the leaching conditions, mineral matter.

Drying the fuel to remove the residual moisture is optional. For some uses, it may be desirable to burn the fuel wet, in others, removal of a part or all of the residual moisture may be desirable. In any event, the coal can be dried in commercially available dryers.

The spent leachant containing the extracted sulphur as sodium sulphide (Na_2S) can be regenerated for recycle in several ways; one approach involves treatment of the spent leachant with carbon dioxide to remove the sulphur as H_2S which is subsequently converted to elemental sulphur by the Claus or Stretford process. The resulting liquor containing soidum carbonates is treated with lime to convert the carbonate back to sodium hydroxide which is recycled to the desulphurization segment after adjusting the concentration. The calcium carbonate is thermally decomposed to lime and carbon dioxide for recycle to the leachment regeneration segment.

2.6.8.1. Efficiency of the Process

The hydrothermal Coal Process is able to extract greater than 90% of the pyritic sulphur and up to 50% of the organic sulphur from certain coals.

Btu recovery as solid clean fuel is in the range of 90 - 95% depending on the coal and processing conditions.

The coals cleaned by hydrothermal coal process can be used directly as a source of energy without further cleaning during the combustion process, assuming all of the sulphur is emitted to the atmoshpere.

During the desulphurization operation, the coal structure is opened up to give a product having a sponge-like morphology. The porous structure allows the alkali to penetrate the coal particles and subsequently to react with the functional groups, for example, carboxylic acid groups, of the coal molecules. Also the alkali is physically deposited within and on the coal particles. Consequently, in addition to having a reduced sulphur content, the hydrothermally treated coal is impregnated with alkali which acts as a sulphur scavenger during the combustion process. In some recent combustion studies about 41.4 to 75.7% of the sulphur remaining in the treated coal was captured by the alkali in the treated coals. This added feature of the hydrothermal process greatly increase the number of coals which can be used as a source of clean energy especially those coals containing relatively high concentrations of organic sulphur not subject to removal by mechanical cleaning.

Hydrothermal treatment results in the extraction of a number of the trace metals. Examples of these are shown in the table below (Table 28).

Table 28 : Toxic metals extracted by hydrothermal treatment of Coal (average values from 3 Ohio Coals).

Metal	Concentration (ppm)	
	Raw Coal	Treated Coal
Arsenic	25	2
Beryllium	10	3
Boron	75	4
Lead	20	5
Thorium	3	0.5
Vanadium	40	2

Therefore, trace metal emissions from the combustion of hydrothermally treated coal should be lower than those from the combustion of the corresponding raw coals.

2.6.9. Coal Desulphurization by Low-Temperature Chlorinolysis

Chlorinolysis is one of the chemical methods of sulphur removal from coal [93,94].

Laboratory Processing

Chlorine gas is bubbled through a suspension of powdered (100 - 200 mesh) moist high sulphur coal in methyl chloroform at 74°C and atmospheric pressur for 1 - 4 hours. The coal slurry is distilled for solvent recovery. The chlorinated coal is hydrolysed with water at 50° - 70°C for 2 hours and then filtered. The coal filter-cake is dried and dechlorinated by heating at 300° - 500°C in a steam or vacuum atmoshpere for approximately 1 hour. Analysis of the original and processed coal was made for organic, pyritic and sulphate sulphur as well as an ultimate analysis of the coal including an analysis for trace metal removal by the desulphurization process.

Experimental data obtained with high sulphur (4.77%) bituminous coal from Hillboro, Illinois are presented in the following (Table 29).

Table 29: Preliminary Chlorinolysis data for bituminous coal (Hillsboro III) Desulphurization

Sulphur Form	Raw Coal (% Sulphur)	Treated Coal (% Sulphur)	Sulphur removal (%)	
			achieved	expected
Pyritic	1.89	0.43	77	77
Organic	2.38	0.72	70	70
Sulphate	0.50	0.35	30	100
Total	4.77	1.50	69	76

On the basis of available information in literature it could be seen that the low-temperature chlorinolysis desulphurization method provides a number of basic advantages like

1) A high degree of sulphur removal, particularly organic sulphur removal;

2) Additional removal of trace metal concentrations contained in the original coal that reduces potentially hazardous emissions to the atmosphere;

3) A relatively simple coal processing operation mostly at low temperatures and atmospheric pressure which translates into a low-cost process;
4) A beneficiation of the coal for use in direct-fired combustion as well as in gasification processes;
5) Improved coal feedstocks for combustion and gasification operations since treated coal is non-caking and non-swelling.

2.7. Microbiological Removal of Pyritic Sulphur

Bacterial removal of pyritic sulphur from coal has become an engrossing study within the past twenty to twenty-five years. Interest has increased in the effect of iron- and sulphur- oxidising bacteria upon the minerals in coal. Russian Workers [95] reported that the bacterium Thiobacillus ferro-oxidans removed from 23 to 27 percent of the pyrite from Donetz Basin coal in 30 days.

Silverman, Rogoff, and Wender [96], studied the effect of Ferrobacillus ferrooxidans on bituminous and sub-bituminous coal and lignite. They found the bacterial action was most effective on the finer particle sizes of coal. In some cases over 80 percent of the pyrite was removed from the coal in three to four days. Bituminous coal was most easily desulphurized, and lignite was the most resistant to the bacterial action. These bacteria are acidophilic and pH values of the solution ranging from 2.5 to 4.2 were found to be the most efficient. Further, they pointed out that some of the difficulties in using bacteria for removal of pyrite are the handling of the necessarily-small sizes of the coal, the handling of the acid mixture, and the disposing of the products of pyrite oxidation (sulphuric acid and ferric sulphate) without adding to water pollution in the environment.

Addition of an essential bacterial nutrient and a surfactant and removing calcite from coal strongly favour the leaching of pyrite from coal by acidophilic bacteria. Besides pyritic sulphur, significant amount of sulphate and a small amount of organic sulphur are removed.

CHAPTER THREE

DESCRIPTION OF STUDY AREA AND COLLECTION OF SAMPLES

3.1. THE STUDY AREA

The major areas of coal deposit in Nigeria which offer huge potentials for commercial mining and metallurgical coke preparation are Enugu, (Anambra State), Lafia (Plateau State) and Okaba (Benue State) as in Figure 16; hence this study spans three states in Nigeria.

Enugu is a city sitting on huge coal deposits and popularly known as the Coal city. It owes its large population as well as socio-economic development to the coal industry which dated back to 1915. Enugu is also the capital of Anambra State.

The town of Lafia after which the coal deposit in this area has been named is in Plateau State and lies some 85 km due North of Makurdi (capital of Benue State). The coal deposit here extends a broad belt 30 km wide to the South-East of Lafia town. The overall deposit is estimated to cover an area of 2,500km^2 although this study has been restricted to an area covering 50km^2.

The area covering the Coal deposit in Lafia is relatively flat, varying in elevation from 120m around the numerous seasonal streams that intersect the area to 250m above sea level on the areas of higher ground associated with outcrops of sandstone which lie within the sequence. Both Obi Village and the town of Lafia are located on such features. Seasonal and all year round Streams and Rivers like Dep, Bui and Ankwai drain into River Guma which discharges into River Benue east of Makurdi (Figure 8).

The third area in this study is Okaba, located in Ankpa Local Government Area of Benue State. Here, the Igala Plateau which is about 500m high has a high steep slope through a range of about 100m and then slopes gently for about 3km eastward to a height of about 300m where Okaba situates 16km from Ankpa town. Enugu escarpment rises from here and the coal field is in the outskirts of Okaba town.

3.2. COLLECTION OF COAL SAMPLES

3.2.1. Enugu

Coal Stockpile with nominal upper size of 75 - 100mm from seam 3 of Onyeama Coal Mine was sampled by hand with hand-shovel of blade size 250mm. 65 Coal sample increments, with each increment weighing 6kg coal were collected from different points on the stockpile. The gross sample was transported to the laboratory from the field in metal drums whose inside were lined with polythene sheets. Before wrapping up the coal in the drums with polythene sheets and sealing the durms, water from the mining pit was sprinkled on the coal to keep it wet. This was done to maintain the coal during transit in an environment similar to its natural subterranean condition and prevent desiccation. Care was taken to prevent breakage of coal during transit and minimize generation of fines.

3.2.2. Lafia

The general thickness, range, and persistence of particular coal horizons in Lafia/Obi area is indicated in Table 30. A total of 117 boreholes were drilled in Lafia/Obi area and two shafts were sunk reaching the seams by driving tunnels through the rock strata.

100 of these boreholes were drilled in Obi area with 8 boreholes per km^2 on the crest of the Giza anticline. The depth of the boreholes vary but the deepest was 520m. The density of Boreholes outside Obi area were generally less than 1 per 50km^2.

Coal samples were taken of the entire thickness of seam 12 upper (which is the commercial seam) but avoiding the admixture of roof or floor earthy material. Coal samples were obtained by cutting vertically down the horizontal stretch of the Coal Seam with pick axe and shovel. The depth of sampling was 26.20m to 27.05m. The incremental Coal Sample so cut was not crushed. It was put into metallic barrels having 180 - 200 litres volume capacity. 20 barrels were loaded with the seam sample and a barrel contained about 150kg of coal. Water from the coal shaft was poured into the barrel to covery its coal content. The barrel was sealed air-tight by welding and each barrel was labelled appropriately.

With details of location and content, the barrels were transported in a tipper lorry from the coal field to the laboratory in Jos.

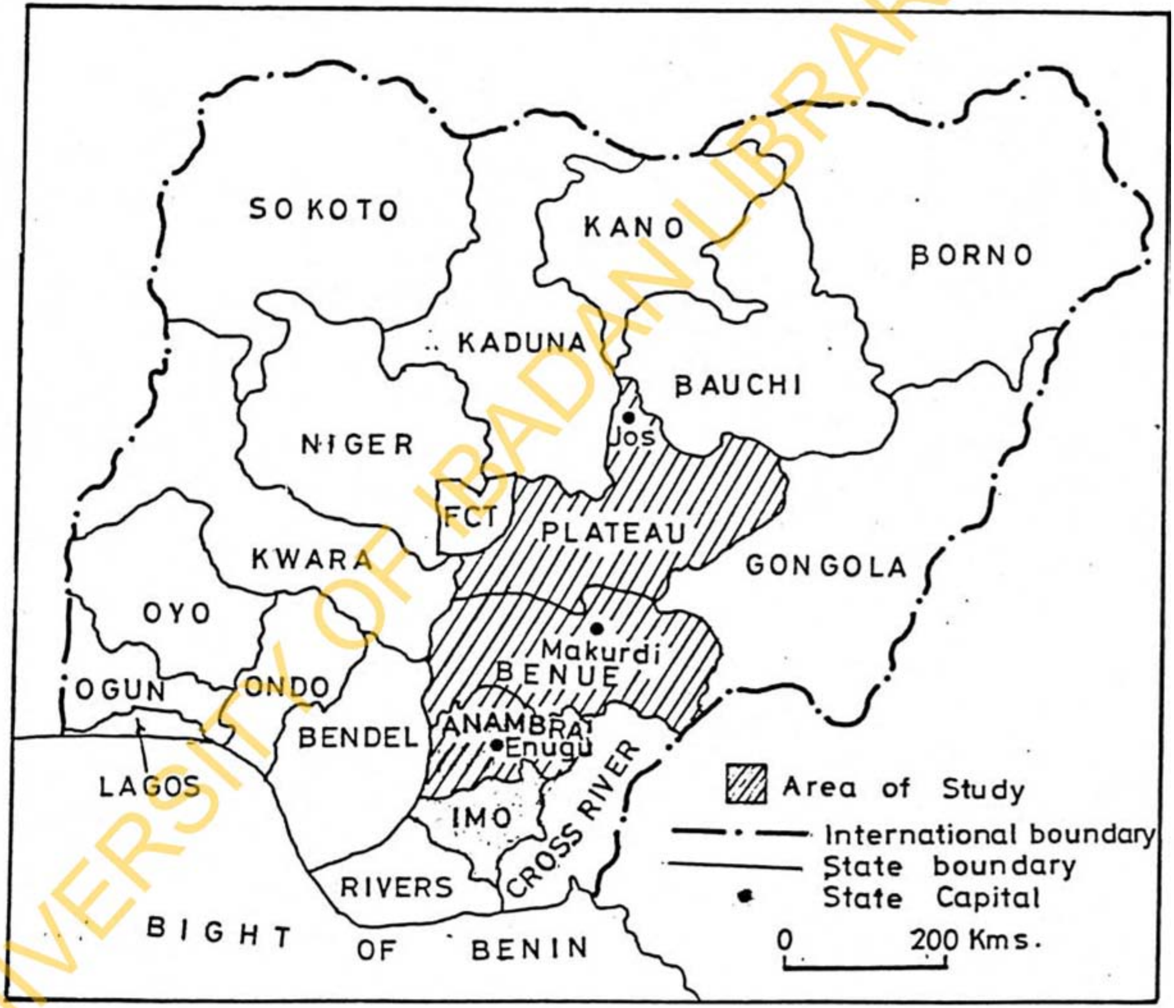


Fig. 16: Map of Nigeria showing Area of Study

TABLE 30

THICKNESS AND PERSISTENCE OF COAL HORIZONS IN LAFIA

Horizon No.	Thickness Of Coal Horizon (m)				No. Of Boreholes Intersections Of Horizons		Distance Above (+)/ Below (-) Horizon 12
	Total		Commercial				
	Range	Average	Range	Average	Total	Zero Thickness x	
36	0.10 to 0.63	0.30	-	-	4	2	+ 435
35	0.10 to 0.60	0.30	0.10 to 0.60	0.24	11	3	+ 345
34	0.15 to 0.49	0.28	0.15 to 0.49	0.28	13	9	+ 340
33	0.10 to 0.38	0.10	0.10 to 0.15	0.15	13	5	+ 335
32	0.10 to 0.20	0.11	0.10 to 0.20	0.10	22	16	+ 265
31	0.20 to 0.35	0.28	0.20 to 0.35	0.20	28	21	+ 245
30	0.10 to 0.94	0.32	0.05 to 0.45	0.16	30	13	+ 230
29	0.17 to 0.85	0.32	0.11 to 0.38	0.19	36	16	+ 210
28	0.05 to 1.29	0.38	0.05 to 0.46	0.22	36	14	+ 205
27	0.15 to 0.66	0.33	0.15 to 0.45	0.11	38	12	+ 188
26	0.05 to 1.35	0.35	0.05 to 0.50	0.11	39	17	+ 185
25	0.05 to 0.80	0.34	0.05 to 0.50	0.15	40	27	+ 170
24	0.05 to 1.90	0.51	0.05 to 0.75	0.26	45	6	+ 150
23	0.04 to 0.55	0.25	0.04 to 0.50	0.15	50	20	+ 137
22	0.05 to 1.04	0.41	0.05 to 0.55	0.18	51	10	+ 125
21	0.05 to 0.45	0.17*	-	-	54	30	+ 72
20	0.10 to 3.90	1.74	0.10 to 1.16	0.69	63	7	+ 60
19	0.05 to 1.70	0.53	0.05 to 0.86	0.27	65	4	+ 45
18	0.09 to 2.02	0.66	0.09 to 0.70	0.40	66	8	+ 39
17	0.05 to 0.60	0.22	0.05 to 0.48	0.10	69	36	+ 35
16	0.03 to 0.85	0.32	0.03 to 0.20	0.08	70	41	+ 31
15	0.04 to 1.10	0.26	0.04 to 0.52	0.08	68	36	+ 23
14	0.10 to 1.40	0.45	0.05 to 0.49	0.29	71	6	+ 17
13a	0.09 to 0.75	0.24	0.09 to 0.65	0.12	75	32	+ 10
13	0.10 to 1.35	0.72	0.10 to 0.74	0.50	75	2	+ 3
12	0.50 to 5.10	3.16	0.10 to 1.95	1.13	78	-	-
11	0.05 to 2.24	0.42	0.05 to 0.72	0.16	73	24	- 3
10	0.05 to 1.00	0.31	0.04 to 0.30	0.11	61	26	- 7
9	0.02 to 0.15	0.13*	-	-	45	42	- 12
8	0.05 to 0.47	0.22	0.05 to 0.36	0.10	35	17	- 20
7	0.10 to 0.90	0.34	0.10 to 0.48	0.24	32	5	- 30
6	0.10 to 1.10	0.29	0.10 to 0.49	0.14	17	4	- 35
5	0.05 to 1.27	0.43*	-	-	13	7	- 50
4	0.10 to 0.35	0.15*	-	-	8	2	- 65
3	0.05 to 0.19	0.12*	-	-	5	3	- 80
2	0.05 to 0.10	0.10*	-	-	2	-	- 95
1	0.15 to 0.15	0.15*	-	-	1	-	- 110

Source: v/o Technoexport report on Lafia Coalfield, November, 1976

* Horizons are represented by coaly argillite only.
x Inferred horizon intersection where horizon is missing.

3.2.3. Okaba

Since Okaba Coal Seams are near the surface, open-cast mining was done to collect the samples. A crane was used to collect coal samples randomly over a large field. The samples collected were loaded into metal drums which have been lined with polythene sheets. As usual, mine water was used to keep the coal wet before the latter was wrapped up in polythene and the drums sealed. The metal drums containing the Coal Samples were conveyed from the field to the laboratory in a Peugeot Pick-up Van.

3.3. LABORATORY SAMPLE PREPARATION FROM FIELD GROSS SAMPLES

3.3.1. Laboratory Sample Preparation for Enugu Coal

For Coal Samples from Enugu, the laboratory sample for laboratory tests was prepared from the common sample in three stages with sample drying in between [97]. The sample preparation guidelines for coal in Tables 31 and 32 were followed. From the residue of the Common Sample after extraction of the total moisture sample the retained sample weighing 60kg was air-dried and then crushed to 10mm.

Table 31: Minimum Mass of Sample to be retained during sample division

Maximum particle size after reduction	Two stage preparation		Three stage preparation	
	cleaned coals with 10% ash	All other coals.	cleaned coals with 10% ash	All other coals.
mm	kg	kg	kg	kg
10	1.5	10	2.5	15
5	0.5	3.5	0.75	5
3	0.3	2.0	0.45	3
1	0.15	0.6	0.25	1

Table 32: Air-drying times of Coal

Temperature not exceeding	Time not exceeding (hr)
up to 15°C above ambient, to 25°C, whichever is the lower.	24
30°C	6
45°C	3

Air-drying was done to ensure that the sample would pass through the mills and sample dividing equipment freely and without loss of moisture or contamination. Air-drying was carried out at any stage of the sample preparation procedure at which it was found to be necessary. The sample was exposed until visibly dry. The temperature and times of air-drying given in the table below were seen to be sufficient but, if it had been necessary, a longer drying time, but not a higher temperature, could have been used. When sample was dry, 15kg of it was taken and further crushed to 3mm and again air-dried. After drying, the Coal was riffled to obtain 3.75kg which was further crushed to 0.2mm, mixed, and divided to obtain a sample weighing 150kg which was the laboratory sample.

3.3.2. Laboratory Sample Preparation for Lafia/Obi Coal

The twenty barrels from Lafia/Obi containing coal shaft samples immersed in water were opened in the laboratory. The water was decanted till coal content showed in the drum. The coal content was then brough out using a small plastic bucket.
The contents of the twenty drums were heaped on nylon sheets spread over the clean cement pavement of the sample preparation shed.

The heap was spread out evenly at about 100mm in thickness of coal pieces. A rake was used to stir the coal layer from time to time to expose fresh coal surface to the laboratory air for drying. It was allowed to dry for 24 hours in the laboratory ambient air of 25°C.

The coal was then mixed together by shovelling it up into a cone and levelling the heap repeatedly ten times.

Finally, the air-dried consignment was spread evenly about 150mm to 200mm thick and the spread divided up into 33 squares as shown below (Table 33).

Table 33: Squares marked to facilitate random sampling of coal

1	2	3	4	5	6	7	8	9	10	11
12	13	14	15	16	17	18	19	20	21	22
23	24	25	26	27	28	29	30	31	32	33

Fifteen nylon bags were used as containers into which sample increments were placed. Using a shovel 150mm wide 3kg increments were taken from each numbered square and placed in each sample bag. The squares were sampled randomly but using no numbered square twice till all have been used once.

66 increments were taken to form a gross sample in each case. 15 gross samples were prepared and each was tied up in nylon bags to ensure it was air-tight. Labels were inserted inside each sample bag as well as being affixed outside the bag. Each bag of sample was weighed and after weighing the sample was in each case transferred back into clean metal drums that had covers. Water was introduced into each drum containing known quantity of coal till the latter was submerged. The covers were sealed to the drums by soldering to ensure air-tight condition to avoid coal oxidation.

3.3.3. Laboratory Sample Preparation for Okaba Coal:

Okaba Coal is young, reactive and very liable to air oxidation. Laboratory sample preparation for Okaba Coal was, therefore, only done within two hours to the time the work on the sample would be carried out.

Analysis sample weighing 60kg was crushed to 10mm and air dried. By coning and riffling manually 15kg of the gross analysis sample was taken and further crushed and ground to 1mm. Using mechanised riffling 3.75kg was taken out and ground to 0.2mm. 300g of laboratory analysis sample was finally taken up in a polythene sample bag for various tests.

CHAPTER FOUR

EXPERIMENTAL

4.1. Characterization of Lafia Coal

Lafia coal was characterized by physico-chemical methods to elucidate its nature, structure and composition. The results of the characterization guided subsequent decision on how best to handle the coal towards its desulphruization. Shaft samples and mor eparticularly the numerous bore-hole core samples were all examined in detail.

The approach adapted was to carry out proximate analysis, ultimate analysis, petrological microscopy and trace elements analysis. From the proximate analysis information was obtained on the coal with regards to its moisture, ash, volatile matter, and sulphur contents. The forms of sulphur in the coal were determined as to what proportion of the total sulphur was pyritic, organic or sulphate sulphur. The ultimate analysis involved the determination of elementary constituents of the coal like carbon, hydrogen, nitrogen, oxygen and sulphur varieties. Petrological microscopy was the means used to study the fossilised plant remains that make up the coal in their variegated assortments of maceral groups, macerals and general lithotypes.

Some physical constants examined were the density, friability and the behaviour under heat treatments. Precise and details of methods of how the parameters mentioned were carried out are given in the section on general methods of investigations. Also, in this section it is the proximate analysis results that is discussed in details as other results are described and commented on in the appropriate sections.

4.2. METHODOLOGIES

4.2.1. Introduction

Unless otherwise stated, British Standards Institution (BSI) Methods of Coal analysis, [98] were used for the determination of the parameters described here.

Since the study theme is on the desulphurization of Coal, a review of analytical methods for the determination of types of sulphur and total sulphur in coal will first be given, before giving detailed analytical procedures for determination of various physico-chemical parameters in coal.

4.2.2. Review of the Methods of Determination of forms of Sulphur in Coal

In most cases, coal analysis for forms of sulphur - sulphate, pyritic and organic - the sulphate and pyritic sulphur are determined directly and the organic sulphur is taken as the difference between the total sulphur and the sum of the sulphate and pyritic sulphur [99].

The methods of most general use are based on the well-known Powell and Parr procedure [104] developed in the early thirties. The British Standard [100], Fuel Research Board [101], U.S. Bureau of Mines [102], and Spanish [103] methods are based on this work.

In the Powell and Parr method the sample is cold digested at room temperature with hydrochloric and nitric acids over a long period of time for the determination of sulphate and pyritic sulphur. Powell pointed out that time of determination could be shortened if the sulphate determination only is done with hot hydrochloric acid digestion.

Teichmann [105] suggested use of stronger (16% or 5M) Hydrochloric acid for a shorter period of 1 hour or 30 minutes to avert possible oxidation and solution of the pyrite that goes with prolonged extractions with dilute hydrochloric acid.

He used coal powder of minus 240 mesh BS for one hour extraction at the boiling point.

Mott [106] extracted the pyritic and sulphate sulphur varieties with both hydrochloric and nitric acids at boiling temperature and thus ensured that the hydrochloric acid did not attack the organic sulphur. He saved time by this procedural modifications which took only 30 minutes for extraction.

Edwards, Daybell, and Pringle [107] carried out a comprehensive review of methods of determination of forms of sulphur and confirmed that the British Standards and the non-simultaneous procedure of the Fuel Research Board give practically identical results, but both are liable to a slight error affecting both sulphate and pyritic sulphur. The British Coke Research Association method is not so subject to oxidation effects and is recommended where highest accuracy is required. The conclusion of Teichmann was confirmed that some oxidation of pyrite takes place by prolonged hydrochloric acid digestion for determining non-pyritic sulphur.

Radmacher and Mohrhauer [108] reduced pyrite with granulated zinc and chromium in acid solution to hydrogen sulphide, H_2S, by the action of nascent hydrogen. The coal has to be powdered finely to liberate all the pyrite for easy access by the metallic reductants to react to give off nascent hydrogen as H_2S.

Organic sulphur in coal had been determined by taking a 50mg sample which was put into 10ml of 25% by volume of nitric acid for 24 hours. Thereafter the sample was filtered, washed, dried and analysed for sulphur in the Leco combustion furnace. The sulphur so determined was considered to be organic sulphur. The difference between the total sulphur and the organic sulphur in the sample was designated inorganic sulphur.

4.2.3. A Review of Methods of Determination of total sulphur in coal

Methods of analysis for sulphur include combustion, reduction, X-ray analysis, neutron activation and cool flames.

Combustion methods are generally carried out in an oxidizing environment and result in production of oxides of sulphur which are eventually converted to the sulphate form that can be determined either gravimetically or volumetrically.

Eschka method [109] for total sulphur determination, developed over a century ago, continues to be one of the best known and is accepted as a national standard in several countries. It has distinct advantages in that special apparatus is not required and only simple analytical techniques are employed.

Basically, the method consists of incinerating initmate mixture of coal or coke with a mixture of 1 part sodium carbonate and 2 parts of light calcined magnesium oxide. The sulphur compounds evolved during combustion react with the sodium carbonate and under oxidizing conditions are retained as sodium sulphate. The ligh magnesium oxide reduces the bulk density and allows access of air to the mass. The sulphate in the residue is extracted in either alkaline or acid solution and determined gravemetrically as barium sulphate or by volumetric methods. With suitable heating rates and adequately ventilated electric muffles, loss of sulphur during incineration does not occur. In addition, by supplying enough air, the sulphur is completely oxidised to sulphate, so that use of bromine to oxidize sulphites is not required [110, 111].

Bomb-combustion methods have long been used for determining total sulphur in solid fuels and are specified as an alternative method in several national standard [112]. The method is favoured by many analysts because of its speed and the assurance that no sulphur is lost during the combustion. It is especially favoured when the determination of calorific value is required as well as sulphur. The two methods of decomposing the sample in the bomb are the Parr fusion procedure [104] with sodium peroxide and secondly the combustion in oxygen at pressures of 20 to 30 atmospheres.

The Parr method gives results in agreement with the Eschka method but required special apparatus.

The oxygen bomb-combustion method is objected to from time to time because it is found with certain coals that a significant portion of the sulphur is retained in the fused cenoshpere of ash [113]. To obtain correct results, the sulphur retained in the ash must be extracted and added to the bomb washings. To overcome this problem, Strambi [114] introduced a modified method in which 1g of coal mixed with Eschka mixture and sucrose and burned in a calorimeter bomb under a pressure of 25 atmospheres of oxygen. Sucrose was added to insure complete combustion of fuels difficult to ignite. The sulphur oxides formed were absorbed in a solution of sodium carbonate in the bomb and the sulphate formed determined gravimetrically. Strambi reported that the method gave higher results for high sulphur coals than the Eschka method and with better precision. The method also is reported to be more rapid than the Eschka when only a small number of samples is to be analysed. The titrimetric finish with barium chromate also can be applied.

A high temperature combustion method based largely on the work by Suthe [115] and by Beet and Belcher [116] has been adopted in many laboratories, particularly in Europe, and is an alternative German and British standard method [117].

One of the particular advantages claimed for the method is that chlorine can be determined at the same time. Briefly the method consists in heating 0.5g sample of coal in a boat placed in an electric furnace heated to 1250°C or 1350°C. The coal is usually covered with kaolin, ferric phosphate, or aluminium oxide to assist in the decomposition of the sulphate and prevent its retention in the ash [118]. Oxygen is passed through the furnace, and the oxides of sulphur are absorbed in hydrogen peroxide which oxidises any SO_2 to SO_3. At the end of the test, the solution is titrated with 0.05M sodium borate solution which gives the total acidity due to the hydrochloric and sulphuric acids formed. The chloride is then determined by adding mercuric oxycyanide solution to convert the sodium chloride to sodium hydroxide according to the equation.

$$\mathrm{Nacl} + \mathrm{Hg(OH)CN} \longrightarrow \mathrm{HgClCN} + \mathrm{NaOH} \qquad \_\_\_\_\_\_ \quad \text{eqn. } 15$$

The liberated alkali then is titrated with 0.025M sulphuric acid. The calculations for obtaining the percentage of sulphur and chlorine are as follows:

If a cm^3 of 0.5M sodium borate is used in the determination,

a_1 = cm^3 of 0.05M sodium borate used in the blank,

b = cm^3 of 0.025M-H_2SO_4 used in the determination,

b_1 = cm^3 of 0.025M-H_2SO_4 used in the blank and

w = weight of coal in grams, the % of sulphur,

$$\%S = \frac{0.08}{w}\left[(a - a_1) - \frac{(b - b_1)}{2}\right] \qquad \underline{\hspace{2cm}} \qquad \text{eqn. 16}$$

and the % of chlorine

$$\%\ Cl = \frac{0.08875\ (b - b_1)}{w} \qquad \underline{\hspace{2cm}} \qquad \text{eqn. 17}$$

In the reductive fusion method the sulphur compounds in coals are converted to hydrogen sulphide by fusion with metals. Usual analytical methods are employed to estimate the sulphur evolved from the hydrogen sulphide obtained in the first step. Use is cadmium acetate to absorb H_2S evolved. The lithium metal was employed for this purpose [119]. It has been reported that fusion with lithium gave results as accurate as those from the Eschka and high-temperature combustion methods.

Edgecombe [120] determined sulphur and chlorine in coal simultaneously by converting them to sulphuric and hydrochloric acids, separating them by ion-exchange and determining them titrimetrically or potentiometrically.

Fluorescent X-ray photons with wavelengths characteristic of the element are generated when the sample is irradiated with high intensity x-rays. The frequency of occurrence of such photons is proportional to the amount of the element present.

The apparatus consists of a high-energy x-ray tube, the power plant for the x-ray tube, and a crystal spectrograph. The measurements are carried out electronically for which proportional detectors or counters, scintillation detectors or counters, or Geiger detectors/counters may be used. Sodium chloride has been found to be the most suitable crystal for sulphur.

The sample is palced in the x-ray beam and the intensity of the sulphur K& line at 5.372°A is measured.

However, attention has been drawn to the disturbing influence of particle isze in x-ray fluorescence analysis of sulphur in coal. In the case of coals having an ash content below 10% these disturbing influences can be eliminated by suitable grinding.

British Coke Research Association [121] (Chesterfield, Derbyshire) carried out a rapid determination of sulphur in coal by x-ray fluorescence spectrometry. Coal samples are crushed to pass through a minus 240 mesh sieve (particle size 0.065mm) and the powder is compressed into an aluminium sample holder. The intensity of the sulphur K& line is measured. Results in good agreement with those of chemical analyses were obtained when correction were applied for the incoherently scattered radiation (Cr K& compton) of the x-ray tube and for Si K& radiation.

Goodby and Stephens [122] found that x-ray fluorescence gave adequate accuracy and sensitivity (lower limit of detection about 50ppm) with fly-ash containing between 0.1 and 0.5 percent of sulphur.

Brown [123] compared the sulphur K& count-rates of 31 soils with their chemically determined sulphur contents. 16 Malaysian coastal swamp soils were chosen for their wide range of sulphur content, (0.05 to 3.56 percent) and loss on ignition (10 to 90 percent); 12 Rothamsted soils with loss on ignition less than 10 percent and 3 Australian soils with small loss in ignitions were used.

When there are no matrix effect and the chemical and x-ray determinations are free from erros (including sampling errors) a graph of sulphur K& count-rate against sulphur content, determined chemically, would be a straight line. When such a relationship exists, as it does approximately for the Rothamsted and Austrialian soils, the sulphur content can be determined directly from the S K& count-rate. For soils that did not conform to this relationship a simple method of correction based on the loss of ignition at 450°C was developed [124].

Volumetric methods have been suggested for determination of sulphate ion because they are more rapid [125]. These methods include i) the use of barium chromate, ii) conductometric titration of bomb washings, iii) direct titration with barium perchlorate, iv) titration with standard barium chloride, v) precipitation of sulphate with 4-amino-4=chlorobiphenyl hydrochloride, vi) dissolution of barium sulphate in standard versenate solution followed by titration against standard solution of magnesium chlorite, and vii) amperometric titration with lead nitrate instead of the gravimetric finish in the Eschka method.

Radmacher and Morhauer [126] used $BaCrO_4$ to precipitate $BaSO_4$ with the excess $BaCrO_4$ determined volumetrically.

Elphick and Gerson [127] described a method for estimating sulphur in coal by conductometric titration of the washings from a bomb calorimeter.

Wilkinson [128] precipitated the sulphate with 4-amino-4-chlorobiphenyl hydrochloride and titrated a suspension of the precipitates with standard NaOH.

Majumdar and Benergee [129] suggested a method in which the barium sulphate precipitate is dissolved in an excess of a standard solution of versenate and the excess versenate titrated against a standard solution of magnesium chloride.

Gordiyeviskii and Radnayeva [130] replaced the final gravimetric step in the Eschka method by amperometric titration with lead nitrate, $Pb(NO_3)_2$.

Hicks (1973) [131] has suggested a titrimetric method for rapid determination of sulphur in coal. In this method, after the sample has been burnt in the combustion bomb under 25 atmospheres oxygen to determine the calorific value, the bomb washing are carefully transferred to a beaker and neutralized, and the sulphate produced during combustion is estimated by titration with a standard solution of lead perchlorate.

Petrographic methods are used for examination of sulphur in coal and its desulphurization products. The distribution of pyritic grains can be examined by the microscope. A microscopic examination is helpful in giving information as to the abundance or scarcity of pyrite. If the pyrite sulphur content is high, mechanical cleaning processes would be helpful.

Scanning electron microscope (SEM) sees and compares the relative density and distribution of all sulphur species in a coal sample. Pieces of information obtained from SEM serves as a good guide in coal cleaning, preparation and utilization. The distribution of inorganic elements e.g. Fe and Ca in association with the sulphur can also be studied under SEM.

Gas chromotography can be applied to the determination of volatile sulphur products of combustion and/or hydrogenation. Oxidation to sulphur dioxide by combustion and reduction to hydrogen sulphide by hydrogenation have been employed. In this instrument, a solute undergoes distribution between two phases, one fixed (stationary) and the other moving (mobile phase). The moving phase is a gas and the fixed phase is a liquid or solid, or a liequid deposited on a solid support. The carrier gas moves the solute (a particular compound), through the column. The compund, separated from other compounds (according to its retention), comes out from the end of the column. The concentrations of the emerging components are successively measured and automatically recorded on a chart.

For measuring sulphur dioxide formed by combustion of organic sulphur compounds a 6m column containing 30% dinoylphthalate at 92°C was found useful (Beauerman and Meloan, 1962) [132]. By this process, sulphides, disulphides, sulphoxides, sulphones, thiones, and thioethers could be estimated, but sulphates cannot be converted to sulphur dioxide quantitatively. Total sulphur can also be determined as hydrogen sulphide by a combination of hydrogenation and gas chromatography (Thompson and Allbrigh, 1970) [133].

4.3. SAMPLE PREPARATION FOR LABORATORY ANALYSIS

Run-of-mine, shaft and core samples were collected from the coal fields. Once the core samples got to the laboratory they were equilibrated with the laboratory atmosphere by spreading them on a clean card-board and left for twenty-four hours. It was only the bed-moisture that was immediately determined as the sample arrived in the laboratory. When the samples had equilibrated with the prevailing atmosphere of the laboratory they were prepared for analysis. Roll crushers or manual grinding were used to pulverize the coal. For laboratory analysis sample preparation minus 72 mesh sieve was used to sieve 100% of the pulverized sample. Proximate and ultimate analyses were then carried out on the minus 72 mesh size coal samples. The methods used to carry out both the proximate and ultimate analyses of the coal analysis samples are described below.

4.4. METHODOLOGIES FOR PROXIMATE ANALYSIS OF COAL

4.4.1. Determinaiton of the Specific Gravity of Coal

Principle

Archimedes principle was applied in the determination of the specific gravity [134] of coal piece suspended in water. It experienced an upthrust equal to the volume of water it displaced.

Apparatus

Analytical balance made by Satorius with digital automatic weight read-out was used for weighing. Distilled water was used as the medium in which the coal piece was suspended and weighed. Tough fine thread was tied around the coal piece. A 250-ml glass beaker served as the container for water.

Procedure

The tough fine thread was tied round the coal piece which was then suspended in the still air of the weighing balance and its weight, w_1g, read out from the balance. Next the suspended coal piece was immersed in distilled water in a beaker and its weight w_2g read off from the balance.

Calculation

weight of coal piece = w_1 g

weight of coal piece in water = w_2 g

Upthrust of water on coal piece $(w_1 - w_2)$g.

Therefore, the Specific Gravity of coal piece $= \frac{w_1}{w_1 - w_2}$

The repeatability of duplicate determinations was limited to ±0.1.

4.4.2. DETERMINATION OF MOISTURE IN COAL

Principle

Direct gravimetric determination was the method used. A known mass of the sample was heated in a glass tube under a current of nitrogen at 105°C to 110°C. The water evolved was collected in a pre-washed absorption tube. The mositure in the sample was calculated from the increase in mass of the absorption tube. This moisture was required for calculating other results to dry basis.

Special Reagents

The special reagents used were:)1 ultra-pure nitrogen gas containing less than 10ppm of oxygen; 2) magnesium perchlorate with particle size of 0.9mm to 1.2mm.

Apparatus

The set-up of the apparatus used was as indicated on figures 17 and 18 and the components of it were:

1) A block type oven capable of maintaining its temperature within the range 105°C to 110°C and of accommodating a glass retort tube containing the coal sample;

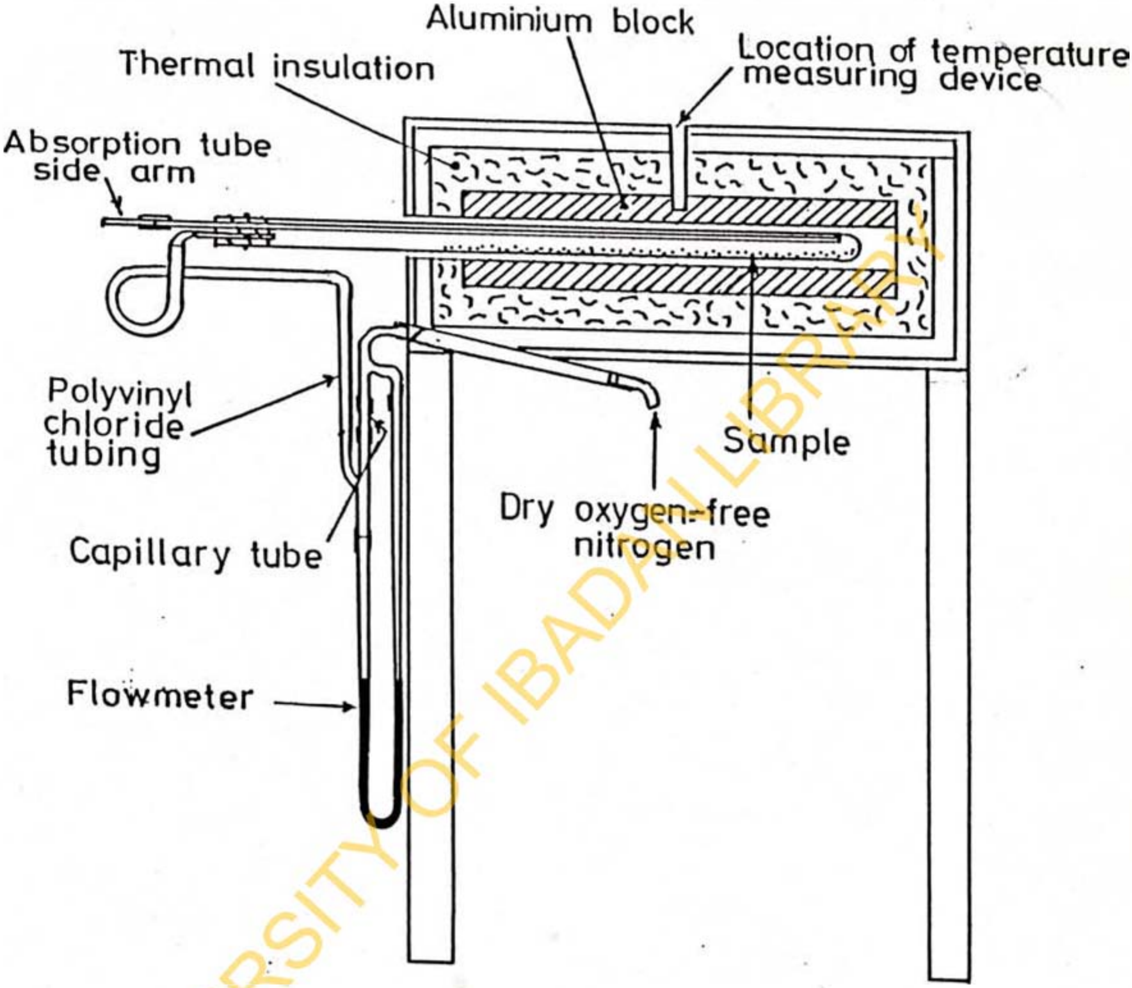
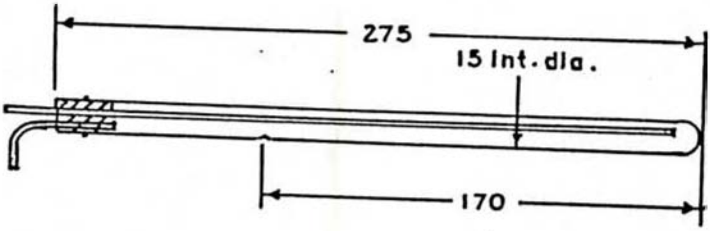


Fig. 17. Apparatus for the direct determination of moisture.



Dimensions in millimetres

Fig. 18: Retort tube

2) A glass retort tube, 275mm long and having a 15mm internal diameter with a depression 170mm from the sealed end, fitted with a flexible, heat resistant stopper through which passed two glass tubes. One of the tubes, the inlet tube, was connected to the flow-meter and terminated just inside the stopper. The other, the outlet tube, was connected to the absorption tube and passed down the retort tube almost to the sealed end. The connection between the flow-meter and the inlet tube was made with polyvinyl chloride tubing. The connection between the outlet tube and the absorption tube was made with a short length of rubber tubing;

3) A drying tower, 250ml capacity, packed with magnesium perchlorate for drying the nitrogen.

4) A flowmeter containing a non-volatile oil and capable of measuring a flow rate of 100 ml per minute.

5) An absorption U-tube fitted with glass stoppers and filled with magnesium perchlorate retained by plugs of glass wool.

Procedure

The rate of flow of the nitrogen passing through the flowmeter was adjusted to 100 ml/min. An empty retort tube was connected to a closed absorption tube and to the nitrogen stream from the flowmeter. The connections were made leak-proof.

The retort tube was inserted into the oven heated to 105°C to 110°C. After 15 minutes the taps of the absorption tube were closed, disconnected, wiped with a clean dry cloth free from loose fibres and the absorption tube was allowed to stand for 20 min in the balance room. The taps of the absorption tube were opened momentarily to equalize the air pressure and it was then weighed to the nearest 0.1mg.

While the absorption tube was standing prior to weighing, the retort tube was removed from the oven and allowed to cool with a stream of dry nitrogen passing through it. When cooled the retort tube was removed, with the stopper and tubes still in it, to the balance room.

About 1g of the coal sample was weighed on a scoop and transferred to the retort tube, removing the stopper and tubes just prior to the introduction of the sample.

The coal was spread in an even layer between the sealed end and the depression in the retort tube. The stopper and tubes were replaced. The apparatus was re-assembled and the connections made leak-proof. Passage of nitrogen at the rate of 100ml/min was resumed.

The charged retort tube was inserted into the oven. After 60min the taps of the absorption tube were closed, disconnected, cooled and weighed on a satorius weighing balance.

A blank determination was done using the same procedure as for the moisture determination but omitting the coal sample.

Calculation

m_1 = mass of coal used (g)

m_2 = mass of absorption tube before the determination (g)

m_3 = mass of absorption tube after the determination (g)

m_4 = mass of absorption tube before the blank determination (g)

m_5 = mass of absorption tube after the blank determination (g)

Mad = percentage of moisture in the analysis sample

then

$$\text{Mad} = \frac{(m_3 - m_2) - (m_5 - m_4)}{m_1} \times 100$$

The difference allowed between duplicate determination

4.4.3. DETERMINATION OF ASH IN COAL

Principle

A known mass of the sample is heated in air to 500°C in 30 min, from 500°C to 815°C in 60 to 90 min and is then kept at 815°C until constant in mass. The percentage of ash is calculated from the mass of the residue remaining after incineration.

Apparatus

The apparatus used in determining ash in coal included weighing balance, muffle furnace, and dish.

Satorious, analytical balance was used for weighing.

The muffle furnace used was made by Carbolite of Great Britain. It gave an adequate zone of uniform temperature that could be raised from room termperature to 500°C ± 10°C in 30 min and from 500°C to 815 ± 10°C in a further 60 to 90 minutes and that maintained that maximum temperature to the end of the experiment. The ventilation was such that gave at least four atmosphere changes per minute at 815°C.

A silica dish, 10mm to 15mm deep, 20mm diameter with cover was used.

Procedure

A clean dry empty dish was weighed together with its cover to the nearest 0.1mg. About 1g of coal sample was spread into the dish in an even layer and the cover was replaced. It was weighed to determine the mass of the sample taken.

The cover was placed in a desiccator while the uncovered dish and its contents were placed in the muffle furnace at room temperature. The temperature was raised to 500°C in 30 minutes and to 815°C in a further 60 to 90 minutes and maintained at that temperature until constant in mass. When incineration was completed the dish was removed from the furnace, the cover was replaced and allowed to cool, first on a thick metal plate for 10 minutes and finally in a desiccator for a further 15 minutes. The covered dish was weighed, the ash brushed out completely and the empty dish and cover reweighed. The mass of the ash was obtained by calculating the difference.

Calculation of result

m_1 = mass of dish plus cover (g)

m_2 = mass of dish plus cover plus sample (g),

m_3 = mass of dish plus cover plus ash (g),

m_4 = mass of dish plus cover after ash has been brushed out (g),

A = percentage of ash in the analysis sample,

then

$$A = \frac{m_3 - m_4}{m_2 - m_1} \times 100\%$$

Repeatability (% absolute) was 0.15, 0.20 and 0.25 respectively for samples with ash less than 10%, 10% to 20% and more than 20%.

4.4.4. DETERMINATION OF VOLATILE MATTER CONTENT IN COAL

Principle

A known mass of the coal analysis sample was heated at 900°C out of contact with air at 900°C for 7 min. It was essential to exclude air from the coal during heating to prevent oxidation and therefore the fit of the lid of the crucible was critical. The volatile matter was claculated from the loss in mass of the sample; a deduction was made for the loss in mass due to the moisture. The moisture in the analysis sample was determined along with the volatile matter content determination.

Reagents

Desiccant of self-indicating silica gel was used in the desiccator.

Apparatus

The apparatus used included weighing balance, muffle furnace, thermocouples, crucible and lid, crucible stand and asbestos discs.

1) Weighing balance

The weighing balance was manufactured and marketed by Satorius of Great Britain. It read to the nearest 0.1mg automatically.

2) Muffle furnace

An electrically-heated muffle furnace, manufactured by Carbolite of the United Kingdom, in which an adequate zone could be maintained at a uniform temperature of 900 ± 5°C was used for heating up the samples. The internal dimensions of the muffle was 250mm long by 100mm wide by 65mm high. It was provided at the front with a well-fitting, readily manipulated door. Its heat capacity was such that with an initial temperature of 900°C a minimum temperature of 885°C was regained within 3 min of the insertion of a cold stand and its crucible. Both temperatures were measured with an unsheathed thermocouple. The temperature was automatically controlled and the power rating of the furnace was 1750Watts. A position was chosen for the crucible stand within the zone of uniform temperature and this position was used in all determinations.

3) Thermocouples

Two thermocouples were used, one sheathed and permanently fixed in position and the other unsheathed and inserted periodically for checking the temperature at the base of the crucibles. This method of checking at the base of the crucible was used because the sheated thermocouple cannot be inserted under the crucible and a thermocouple with a bare junction constantly exposed under the specified conditions may change its characteristics.

The temperature of the uniform zone of the muffle furnace was checked periodically by means of an unsheathed thermocouple of wire not thicker than 1mm. The thermocouple was inserted midway between the base of the crucible in its stand and the floor of the muffle furnace and its reading related to that of the sheathed thermocouple whose position is shown in Figure 18. The sheathed thermocouple also served to locate the stand in the zone of uniform temperature.

4) Crucible and lid.

A cylindrical crucible with capsule type lid, both of translucent silica, having dimensions approximately to those shown in Figure 19 was used. The crucible and lid were matched so that their combined mass was between 10g and 14g. Since the fit of the lid was critical the lid was selected for each crucible so that when placed in position the maximum horizontal clearance around the lid of the crucible was 0.5mm. After selection each crucible and lid was ground together to give smooth surfaces and then identified with a distinguishing number (or mark).

5) Stand.

A stand constructed from heat-resistant steel wire [135] for single and for multiple determinations as shown in Figures 20 and 21 were used. The upper and lower surfaces of the handle of the single crucible stand were ground flat to facilitate handling with tongs.

6) Asbestos discs

The asbestos discs used were each 25mm in diameter and 1mm thick.

Procedure

The empty crucible and lid supported on two asbestos discs in the stand were heated at 900°C for 7 minutes. The crucible was removed from the furnace and cooled, first on a metal slab for about 5 minutes and finally in a desiccator besides the weighing balance. As soon as it was cold the empty crucible and lid were weighed to the nearest 0.1mg. About 1 ± 0.01g of the coal analysis sample was weighed on a scoop. The sample was carefully and completely transferred into the crucible. On the clean hard glass surface of the weighing table the crucible was given some taps until the coal formed a layer of even thickness on the bottom of the crucible.

The temperature of the muffle furnace was adjusted to 900°C as indicated by the correctly located unsheathed thermocouple and containing a stand and an empty crucible and its lid. The empty crucible, its lid and stand were removed and the door of the furnace closed to restore steady conditions, (Figures 19,20, 21, 22).

Next the covered crucible containing the sample was placed in a cold stand fitted with two asbestos discs.

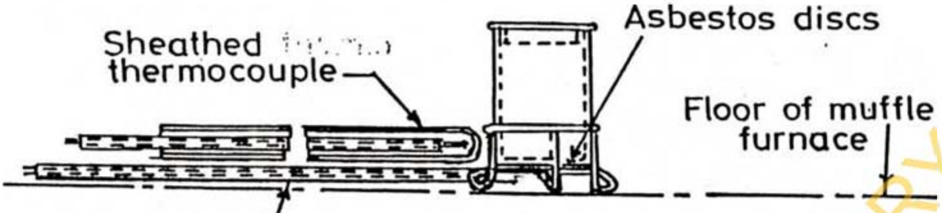
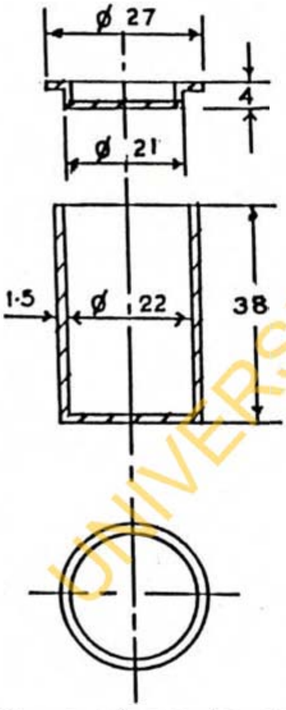


Fig. 19. Determination of Volatile matter: Location of thermocouples for single crucible stands.



Dimensions in millimetres

Fig. 20: Silica crucible and lid.

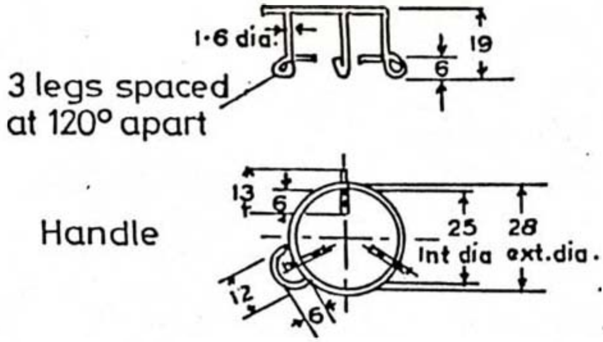
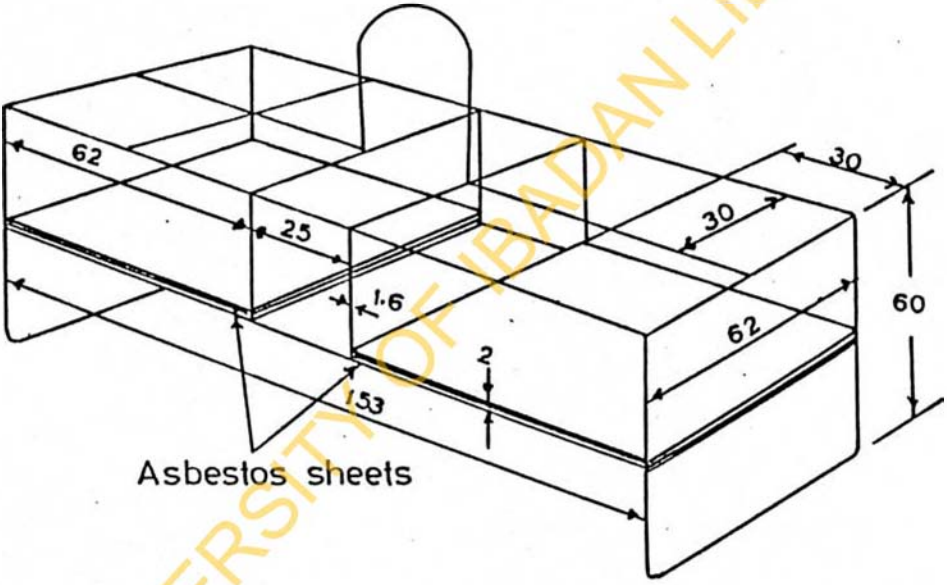


Fig. 21. Single type crucible stand.



Dimensions in millimetres

Fig. 22. Multiple type crucible stand

The stand and crucible were transferred into the muffle furnace and heated for exactly 7 minutes from the time of insertion. The stand and crucible were then removed and allowed to cool for about 5 minutes on a metal slab after which the crucible and its contents were cooled in a desiccator. The covered crucible and its contents were then weighed.

Calculation of result

m_1 = mass of crucible plus lid (g),

m_2 = mass of crucible plus lid plus sample before heating (g),

m_3 = mass of crucible plus lid plus sample after heating (g),

Mad = percentage of moisture in the analysis sample,

V = percentage of volatile matter in the analysis sample,

then,

$$V = \frac{m_2 - m_3}{m_2 - m_1} \times 100 - \text{Mad}.$$

Volatile matter on dry ash free (d.a.f) basis,

$$(V^r) = \frac{V}{100-A} \%$$ where A = % ash of sample

In samples with CO_2 higher than 2%

$$V = \frac{m_2 - m_3}{m_2 - m_1} \times 100 - Mad - \%(CO_2)$$

Repeatability of duplicate determination was limited to 0.6% where V was less than 45%.

4.5. METHODOLOGIES FOR ULTIMATE ANALYSIS OF COAL

4.5.1. Introduction

The ultimate analysis of coal involved the determination of carbon, hydrogen, nitrogen and sulphur contents of the coal. The sample used was the analysis sample that had a top size not greater than 0.2mm (ground to pass 0.212mm test sieve). The sample was brought into equilibrium with the laboratory atmosphere by exposing it in a thin layer on a tray for the minimum time required to avoid oxidation (which would otherwise alter the values of some parameters). The sample was thoroughly mixed manually in a cylindrical glass bottle immediately before the determination. Duplicate determination were made.

4.5.2. DETERMINATION OF CARBON AND HYDROGEN IN COAL SAMPLES

Principle

A known mass of coal was burned in a rapid current of oxygen at a temperature of 1350°C so that all the carbon was converted to carbon dioxide (CO_2) and all the hydrogen to water (H_2O). Chlorine and oxides of sulphur were retained in the apparatus by silver ganze.

The water formed was absorbed by magnesium perchlorate and the carbon dioxide by soda-lime. A determination of moisture was carried out at the same time and the value for hydrogen was corrected for that present as moisture in the sample.

Reagents

All reagents used were of analytical grade quality. Deionized water was used in preparing solution. Other reagents used were:

1) Magnesium perchlorate
particle size of the perchlorate was 1.2mm to 0.7mm.

2) Soda lime
2.0mm to 1.2mm particle size of it was used.

3) Aluminium oxide
The oxide was finely divided, approximately 0.1mm.

4) Sodium borate solution, 0.05M.

5) Hydrogen peroxide solution
The solution contained 3 parts of 100 volume H_2O_2 and 97 parts of water.

6) Screened indicator
0.125g of methyl red was dissolved in 100ml of ethanol (95 v/v) and marked solution A while 0.083g of methylene blue was dissolved in 100ml ethanol (95 v/v) and marked solution B, stored in a dark glass bottle. Solutions A and B were mixed in equal volumes immediately before use.

7) Pure silver gauze, one mesh per millimetre, of 0.3mm diameter wire.

8) Oxygen gas that was hydrogen free.

Apparatus

The apparatus used are shown in Figures 22 and 23.

1) Purification train made up of
   - a) magnesium perchlorate for removing water,
   - b) soda-lime for removing carbon dioxide and
   - c) magnesium perchlorate for removing water produced in the reaction between carbon dioxide and soda lime.

2) Electrical heating furnace designed to carry a tube of 28.5mm outside diameter and to heat it over a length of about 128mm to give a temperature of 1350°C at the centre of the hot zone and a temperature distribution curve shown in Figure 23.

3) Combustion tube.

   This was a tube, 22mm internal diameter and 28.5mm outside diameter by 0.6mm long, made of refractory aluminous porcelain that was not permeable to gases at 1400°C.

4) Combustion boat

   The combustion boat was of iron-free unglazed porcelain, 70mm long by 12.5mm wide by 10mm deep, that did not blister, discolour or change in weight on heating at 1350°C in oxygen for 3 hours.

5) Silica pusher

The pusher was constructed of rod of silica, 6mm diameter by approximately 450mm long, flattened at one end to form a disc of 12mm diameter for pushing the boat into the furnace. The pusher was marked in millimetres from the disc end for convenience in measuring how far the boat had been pushed into the combustion tube.

6) Nickel-Chromium wire

This was a length of stiff nickel-chromium wire with a hooked end, to extract the boat from the furnace on to a piece of refractory tile.

7) Absorption train

The train [Figure 24] contained in glass bottles and in the order magnesium percholrate to absorb water, soda lime to absorb carbon dioxide, and magnesium perchlorate to absorb water produced in the reaction between carbon dioxide and soda lime. Glass wool was placed above and below the absorbent to prevent carry-over of dust by the rapid flow of oxygen and also to reduce the risk of the heat of the reaction cracking the tube containing the soda lime.

8) Flow-meters

Two flow-meters were used, one capable of measuring rates of flow of oxygen of up to 300ml/min and the other capable of measuring rates of flow of air of up to 250ml/min.

9) Pressure gauze

A manometer that measured the back pressure on the system, between 5mbar and 7mbar (1 mbar = 10^2 N/m^2).

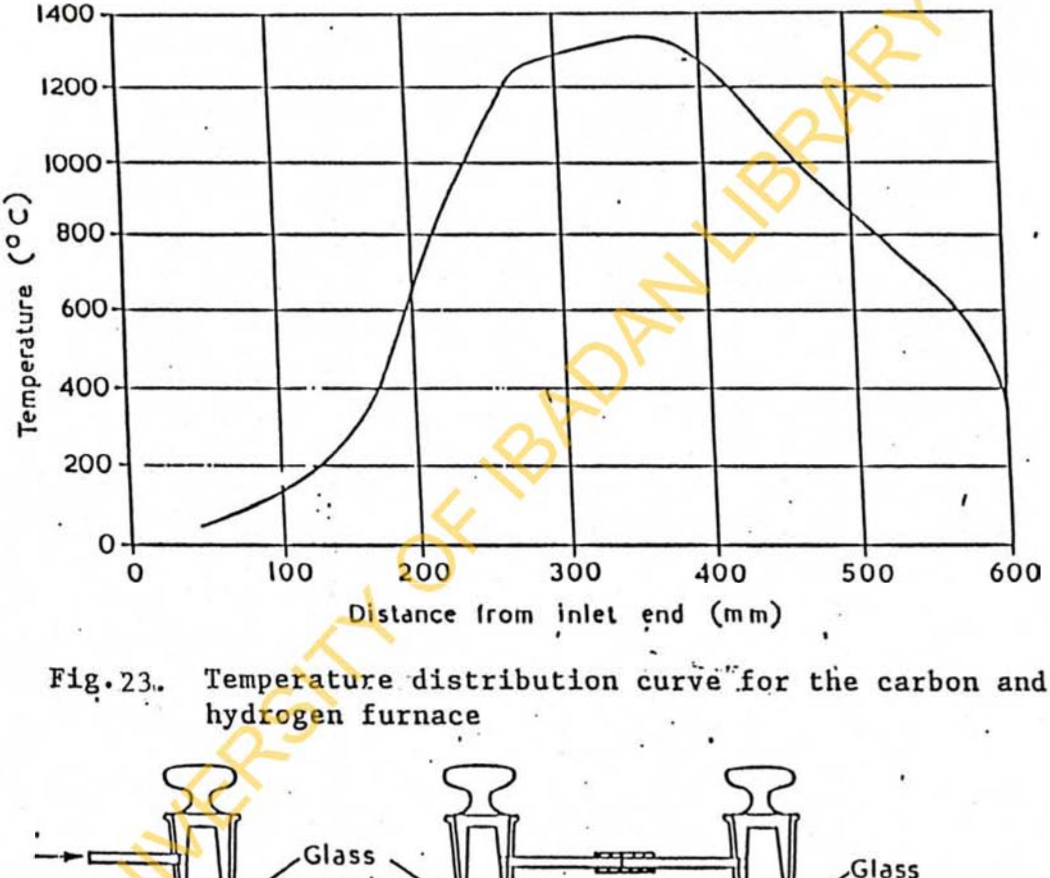


Fig. 23. Temperature distribution curve for the carbon and hydrogen furnace

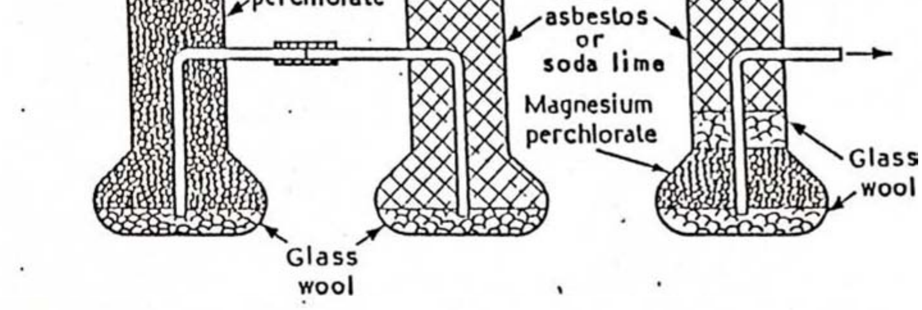


Fig. 24, Absorption train for carbon and hydrogen determination

10) Heat resistant stopper which was used for attaching the absorption train to the combustion tube.

11) Silver gauze roll

    The silver gauze roll had a stout silver wire passed through it for ease of removal. 75mm long and of sufficient diameter to ensure a close sliding fit in the combustion tube.

CALIBRATION OF APPARATUS

The combustion tube was inserted into the furnace so that it projected 75mm at the exit end. The projecting portion was wrapped with asbestos string to prevent condensation of water in the tube. The rubber stopper carrying the silica pusher at the inlet end of the combustion tube was inserted and oxygen supply was connected to the limb of the T-piece. The silver gauze roll was inserted into the exit end of the combustion tube to a distance of about 75mm.

With furnace at its working temperature of 1350°C a bubbler containing hydrogen peroxide solution was connected by means of the heat-resistent stopper to the exit end of the combustion tube.

With oxygen flow rate adjusted for the optimum, about 0.5g of the coal sample was burnt. The amount of oxide of sulphur passing the roll was determined by titration of the hydrogen peroxide with the sodium borate solution using the screened indicator. Combustion of 0.5g portions of the sample was repeated moving the silver gauze roll in 5mm steps, until a position was found where the titre of the sodium borate solution was not more than 0.2ml. That indicated retention of at least 99% of the oxides of sulphur by the silver gauze roll. This location was used in all determinations. When the combustion tube was not in use, it was protected from atmospheric contamination by fitting stoppers.

PROCEDURE FOR BLANK DETERMINATION

The temperature of the furnace was raised to 1350°C and oxygen was passed through the combustion tube at a rate of 300ml/min. Absorption train, (Figure 24) was connected to the apparatus by means of a heat resistant stopper and the set-up purged for 10 minutes. It was ensured that the inlet of the water-absorption tube was flush with the inner end of the heat resistant stopper in the combustion tube and that the joints between the absorption tubes were made glass to glass, using the rubber tubing merely as a seal.

The absorption train was disconnected from the combustion tube and connected to the air purification train. Purified air was drawn through the train at a rate of 200cm^3/min for 10 minutes. The absorption train was disconnected, each absorber was wiped with a clean dry cloth free from loose fibres, and allowed to cool to the balance room temperature in an enclosure free from dust and each was weighed. The absorption train was reassembled and reconnected to the combustion tube. Purified oxygen was passed through the apparatus at 300cm^3/min for 10 minutes. The absorption train was again disconnected and the oxygen in it was displaced by purified air, then cooled and each unit was weighed as in the earlier instance. When the total change in mass of the absorption tubes was less than 0.5mg the apparatus was certified to be in proper condition for use.

PROCEDURE FOR THE DETERMINATION OF CARBON AND HYDROGEN IN THE COAL ANALYSIS SAMPLE

The temperature of the furnace was raised to 1350°C and the silve gauze roll was ascertained to be in its correct position. The absorption train (Figure 24) was connected to the combustion tube and oxygen passed at a rate of $300cm^3$/min for 10 minutes. The absorption train was disconnected, purged with purified air, wiped, cooled and weighed as described in the blank determination.

About 0.5g of the sample was weighed to the nearest 0.1mg and spread evenly over the bottom of a clean dry boat.

Moisture determination was done for the same coal analysis sample simultaneously.

About 0.5g of aluminium oxide was added to cover the sample in the boat completely. The total moisture in the aluminium oxide was determined by heating 2g to 1350°C in the apparatus and absorbing the water in magnesium perchlorate.

The weighed absorption train was reconnected. The rubber stopper was disconnected. The charged boat was inserted into the combustion tube to a position such that its centre was 240mm from the centre of the hottest zone. With the silica pusher fully withdrawn, the rubber stopper was replaced and oxygen passed at $300cm^3$/min.

At the end of each of the next 6 one-minute intervals the boat was pushed forward 40mm, the silica pusher was withdrawn each time to prevent its distortion. The boat was allowed to remain in the hottest zone for a further 4 minutes. The absorption train was disconnected, purged with purified air, wiped and cooled as before but for 1 hour, and it was weighed as in the blank determination.

Calculation of results

$$\mathrm{Cad} = \frac{27.29M_3}{M_1}$$

and

$$\mathrm{Had} = \left(\frac{11.19M_2}{M_1} - 0.1119\mathrm{Mad}\right)$$

where M_1 is the mass of the sample taken, (g).

M_2 is the increase in mass of the water absorption tube, less the mass of the moisture in the aluminium oxide used (g).

M_3 is the total increase in mass of the soda lime tubes (g).

Cad is the percentage of total carbon in the analysis sample

Had is the percentage of total hydrogen in the analysis sample

Mad is the percentage of moisture in the analysis sample.

4.5.3. DETERMINATION OF TOTAL SULPHUR IN COAL BY THE ESCHKA METHOD

Principle

A known mass of the sample was ignited in intimate contact with Eschka mixture in an oxidizing atmosphere at 800°C to remove the organic material and to convert all the sulphur to sulphate [110]. The sulphate was then extracted and determined gravemetrically by precipitation with barium chloride.

Reagents

All reagents used were of analytical grade quality. Deionised water was used to prepare aqueous solutions. Other reagents used were as follows:

1) Hydrochloric acid, specific gravity 1.18
2) Ammonia solution, specific gravity 0.88
3) Eschka mixture

   A mixture composed of two parts by mass of light magnesium oxide and one part by mass of anhydrous sodium carbonate.

4) Barium chloride solution

   100g of crystalline barium chloride ($BaCl_2 2H_2O$) dissolved in water and diluted to 1 litre.

5) Potassium sulphate solution

   2g of potassium sulphate was weighed accurately after it had been dried at 110°C for 2 hours and then dissolved in water and diluted to 1000 ml in a volumetric flask.

6) Methyl red indicator

0.02g of methyl red was dissolved in 60ml of ethanol (95%) and diluted to 100ml with water.

Apparatus

The following apparatus were used:

1) Muffle furnace

An electrically heated muffle furnace capable of maintaining a zone within the range of 800 ± 25°C. The ventilation was such as gave about five air changes per minute at 800°C.

2) Crucibles

A shallow crucible of glazed porcelain of approximately 25ml capacity was used.

3) Silica plate

A silica plate, 6mm thick, that was an easy sliding fit in the muffle furnace was used.

Procedure

About 1g of the coal analysis sample was weighed to the nearest 0.1mg and mixed intimately with 3g of the Eschka mixture in the crucible. The content of the crucible was levelled and it was covered with a layer of 1g of the Eschka mixture.

The charged crucible was placed in the cold furnace. The temperature of the muffle furnace was raised slowly at first to allow the slow expulsion of volatile matter and finally to 800°C in 1 hour. The furnace was maintained at this temperature for 1 hour. It was then withdrawn and allowed to cool. The charge was then transferred to a 400ml beaker containing 25ml of water. The crucible was thoroughly washed with about 50ml of hot water and the washings were added to the contents of the beaker. The beaker was covered with a clock glass and while stirring, sufficient hydrochloric acid (14ml) was carefully added to dissolve the solid matter, warming the contents of the beaker to effect complete dissolution. It was boiled for 5 minutes to expel carbon dioxide and then filtered into a 400ml beaker, using a retentive filter paper. The filter paper was washed with four 20ml portions of hot water.

To the combined filtrate and washings two drops of the methyl red indicators were added. Ammonia solution was added cautiously until the colour of the indicator changed and a trace of precipitate formed. Just enough hydrochloric acid was added drop by drop to redissolve the precipitate and then 1ml of the acid was added in excess. The solution was diluted to approximately 200ml with water.

The beaker was covered and heated until the solution boiled. The heat was reduced until the ebullition of the solution just ceased. 10ml of cold barium chloride solution was added from a pipette with a delivery time of 20 seconds, held so that the liquid fell into the middle of the hot solution while it was agitated. The contents of the beaker were kept just below the boiling point for 30 minutes.

The precipitate was filtered through an ashless close-textured, double-acid-washed whatman filter paper. It was then washed with water until free from chlorides.

The wet filter paper was folded together with its contents and it was placed in a previously heated and weighed silica capsule. The silica capsule was placed on the silica plate and both were transferred into the muffle furnace at 800°C. It was heated for 15 minutes and then the capsule was removed, cooled on a metal slab and finally in a desiccator and weighed.

Determination of the blank

A blank was determined at the sametime under the same conditions as the actual determination but omitting the sample. 25cm³ of the potassium sulphate solution was however, added to the blank filtrate before precipitating with barium chloride solution.

Calculation of result

$$S = \frac{m_2 - (m_3 - 0.0335m_4)}{m_1} \times 13.74.$$

where m_1 was the mass of the sample taken (g).

m_2 was the mass of barium sulphate found in the determination, (g)

m_3 was the mass of barium sulphate found in the blank determination, (g),

m_4 was the mass of potassium sulphate per litre of solution, (g),

S was the percentage of total sulphur in the analysis sample.

4.5.4. Determination of Sulphate Sulphur in Coal

Principle

A known mass of the sample is extracted with boiling hydrochloric acid for 30 minutes to bring the sulphate sulphur and non-pyritic iron into solution. The filtered extract is made alkaline to precipitate non-pyritic iron, which is then removed by fitration. The sulphate sulphur is precipitated from the filtrate as barium sulphate and determined gravimetrically.

Reagents

All reagents used were of analytical grade and deionised water was used for preparing aqueous reagent solutions. Other reagents used were the following:

1) Hydrochloric acid, specific gravity = 1.18
2) Hydrochloric acid, 5M
3) Hydrogen peroxide, 100 volume
4) Ammonia solution, 5M
5) Barium chloride solution 100 g of crystalline barium chloride dihydrate, $BaCl_2.2H_2$

6) Potassium sulphate solution

   2g of potassium sulphate was dissolved in water and diluted to 1 litre.

7) Methyl red indicator

   0.02g of methyl red was dissolved in 60ml of ethanol (95% v/v) and diluted to 100ml with water.

Apparatus

The following apparatus were required:

1) Condenser

   A cold finger condenser, Arnold bubbler, which fitted loosely into the neck of a 250ml conical flask was used.

2) Carbolite muffle furnace

3) Silica plate

   A silica plate, 6mm thick, that was an easy sliding fit in the muffle furnace.

Procedure

Preparation of solution

About 5g of the coal analysis sample was weighed into a 250ml conical flask. 50ml of the hydrochloric acid was added. Arnold bubbler condenser was inserted through which a slow stream of tap water was passing and the mixture was boiled for 30 minutes.

The condenser was removed and the mixture was filtered. The coal residue was washed six times with the 5N hydrochloric acid using 20ml in all. 2ml of the hydrogen peroxide was added to the filtrate and boiled for 5 minutes.

Procedure Involving Titrimetric Determination of Iron

Titrimetric determination of iron on the coal extract was used. Two drops of methyl red indicator were added to the solution followed by the ammonia solution, drop by drop, until the solution was just alkaline and then five drops were added in excess to ensure precipitation of Iron in the extract. It was filtered into a 400ml beaker through a fast hardened whatman filter paper and the precipitate was washed with three 20ml portions of hot water. The precipitate was reserved for the determination of non-pyritic iron.

Concentrated hydrochloric acid was added, drop by drop, to the filtrate until it just turned acidic and then 1ml was added in excess. $25cm^3$ of potassium sulphate solution was added from a pipette and the solution was diluted to about 200ml with deionised water.

The covered beaker was heated until the solution boiled and then the heat was reduced until boiling just ceased. 10ml of barium chloride solution was added from a pipette with a delivery time of about 20 seconds held so that the liquid fell into the middle of the hot solution while the latter was agitated. The contents of the beaker were kept just below the boiling point for 30 minutes.

The contents of the beaker was filtered through an ashless close textured, double-acid-washed whatman filter paper and the precipitate was washed with water until free from chloride. The filter paper was folded over the precipitate it contained and it was transferred into a previously ignited and weighed silica capsule,stood on the silica plate and both placed in the muffle furnace at 800°c for 15 minutes. The capsul was removed and cooled first on a metal slab and finally in a desiccator. It was the weighed.

Determination of blank

A blank determination was done under the same conditions as the actual determination but omitting the sample.

Calculation of result

$$Ss = \frac{m_2 - m_3}{m_1} \times 13.74$$

where m_1 = mass of the coal taken, (g),

m_2 = mass of the barium sulphate found in the determination, (g),

m_3 = mass of barium sulphate found in the blank determination, (g),

Ss = the percentage of sulphate sulphur in the analysis sample.

4.5.5. DETERMINATION OF PYRITIC SULPHUR IN COAL

Principle

The coal analysis sample to be used for the determination of pyritic sulphur was ground to minus 200 mesh for ease of subsequent acid digestion. A known weight of sample was extracted with boiling nitric acid for 30 minutes to bring both pyritic and non-pyritic iron into solution. The extract was filtered and the amount of iron in the filtrate was determined. The amount of iron in the nitric acid extract of the coal analysis sample less the non-pyritic iron present in the hydrochloric acid extract gave the amount of pyritic iron present in the coal sample. The pyritic sulphur content was calculated from the pyritic iron, assuming it to be present in the form, FeS_2.

Reagents

Analytical grade reagent of chemicals and deionised water were used in all cases. The following reagent solutions were prepared and used.

1) Ammonia solution, 5M
2) Hydrochloric acid, 5M

3) Tin (II) chloride solution

5g of tin (II) chloride dihydrate, $SnCl_2 2H_2O$ were dissolved in 100ml of the 5N hydrochloric acid.

4) Mercury (II) chloride solution, saturated, 70g/l

5) Sulphuric/phosphoric acid mixture. 150ml of concentrated sulphuric acid (S.G. = 1.84) was cautiously added to 500ml of deionised water followed by 150ml of orthophosphoric acid (S.G. = 1.75) and the mixture was made up to 1 litre.

6) Potassium dichromate solution, 0.9g of potassium dichromate was weighed accurately after it had been dried at 150°C for 2 hours and then dissolved in deionised water and made up to 1 litre in a volumetric flast. For this solution 1ml titre $\equiv M_1 \times 0.00114$g of iron, where M_1, was the mass of potassium dichromate taken.

7) Methyl red indicator

0.02g of methyl red was dissolvedin 60ml of ethanol (95% v/v) and diluted to 100ml with water.

8) Sodium diphenylamine sulphonate indicator. 0.2g of sodium diphenylamine sulphonate was dissolved in 100ml of water.

9) Nitric acid, 2M

10) Hydrogen peroxide, 100 volume.

Special Apparatus

The special apparatus used was the condenser.

A 'cold finger' condenser which fitted loosely into the neck of a 250cm^3 conical flask was used.

Preparation of Coal Sample Solution

Coal analysis sample to be used for determination of total acid-soluble iron was ground fine to minus 200 mesh. About 1g of the ground sample was accurately weighed into a 25cm^3 conical flask and 50cm^3 of nitric acid was added. The Arnold bubbler condenser was inserted and the mixture boiled for 30 minutes as a slow stream of tap water flowed through the condenser. The condenser was removed, the mixture filtered and the coal residue washed six times with the nitric acid. the filtrate was coloured brown and so 2cm^3 of the hydrogen peroxide were added and the solution boiled until colourless. The iron in the filtrate was determined titrimetrically.

2.5.5.1. Titrimetric determination of total acid-soluble iron

The filtrate from above sample dissolution step was heated to boiling and two drops of methyl red indicator added. Iron was precipitated by adding ammonia solution, drop by drop, until the solution was just alkaline and the five drops were added in excess. the content of the beaker was filtered through a fast, hardened whatman filter paper and the precipitate was washed with three 20cm^3 portions of hot water.

The filter paper was pierced and the precipitate was washed into a 500cm^3 conical flask with a fine jet of hot water. 10cm^3 of the hot hydrochloric acid was poured on the filter paper in small portions to remove the last traces of iron (III) hydroxide and finally washed with a small quantity of hot water. The total volume of solution did not exceed 200cm^3.

The contents of the flask was heated to boiling and swirled as tin (II) chloride solution was added, drop by drop, until the yellow colour disappeared. Five drops were added in excess and the solution was quickly cooled to room temperature and 10cm^3 of mercury (II) chloride solution added rapidly. A silky precipitate of mercury (II) chloride was formed.

15cm^3 of the sulphuric (I) phosphoric acid mixture were added. The solution was diluted to 200ml. Five drops of the sodium diphenylamine sulphonate indicator were added and the solution was titrated with the potassium dichromate solution. Near the end-point the colour of the solution deepened to grenish-blue. When that stage was reached the potassium dichromate solution was added, drop by drop, until the colour changed to intense violet-blue. In some cases when the end-point colour of the solution tended to vary, it was understood that the titration was not complete and then addition of one or two more drops of the potassium dichromate solution made the colour-change permanent confirming completion of chemical reaction.

4.5.5.2. Determination of non-pyritic iron in coal

Principle

Non-pyritic iron was determined by the same method as that used for the total acid-soluble iron in coal. Titrimetric method of determining non-pyritic iron was employed.

The precipitate of iron (II) hydroxide obtained during the determination of the sulphate sulphur was determined by the method described for total acid-soluble iron.

Calculation of Result

$$Sp = \left(\frac{V_1}{M_2} - \frac{V_2}{M_3}\right) \times 0.1308M_1$$

where M_1 = mass of potassium dichromate per litre of solution, (g),

M_2 = mass of the coal taken, (g), for total acid-soluble iron,

M_3 = mass of the coal takne, (g), for the non-pyritic iron

V_1 = Volume (cm^3) of the potassium dichromate solution used in the determination of toal acid-soluble iron

V_2 = Volume (cm^3) of the potassium dichromate solution used in the determination of non-pyritic iron.

Sp = percentage of pyritic sulphur in the analysis sample.

4.5.6. Determination of Organic Sulphur in Coal

The organic sulphur content of coal was obtained by calculation and it was the difference between the total sulphur, determined by the Eschka method, and the sum of the sulphate sulphur and the pyritic sulphur.

$$So = s - (Ss + Sp)$$

where

S = percentage of total sulphur in analysis sample

So = percentage of organic sulphur in analysis sample

Ss = percentage of sulphate sulphur in the analysis sample

Sp = percentage of pyritic sulphur in the analysis sample.

Repeatability (% absolute) for pyritic sulphur less than 0.5% was 0.05 and for values of 0.5% and above repeatability was 0.07. For sulphate sulphur repeatability (% absolute) was 0.02.

4.5.7. Determination of Nitrogen in Coal

Principle

The Kjedahl method of nitrogen determination was used.

A known mass of the sample was heated with concentrated sulphuric acid in the presence of a catalyst to destroy the organic material and to convert the nitrogen to ammonium sulphate. Ammonia was released by steam distillation from an alkaline solution, absorbed in boric acid solution and titrated directly with 0.01M sulphuric acid.

Reagents

All reagents used were of analytical grade. Deinoised water was used in preparing all aqueous solutions. Other reagents included:

1) sulphuric acid, specific gravity = 1.84
and it was nitrogen free.

2) sulphuric acid 0.01M

3) mixed catalyst which contained by mass:
32 parts of potassium sulphate,
1 part of selenium powder, and
5 parts of mercuric sulphate.

The mixture was thoroughly ground in a mortar before use.

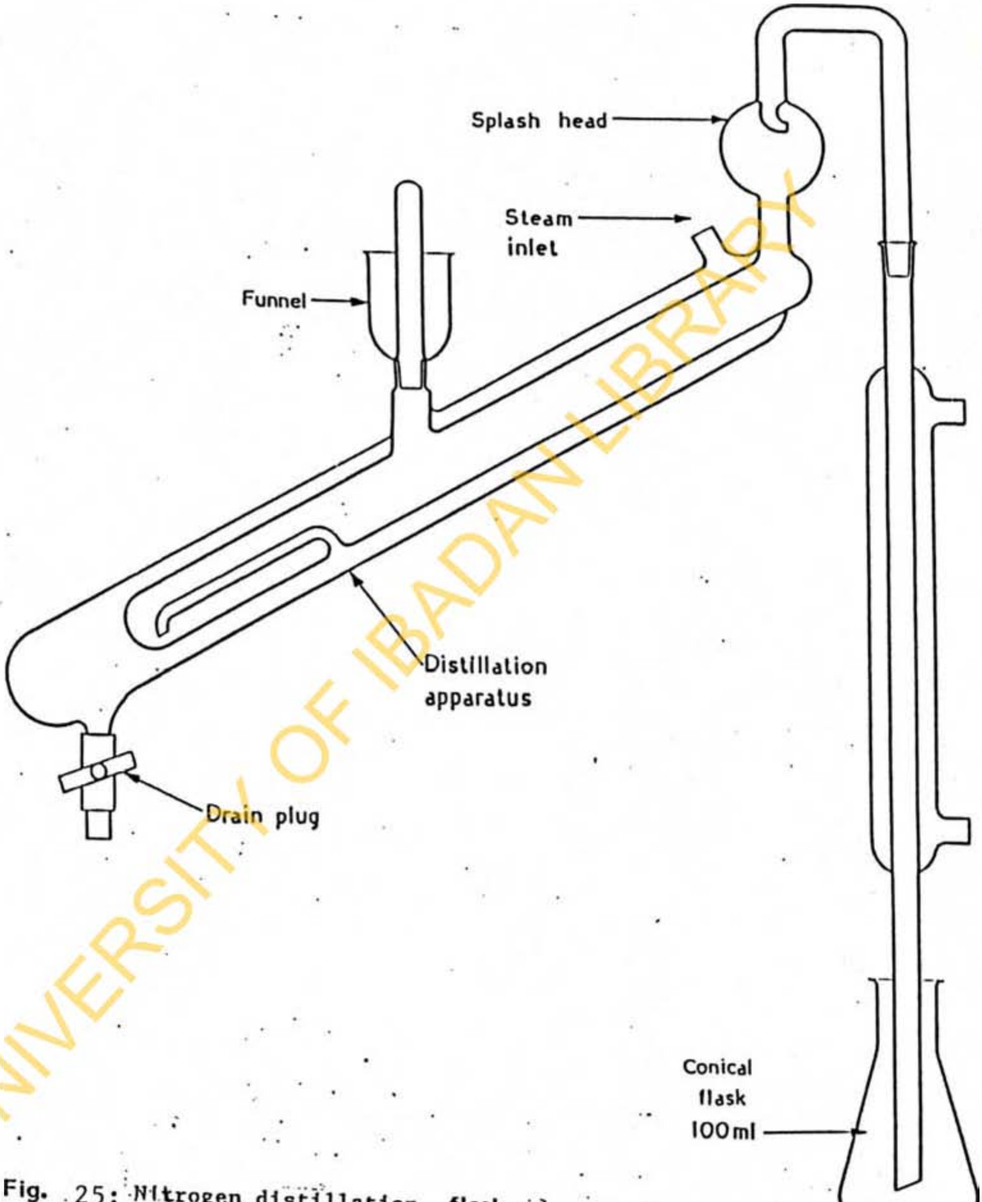


Fig. 25: Nitrogen distillation flask

*A unit using 100 W electric heating mantles to heat a number of flasks simultaneously is obtainable commercially

4) Boric acid solution, saturated

60g of boric acid was dissolved in 1 litre of hot water. It was allowed to stand for 3 days after which it was then decanted and the clear solution used.

5) Alkaline sodium sulphide solution

It was used with the catalyst mixture in (3) above. 400g of sodium hydroxide plus 40g of sodium sulphide ($Na_2S.9H_2O$) per litre.

6) Screened indicator.

The indicator was prepared by mxiing equal volumes immediately before use. The solutions A prepared by dissolving 0.125g of methyl red in 100ml of ethanol (95% v/v) and B which contained 0.083g of methylene blue in 100ml of ethanol (95% V/V) and stored in a dark glass bottle.

7) Sucrose

Apparatus

The apparatus consisted of:

1) Digestion flask

A Kjeldahl flask of borosoliticate glass, which was round-bottom shaped and of 50ml capacity;

2) Stopper

The stopper was loosely fitting, blown glass used for closing the mouth of the flask.

3) Heating Unit

The flask was heated using electrical heater.

4) Distillation apparatus

A suitable distillation apparatus used was of borosilicate glass, fitted with a splash head to arrest the passage of any entrained sodium hydroxide solution, and upright condenser and a 100cm^3 conical flask, as shown in Figure 25.

5) Burette

A 25cm^3 glass burette was used.

Procedure

About 0.01g of the coal analysis sample was weighed to the nearest 0.1mg on a scoop. It was transferred completely to the digestion flask and 2g of the catalyst mixture was added and shaken to mix. 4cm^3 of the concentrated sulphuric acid was added and the reactants were mixed well.

The flask was placed on the electrical heating unit with the heat input adjusted so that the solution became clear in 10 minutes. After the solution became clear, boiling was continued to give a total digestion time of 25 minutes in order to complete the conversion of the nitrogenous compounds into ammonium hydrogen sulphate.

During the digestion, the distillation apparatus was steamed out for 30 minutes and then the connection between the steam generator and the distillation flask was closed and any waste from the previous distillation was blown out. 20cm^3 of the alkaline sodium sulphide solution was poured into the distillation flask through the funnel and the latter was rinsed with water. 2cm^3 of the boric acid solution was measured into the receiver and four drops of the mixed indicator solution were added. the height of the receiver was adjusted so that the end of the condenser tube dips below the surface of the liquid.

The acid digest was allowed to cool for 5 minutes and it was then diluted by adding 10cm^3 of deionised water, at first in drops with shaking, until the reaction was less violent.

The diluted digest was poured into the distillation flask through the funnel. The digestion flask was rinsed into the funnel with water and the tap funnel was itself rinsed with water. The total volume of liquid in the distillation flask did not exceed half the capacity of the flask to forestall frothing and priming.

Steam was passed into the distillation apparatus and it was distilled for 5 minutes from the time the steam entered the condenser, at a rate of about 4cm^3 of distilate per minute.

To ensure complete absorption of the ammonia the contents of the receiver was kept cold. The reciever was lowered so that the condenser end was about 25mm above the surface of the liquid and distillation continued for 1 minute longer. The end of the condenser tube was rinsed with water.

The ammonia present in the distillate was titrated with the sulphuric acid until the grass-green colour changed to steel-grey, a further drop then giving the purple (acid) colour.

Determination of the blank

A blank determination was done at the same time and under the same conditions as the actual determination, but using 0.1g of sucrose instead of coal.

Calculation of Result

$$N_2 = \frac{(V_1 - V_2) \times 0.014}{M}$$

where M = the mass of sample taken, (g),

V_1 = the volume of 0.01M sulphuric acid in the sample determination (cm^3),

V_2 = the volume of 0.01M sulphuric acid used in the blank determination, (cm^3),

N_2 = the percentage of nitrogen in the analysis sample.

4.5.8. DETERMINATION OF THE CRUCIBLE SWELLING NUMBER

Principle

A standard mass of coal was heated in a covered crucible under standard conditions to a final temperature of 820 ± 5°C. The coke button obtained was classified by comparison with the outlines of a set of standard profiles. The number of the profile most closely corresponding to the coke button obtained was the crucible swelling number.

Procedure

The sample was prepared, weighed and heated in an electric furnace to obtain the coke button.

Preparation of sample

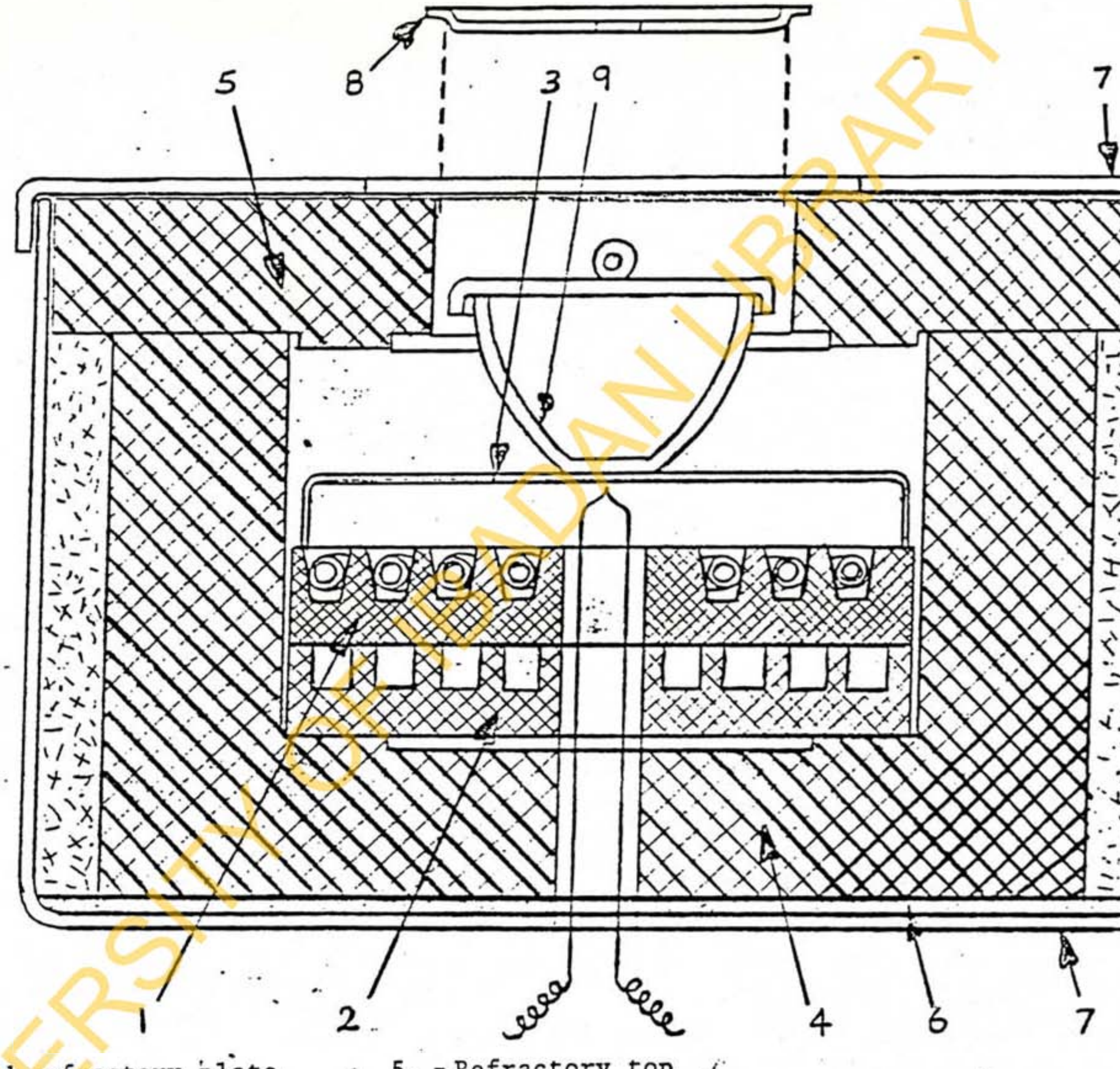
The coal used for the determination of the swelling number was the analysis sample ground to pass minus 72 mesh. Precaution was taken to avoid very fine grinding and undue exposure of the ground coal to the atmosphere since they were liable to lead to incorrect results. The coals were prepared and ground not more than 2 hours before the swelling was done.

Apparatus

The apparatus used were as follows:

1) Furnace

The electrical furnace used is illustrated in Figure 26. The furnace consisted of a grooved refractory plate, approximately 88mm in diameter and 13mm thick, carrying the heating element, which was a spiralled metallic coil. The plate was supported by a second grooved plate. An inverted silica dish of 1mm wall-thickness, 10mm high and approximately 85mm external diameter, was placed over the windings and acts as a support for the crucible. The plates were surrounded by a refractory cylinder of approximately 140mm diameter, bored to a depth of about 60mm with a 90mm diameter hole. The cylinder was fitted with a refractory top, 50mm thick; a hole 50mm diameter was bored in the centre of the refractory top to allow for insertion of the crucible. A refractory lid was used to conserve the heat losses from the furnace between determinations. The refractory cylinder rested on a refractory sheet and was surrounded by a 10mm thick insulation of alumina - silica wool. The whole furnace was placed in an aluminium case.



1. Grooved refractory plate
2. Second grooved plate
3. Inverted silica dish
4. Refractory cylinder
5. Refractory top
6. Alumina-silica wool insulation
7. Aluminium case
8. Refractory lid
9. Silica crucible

Fig. 26:Electrically heated furnace

A hole bored through the base of the furnace enabled a thermocouple to be placed in contact with the underside of the silica dish. The furnace was equipped with a suitable energy regulator and an ammeter.

2) Crucible and lid

Transluscent silica crucible, with a flat base, squat form and silica lid with a ring handle and flat lower surface were used. The dimensions of the crucible were: external height 26± 0/5mm, external diameter at top 41± 0/75mm, internal diameter at base 11mm, mass 11,5g and capacity $16.5cm^3$.

3) Pierced silica lid

The lid had a 6mm hole to accommodate the thermocouple

4) Thermocouples

The thermocouple was of fine wire of diameter not greater than 0.23mm made of platinum and with ends in the form of flattened loop.

5) Weight

500g weight was used

Calibration of Apparatus

The furnace was switched on and the energy input adjusted so that a steady temperature of about 850°C was maintained at the base of a crucible resting on the silica dish. The crucible was removed and a cold crucible was inserted covered with a pierced lid through which passed the fine wire thermocouple so that its unprotected junction and a portion of each wire rested on the base of the crucible. It was ascertained that the standard heating conditions of 800± 10°C in 1.5minutes and 820 ± 5°C in 2.5 minutes were attached from the time of inserting the crucible. The temperature of the furnace as indicated on the thermocouple at the underside of the silica dish was recorded.

Determination of Crucible Swelling Number on Coal

1.00g of the freshly ground coal analysis sample was weighed into a dry crucible and the crucible with its content was lightly tapped about 12 times on the bench to level the surface of the coal. The crucible was covered with the unpierced lid and then placed centrally in the furnace on the silica dish. It was heated until the volatile matter ceased to be evolved which took about 2.5minutes. The crucible was removed from the furnace and allowed to cool.

The residue was carefully removed from the crucible. The residue that was non-coherent, i.e. mostly loose powder, was not examined any further and such was designated swelling number 0.

The residue which was a coherent coke button but was not swollen, was placed on a flat surface and 500g weight was rested on it. Such residue which supported the 500g weight without breaking into more than two or three hard pieces was designated swelling number 1; while those that crumbled or disintegrated were designated swelling number ½.

The coke button that swelled were compared with the standard numbered profiles shown in Figure 27. Its largest profile was presented for comparison. The residue was matched to the most similar outline of the swelling numbers given in Figure 27. Five tests were done in succession. In between tests the refractory lid of the furnace was put in place to minimize heat losses through the top of the furnace. After each test the carbon residue was burnt off and the crucible internally wiped with a clean cloth.

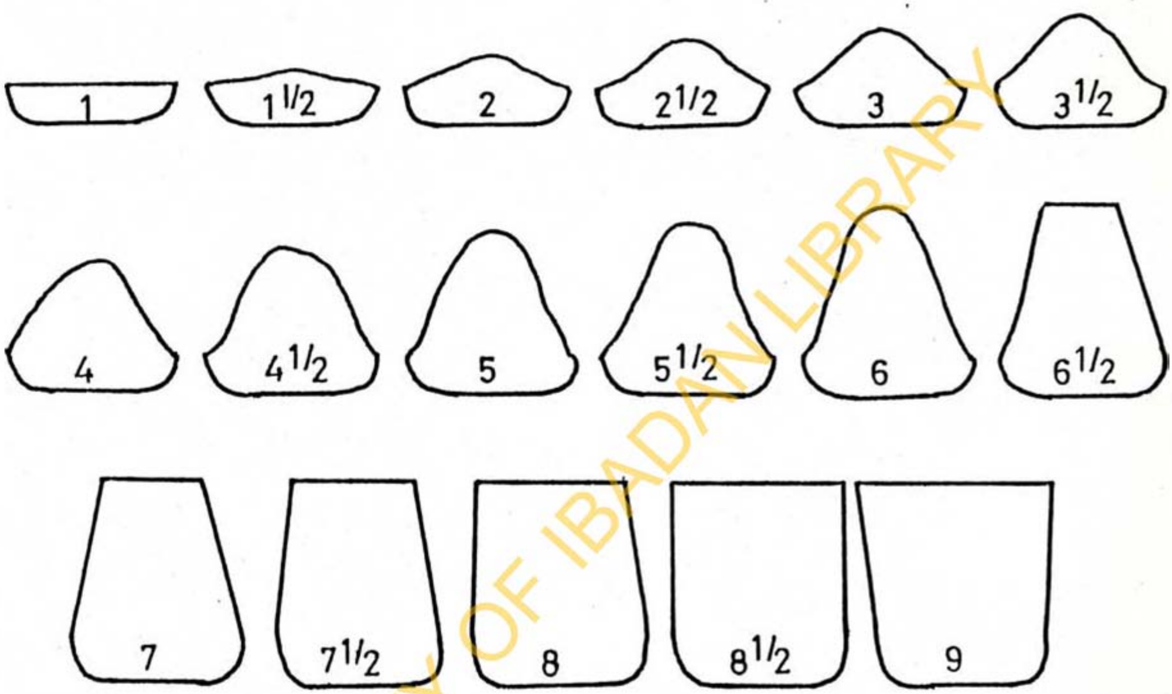


Fig. 27. Determination of crucible swelling number: Standard profiles and corresponding swelling numbers.

Reporting Results

The results were reported as the mean of the five tests to the nearest ½ unit as the crucible swelling number of the coal sample. The swelling number was recorded as follows:

Swelling number 0: non-coherent residue

Swelling number ½: non-swollen coke button that disintegrated under 500g weight.

Swelling number 1: non-swollen coke button that supported the 500g weight without breaking into more than two or three hard coherent pieces.

Swelling number 1 ½ to 9: Number of the profile in Figure 21 that a swollen coke button most closely matched.

Accuracy: Repeatability was limited to ½ unit.

4.5.9. DETERMINATION OF GRAY-KING COKE TYPE

Introduction

The Gray-King coke type is one of the parameters used to clarify hard coal by type in Great Britain and by the United Nations Economic Commission for Europe. It is a way of assessing the caking properties of a type of coal or a blend of coals by carbonizing under standard conditions.

Principle

The coal is heated under standard conditions to a final temperature of 600°C. The coke residue obtained is classified by reference to a series of standard residues. The coke produced is so swollen that it filles the cross-section of the retort tube. The determination is repeated with the coal admixed with a suitable quantity of electrode carbon. For such highly swelling coals the Gray-King coke type is defined by the minimum amount of electrode carbon required to produce a strong hard coke residue of the same volume as the original coal and electrode carbon mixture.

Reagent

The reagent was high temperature electrode carbon with the following characteristics:

Moisture less than	1%
Volatile matter less than	1.5%
Ash less than	5%
Bulk density at 25°C	1.00g/cm^3 to 1.05g/cm^3
Relative density at 25°C	2.05 to 2.09

Size analysis of the electrode carbon shall be:

Minus 212 m plus 125 m	less than 26%
Minus 125 m plus 63 m	10 % to 40%
Minus 63 m	50 % to 85%

Apparatus

A horizontal electric furnace that has 50mm internal diameter and 300mm long, with one end closed and the other carrying a plug of insulating material which is bored centrally with a hole 25mm diameter. The winding of the furnace was such that the middle 200mm was at a uniform temperature within ±5°C at both 300°C and 600°C.

The furnace was insulated and placed in a metal box and was equipped with a suitable thermocouple lying above the retort tube, when the latter was in position and with the junction at the centre of the furnace. An indicator for showing the furnace temperature with an accuracy of ±5°C was provided. A suitable controller of the automatic programmed type was provided which permitted an increase in temperature at a rate of 5°C/minute.

Retort Tube

A heat-resistant glass tube, 20mm internal diameter and 100mm long, closed at one end, with a side arm, 8mm internal diameter and 50mm long, sealed in at a distance of about 20mm from the open end. the tube had a slight taper (19mm to 21mm) such that the open end was larger.

Distance Rod

This was a rod with a flat disc at one end that assisted in the packing of the coal and indicated the free end of the coal sample in the retort tube.

Receiver and Outlet Tube

A U-tube was used which was immersed in water, suitably supported and attached to the side arm of the retort tube, fitted with an outlet tube leading to the atmosphere of the end of which the gas leaving the receiver was burnt.

Procedure for Gray-King Coke Type Determination

The coal used for the determination of the Gray-King coke type was the analysis sample ground 100 percent to minus 212μm. It was thoroughly mixed. The sample was prepared the same day that the determination was carried out.

For Coals with a Gray-King Coke Type Within the Range A to G_2

The temperature of the furnace was raised until it was steady at 325°C. 20g to 20.01g of the coal sample was weighed on a scoop and transferred to the retort tube, held in such a manner that the coal could not enter the side arm. The coal was completely transferred to the far end of the retort tube with a soft brush. The tube was held horizontally and the distance rod inserted so that the face of the disc was 150mm from the closed end of the retort tube.

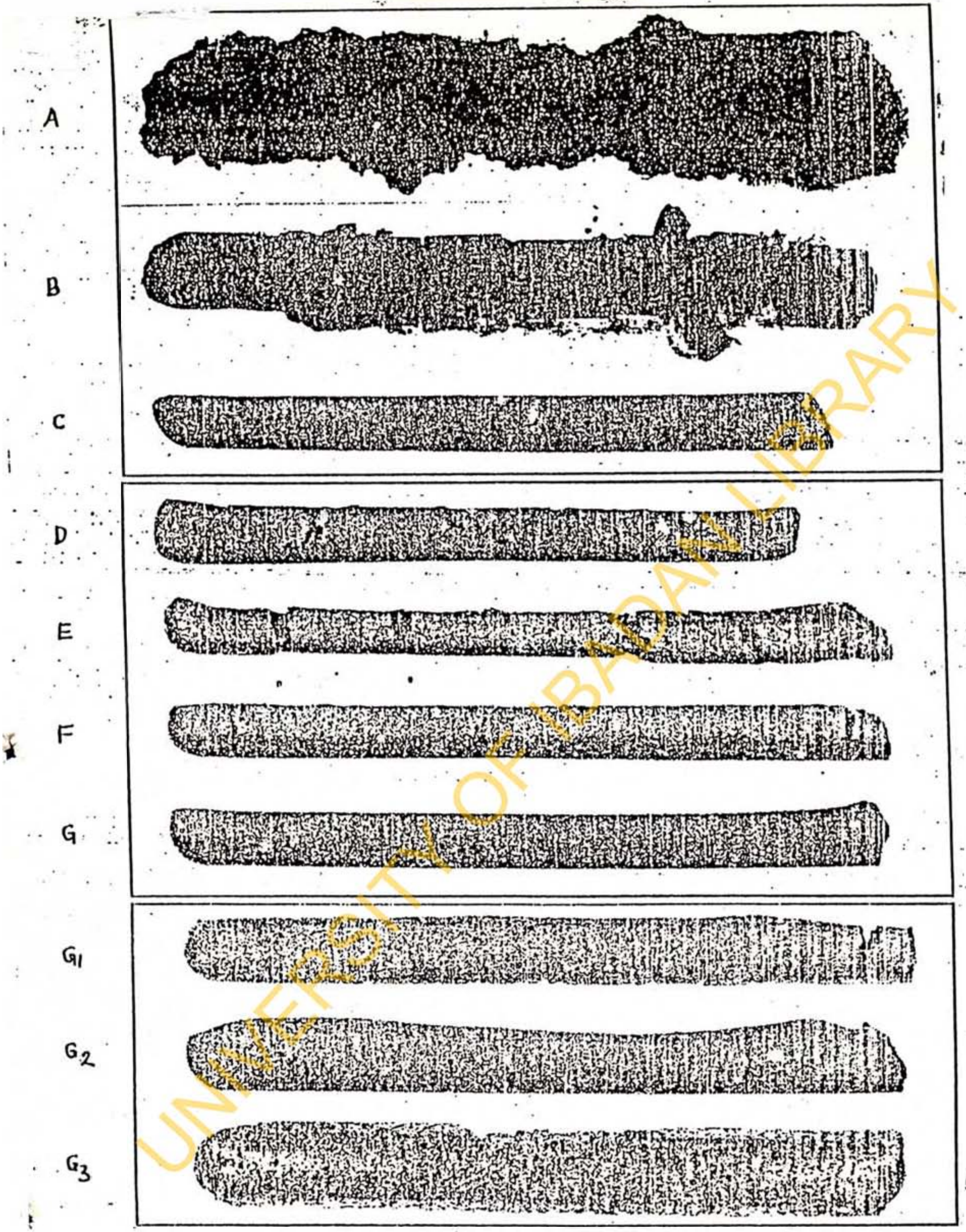


Figure 28: Gray-King Coke Type

KEY

A	Pulverulent
B	Just coherent, breaks into powder on handling.
C	Coherent, but friable on rubbing.
D	Shrunken, moderately hard.
E	Shrunken and fissured, hard
F	Slightly shrunken and hard
G	Hard, occupying same volume as original coal (="standard coke")
G_1	Slightly swollen, hard
G_2	Moderately swollen, hard.
G_3	Highly swollen, fills tube without increase

The coal was spread into a layer of uniform depth by shaking, turning and tapping gnetly. The distance rod was withdrawn and a flattened pad of alumina silica wood was inserted to retain the coal in position. Without disturbing the position of the coal, the open end of the retort tube was closed with a heat resistant stopper. The receiver was connected to the side arm and the retort tube was inserted in position in the furnace so that the centre of the coal layer coincides with the centre of the furnace. The energy input to the furnace was raised in such a manner that the temperature of 325°C was regained in 3 minutes to 7 minutes. A uniform rate of 5±0.1°C per minute temperature rise was maintained with a variation of not more than ±1°C per 50°C rise in any 10 minutes period, thereafter until a temperature of 590°C was reached. At this point, the energy input was regulated to the furnace so that a temperature of 600°C was reached. This temperature was maintained for 15 minutes.

The retort tube was withdrawn and allowed to cool. The receiver was detached, the stopper removed and the coke residue was slided out for examination.

For Coals with a Gray-King type greater than G_2

Xg of electrode carbon was weighed on a scoop. X was an interger. (20-X)G of the coal sample was also weighed. Both the coal and the electrode carbon were transferred into a weighing bottle. The contents were thoroughly mixed. The mixture was transferred to the retort tube and the procedure described above repeated. The amount of electrode carbon was varied in the 20g mixture until a coke residue of type G was obtained using the minimum mass of electrode carbon.

Results of Gray-King coke type Determination

The Gray-King coke types of Lafia, Enugu and Okaba coals were interpreted by referring to the coke residues and to Figure 29. For Lafia coal that gave a coke type with an index greater than G_2, the subscript was the minimum number of grams of electrode carbon added to produce a standard G type coke residue.

Lafia coal seam 12 Gray-King coke type

- Untreated G_3
- Sp.Gr. 1.4 coal concentrate G_8

Enugu coal Gray-King coke type - A

Okaba coal Gray-King coke type - B

4.6. DISCUSSION OF RESULTS OF PROXIMATE ANALYSES OF LAFIA COAL

The mean results of triplicate determination of proximate analyses parameters (moistre, ash, volatile matter, total sulphur) along with crucible swelling number are given in Table 34. Table 35 gives sulphur speciation analysis, specific gravity, moisture, ash and volatime matter content of Lafia Coal seam 12. Normally, coal samples having ash greather than 35% are not analysed for volatile matter content, specific gravity and crucible swelling number. Table 41 gives a measure of precision achieved in the determination of ash content of some reference coal samples (types International Code Numbers 333, 434, and 534).

From the results in Table 35, it is noteworthy that pyritic sulphur is the form of sulphur that predominates in Lafia coal with a concentration range of 1.01 to 40% followed by organic sulphur with concentration range 0.79 to 1.00%. The sulphate sulphur form is present at much lower concentration (0.01% to 0.07%) which is the usual amount common to un-oxidised bituminous coals. Total sulphur values range from 1.91% to 5.6% and on occasions higher values have been encountered. This is to say that of the three main sulphur forms pyritic sulphur constitutes from 52.9% to 84.4% of total sulphur in Lafia Coal.

TABLE 34:

PROXIMATE ANALYSIS OF LAFIA COAL CORE SAMPLES FROM SEAM 12

Serial Number	Sample Number	Sample Depth (m)	Moisture (% wt) (a.d.)	Ash (% wt) (a.d.)	Volatile matter (% wt) (d.a.f)	Crucible swelling Number	Sulphur (Total) (% wt)
1	2472	35.10	3.47	64.06	-	-	4.80
2	2473	35.30	2.30	31.42	28.81	5	3.20
3	2474	37.00	1.47	21.10	28.28	6	2.80
4	2475	37.03	4.48	71.89	-	-	2.81
5	2476	37.35	2.26	35.42	29.77	5	1.76
6	2479	37.95	1.99	19.93	27.44	6	3.53
7	2507	36.10	1.02	38.06	31.94	5	-
8	2477	37.60	6.84	81.60	-	-	-
9	2476	37.65	4.15	51.40	-	-	-
10	2505	35.80	1.42	38.04	-	-	2.12
11	2451	43.05	1.68	29.40	28.50	5	3.09
12	2452	43.85	2.13	45.20	-	-	-
13	2453	44.20	2.20	44.05	-	-	-
14	2454	44.40	4.74	74.17	-	-	3.72
15	2455	44.60	2.04	29.57	29.47	6	2.86
16	2456	44.75	5.29	78.24	-	-	2.19
17	2457	44.95	2.20	43.20	32.72	6	0 96
18	2459	46.30	4.46	77.68	-	-	4.90

TABLE 34 (contd.)

S/NO	Sample Number	Sample Depth (m)	Moisture (% wt) (a.d.)	Ash (% wt) (a.d.)	Volatile matter (% wt) (d.a.f)	Crucible Swelling Number	Sulphur (Total) (% wt)
19	2460	46.35	1.82	33.87	29.77	5	-
20	2461	46.50	5.16	82.43	-	-	0.90
21	2462	47.10	1.34	24.74	-	-	3.53
22	2463	47.35	2.23	38.71	-	-	4.62
23	2482	359.00	3.89	92.50	-	-	1.17
24	2483	359.20	0.75	52.70	-	-	-
25	2484	359.25	1.66	57.76	-	-	2.73
26	2485	359.50	2.30	58.35	-	-	-
27	2486	359.60	2.83	72.71	-	-	5.40
28	2487	360.35	3.91	87.52	-	-	3.12
29	2488	360.70	0.93	42.10	-	-	3.04
30	2490	361.50	1.92	31.22	30.38	5	1.85
31	2491	361.90	1.80	29.76	29.94	6	0.93
32	2492	362.00	4.51	85.02	-	-	-
33	2493	362.05	3.08	70.07	-	-	-
34	2495	358.80	1.44	31.38	27.95	6	-

Legend

1. Sample Numbers 1-9 were core samples from borehole No.97
2. Sample Number 10 was a core sample from borehole No.99
3. Sample Numbers 11-22 were core samples from borehole No.96
4. Sample Numbers 23-34 were core samples from borehole No.93
5. dash (-) means not determined.

TABLE 35:

PROXIMATE ANALYSIS OF LAFIA COAL SEAM 12 SHOWING SULPHUR TYPES

Serial Number	Thickness (m)	Specific gravity	Moisture % wt	Ash (d.b) % wt	Volatile matter (% wt)	Total sulphur stot (% wt)	Pyritic sulphur syr. (% wt)	Sulphate sulphur (% wt)	Organic sulphur sorg. (% wt)	Spyr/Stot
1	0.40	1,591	1.96	32.2	28.1	3.19	2.21	0.07	0.91	69.3%
2	0.32	1,493	1.23	25.7	28.5	2.66	1.82	0.05	0.79	68.4%
3.	1.07	1,915	3.59	57.1	-	1.98	1.57	0.02	0.39	79.3%
4.	0.28	1,479	1.00	24.1	28.8	2.43	1.43	0.07	0.93	58.8%
5.	0.07	1,697	1.30	37.1	28.1	5.06	4.04	0.05	0.97	79.8%
6.	0.32	1,416	0.80	19.4	28.4	2.36	1.35	0.01	1.00	57.2%
7.	0.44	1,640	1.40	35.7	29.6	5.56	4.69	0.06	0.84	84.4%
8.	0.12	1,618	1.80	36.4	28.2	1.91	1.01	0.03	0.87	52.9%

TABLE 36: PROXIMATE AND ULTIMATE ANALYSIS OF ENUGU AND OKABA COALS

Sample No.	PROXIMATE		ANALYSIS		ULTIMATE ANALYSIS					Calorific value Btu/Ib
	Moisture	Ash	Volatile matter	Fixed carbon	C	H	N	S	O	
ENUGU COAL										
1A	10.1	7.1	38.3	44.5	68.1	4.4	1.8	0.4	8.1	11,340
1B	-	-	46.3	53.7	82.2	5.3	2.2	0.5	9.8	13,700
2A	6.8	6.7	41.8	44.7	67.8	5.0	1.9	1.5	10.3	11,980
2B	-	-	48.3	51.7	78.4	5.8	2.2	1.7	11.9	13,850
3A	8.1	10.9	38.5	42.5	63.1	4.5	1.8	0.5	11.1	11,370
3B	-	-	47.5	52.5	77.9	5.6	2.2	0.6	13.7	14,040
4A	7.4	6.5	41.7	44.4	65.1	4.7	1.8	1.0	13.5	12,070
4B	-	-	48.4	51.6	75.6	5.5	2.1	1.2	15.6	14,020
5A	6.0	11.2	40.2	42.6	65.3	4.6	1.9	0.6	10.4	11,670
5B	-	-	48.6	51.4	78.9	5.6	2.3	0.7	12.5	14,090
6A	5.9	6.5	41.3	46.3	69.4	4.7	2.0	0.8	10.7	12,200
6B	-	-	47.1	52.9	79.2	5.4	2.3	0.9	12.2	13,980
7A	11.1	9.1	38.3	41.5	64.1	4.8	1.6	0.9	8.4	11,500
7B	-	-	48.0	52.0	80.5	6.1	2.0	1.0	10.4	14,400
8A	8.3	7.9	41.0	42.8	67.3	5.2	1.5	0.5	9.3	12,150
8B	-	-	48.9	51.1	80.4	6.2	1.8	0.6	11.0	14,500
9A	10.5	3.9	39.9	45.7	68.3	5.1	1.7	0.5	10.0	12,200
9B	-	-	46.6	53.4	79.8	6.0	2.0	0.6	11.6	14,450
10A	6.3	15.8	36.0	41.9	61.5	4.5	1.3	0.8	9.8	11,100
10B	-	-	47.3	52.7	80.7	5.9	1.7	-	11.7	14,600
OKABA COAL										
1A	12.5	10.8	38.9	37.8	55.6	3.9	1.5	0.7	15.0	9,860
1B	-	-	50.7	49.3	72.5	5.1	1.9	0.9	13.6	12,860
2A	8.1	7.8	42.7	41.4	60.4	3.7	1.6	0.6	17.8	10,700
2B	-	-	50.8	49.2	71.8	4.4	1.9	0.7	21.2	12,720

N.B.

A - air dried basis

B - dry ash free basis

TABLE 37: PROXIMATE COMPOSITION OF SOME NIGERIAN COALS COMPARED WITH COALS FROM OTHER COUNTRIES

Origin of the coals:			Ash	Volatile compounds (%)	Sulphur^{5} (%)
Norway		Spetsbergen	7	39	0.78
England			9	30	1.12
USA	A	Massey	-4)	35^{3})	1.02
USA	B	Massey	-^{4})	32^{3})	0.61
West Germany	HA	Ruhr	5^{1})	28^{2})	0.85
West Germany	W		11^{1})	35^{2})	0.79
Poland	A		-^{4})	-^{4})	0.60
Poland	B		-^{4})	-^{4})	0.76
Soviet Union	A	Petchora	15-17^{1})	34-35^{2})	0.70
Soviet Union	B	Petchora	15	36^{2}	0.50
Soviet Union	C	Kuznetsk	12	28^{2})	0.40
Australia		50% Lithgow 50% Katoomba (New South Wales	15^{3})	27^{3})	0.47
Nigeria^{6}		Lafia	19.4-57.1	28.1-28.8	1.91-5.06
		Enugu	3.9-15.8	36.0-41.8	0.40-1.20
		Okaba	7.8-10.8	38.9-42.7	0.60-0.70

1) On dry coal basis
2) On dry ash free basis
3) "as received"
4) No data available
5) On dry coal basis
6) Range of results obtained

The high (0.79 to 1.00)organic sulphur (usual level of organic sulphur in coal is 0.5%) in Lafia coal places a limitation on the level of sulphur removal that could be achieved using physical coal cleaning facilities. This is because the organic form sulphur is a chemical constituent of the coal-forming organic compounds. As such, it is only rigorous oxidative or reductive chemical treatment that could dislodge organic sulphur variety from the coal substance.

The normal level of sulphate sulphur in coal is 0.1% hence the understanding that Lafia coal seam 12 is not significantly oxidized. As the sulphate form of sulphur is soluble in water, it easily dissovles in aqueous coal washery medium.

Again, Table 34 contains the character of Lafia coal as viewed through bore-holes Nos. 99, 97, 96 and 93. In addition to the measures of moisture, ash, volatile matter and sulphur contents which are within the same range as in Table 35, the Table gives the crucible swelling number of some of the coal samples. Values of the crucible swelling number obtained have a range between 5 and 6, which compliments the values of volatile matter content of 28.81, 29.77% and indicates a coal with coking potentials.

This is explained by the fact that, typically, coals with medium volatile matter content (between 28-32) exhibit cokeability as visible from the crucible swelling number which in this sample was between 5 and 6.

The volatile matter content of seam 12 coal is 28.1-29.6% and has mean value of 28.5% on dry basis. It is probably sufficient within the limits of available data to say that Lafia Coal appears to have medium volatile matter content.

Since the ash content of Lafia coal is high (19.4-57.1%) before the coal could be classified according to the International Classification Code for hard coals by type, it is required to de-ash it below 10% ash and then re-measure its volatile matter. The volatile matter would then be calculated on the dry ash free (daf) basis. Then, using the parameters of volatile matter and caking (e.g. Gray King coke type) properties International Classification number could be assigned.

The moisture content of Lafia coal samples are moderate (1 to 4%) and therefore typical of bituminous coals. Also, the specific gravity of Lafia coal varies between values of 1.416 and 1.915 which seem to point at the occurrence of inorganic impurities in the coal.

Coal organic matter has a specific gravity of between 1.2 and 1.8 but since coal has an admixture of organic and inorganic matter and latter raises the overall specific gravity of run-of-mine coal. Such inorganic mineral matter are aluminosilicates (Sp.Gr.2.6), sulphides (S.G. 4.9), carbonates (S.G. 2.9) and sulphates (S.G. 2.4).

Table 36 however gives the results of proximate and ultimate analysis of Enugu and Okaba coals, which are the other two major deposits of coal in Nigeria apart from Lafia coal. Enugu coal, based on air dried basis (a.d.b.) contains 6.3-10.1% moisture, 3.9-15.8% ash, 36.0-41.7% volatile matter content and total sulphur 0.4-1.5%. Okaba coal also contains 8.1-12% moisture, 7.9-10% ash, 38.9-42.7% volatile matter and total sulphur about 0.7%.

Table 37 gives the proximate composition of coal from other countries compared to Nigeria.

In comparison with Lafia coal which is high in sulphur (total sulphur 1.9-5.1%), Enugu and Okaba coals have low ash and low total sulphur values (Tables 34, 35, 36 and 37) which are comparable with coal from Norway, West Germany and Soviet Union.

However, Enugu and Okaba coals are not suitable for metallurgical coke preparation because they are non-coking. Hence the choice of Lafia coal, which though has coking properties, still has high sulphur content which is desirable for metallurgical use. The high sulphur content must nonetheless be reduced by suitable methods of desulphurization in order to make the coal to be of immense industrial and socio-economic benefit to the steel industry in Nigeria.

4.7. ANALYSIS OF NIGERIAN COAL-ASH

Introduction

Coal ash is an important component of the chemical constitution of coke. Coal ash in coke gives strength to the coke. Some metallic elements in coal ash catalyse the combustibility of coke and are thus significant in the thermo-chemical reaction of coke at the furnace high temperatures. Some elements like ferrous iron in coal ash lower the melting point of the slag while some other will dissolve in the hot molten crude steel. For these reasons it is of metallurgical interest to accurately determine the physico-chemical properties of coal ash.

Principle

The coal-ash is fused with lithium metaborate and dissolved in dilute hydrochloric acid. A standard ash and a blank solution were made up and the elements determined on the Atomic Absorption Spectrophotometer using standard ash of calibration.

Reagents

1. Lithium metaborate fusion mixture:
   150 grams of A-R lithium carbonate
   250 grams of A-R boric acid.

The two were mixed and heated for 4 hours at 400°C, cooled and crushed to pass through a 60 mesh sieve.

2. Lanthanum releasing agent (=10% La)
25 grams of lanthanum chloride
$LaCl_3.7H_2O$ dissolved in 100ml of water.

Dissolution

0.1g of the sample (dry and organic-matter-free) is mixed with 0.6g of the fusion mixture in a platinum crucible. The mixture is fused to a clear melt over a bunsen burner for 5 minutes. The crucible is allowed to cool and placed in a beaker containing hot 150cm^3 of distilled water into which about 10cm^3 of concentrated hydrochloric acid had been added. The beaker and its contents were kept boiling and stirred to complete dissolution. The solution is cooled and made up to 200cm^3 with deionised water in a volumetric flask. This is solution A. A solution of the standard ash and a blank are made up at the same time.

Dilutions

Solution	A	1% element = 5ppm
Solution	B	25cm^3 of solution A + 2cm^3 of La(10%)
Solution	C	4cm^3 of solution A + 1ml of conc HCl + 2cm^3 of La(10%) made up to 20cm^3

Conditions

Using 5cm burner (nitrous oxide burner) solution B, the instrument conditions used are as stated in Table 38. Solution C with burner in normal position for the concentration shown or in the 30° rotated for the higher concentrations; the instrument conditions used were as stated in Table 39.

Results and Discussions

Table 40 gives the results of analyses of Nigerian coal ash while Table 41 gives precision studies on the coal ash analysis. Table 41 shows that good precision was obtained in the coal ash analysis as indicated by the coefficient of variation values.

Table 38: Instrument conditions for using AAS for analysis of solution B for coal ash constituent oxides

Element	Lamp current (MA)	Wavelength (nm)	Slit (nm)	Burner height (cm)	Flame air	N_2O	C_2H_2	Linear range
Si	33	251.6	0.2	6	-	30	60	0-65% (SiO_2)
Al	25	396.2	0.7	6	-	30	60	0-30% (Al_2O_3)
Fe	10	372.0	0.2	7	40	-	25	0-50% (Fe_2O_3)
Ti	20	365.3	0.7	6	-	30	60	
Mn	15	279.5	0.2	7	40	-	25	

Table 39: Instrument conditions for using AAS for analysis of solution C for coal ash constituent oxides

	Lamp current (mA)	Wavelength (nm)	Slit (nm)	Burner height (cm)	Flame air	C_2H_2	Linear Range	
							Normal	30° Rotation
Ca	15	422.7	0.7	7	40	25	0-12	0-40% CaO
Mg	15	285.2	0.7	7	40	25	0-13	0-25% MgO
Na	5	589.0	0.7	7	40	25	0-3	0-25% Na_2O
K	6	766.5	2.0	7	40	25	0-5	0-25% k_2O

Table 40: Results of (wt%) of analysis of Nigerian coal ash constituent oxides.

Parameter	Lafia Coal	Enugu Coal	Okaba Coal
SiO_2	60.1	59.2	57.9
Fe_2O_3	15.3	6.3	8.7
MgO	1.1	0.3	0.5
CaO	1.4	0.2	2.4
Al_2O_3	18.4	30.1	24.1
TiO_2	0.9	2.1	2.4
K_2O	1.3	0.6	0.4
Na_2O	0.4	0.1	0.1
SO_3	0.7	0.2	2.4

TABLE 41: Precision studies for analysis of Nigerian coal-ash constituent oxides

%	A53	A53	A53	A53	A53	C.V.	A54	A54	A54	A54	A54	C.V.	A55	A55	A55	A55	A55	C.V.
SiO_2	60.9	60.1	60.6	61.2	60.3	0.73	60.0	60.2	60.1	60.1	59.9	0.20	57.9	58.0	57.9	57.9	58.3	0.29
Fe_2O_3	15.4	15.3	15.4	15.4	15.4	0.26	6.5	6.4	6.5	6.4	6.5	0.77	8.8	8.9	8.8	8.8	8.8	0.0
MgO	1.1	1.1	1.1	1.1	1.1	0.0	0.3	0.3	0.3	0.3	0.3	0.0	0.5	0.5	0.5	0.5	0.5	0.0
CaO	1.4	1.4	1.4	1.4	1.4	0.0	0.3	0.3	0.3	0.3	0.3	0.0	2.4	2.4	2.4	2.4	2.4	0.0
Al_2O_3	18.0	18.1	17.7	17.9	17.9	0.83	30.1	29.8	29.9	30.1	30.1	0.47	24.2	24.1	24.4	24.1	24.3	0.54
TiO_2	0.9	0.9	0.9	0.9	0.9	0.0	2.0	2.1	2.1	2.1	2.0	2.4	2.3	2.3	2.3	2.3	2.3	0.0
Na_2O	0.4	0.4	0.4	0.4	0.4	0.0	0.1	0.1	0.1	0.1	0.1	0.0	0.1	0.1	0.1	0.1	0.1	0.0
K_2O	1.3	1.2	1.2	1.2	1.2	3.3	0.6	0.6	0.6	0.6	0.6	0.0	0.3	0.3	0.3	0.3	0.3	0.0

LEGEND

A53 - Lafia coal ash
A54 - Enugu coal ash
A55 - Okaba coal ash
CV - Coefficient of variation

Table 42 compares the results of Lafia coal-ash with the British Allerton Bywater washed small's and some literature values. It is clear from Table 42 that the percentage of constituent oxides of Nigerian coal-ash are within the range reported for coal-ash in the literature. Silica in Nigeria coal-ash is higher than in the British coal-ash while the latter has higher alkalis and sulphur than the former. Furthermore, a comparison of the values of the major and minor elements in Nigerian coal-ash with those given in the literature for coal-ash and the Earth's crust are given in Table 43.

From the Table it is observed that the level of silicon (28%) in Nigerian coal-ash is much higher than in the coal-ash given in literature but it is only a little higher than the silicon concentration in the earth's crust (25.7%). Iron concentration in Nigeria coal-ash is the same value as that in literature but both are higher than in the earth's crust. Mg, Ca, and Na have by far higher concentrations in the literature coal-ash and the earth's crust than in Nigerian Lafia coal-ash.

Table 42: Comparison of coal ash constituent oxides with literature values

Coal ash constituent oxides	British Allerton Bywater washed smalls (wt%)	Nigerian Lafia coal ash (wt%)	Literature (2) concentration range of oxides in coal (wt%)
SiO_2	43.7	60	20-60
Fe_2O_3	19.3	15.3	5-35
MgO	1.4	1.1	0.3-4
CaO	2.20	1.4	1-20
Al_2O_3	25.3	18.4	10-35
TiO_2	0.93	0.9	0.5-2.5
Na_2O	2.00	0.4)	1-4
K_2O	3.00	1.3)	
Mn_3O_4	0.05	-	-
SO_3	1.6	0.7	0.1-12
P_2O_5	0.26	-	-

Table 43: Comparison of concentrations of elements (12) in coal ash and the earth's crust

Element	Nigerian Lafia coal ash (% wt)	Literature coal ash (% wt)	Earth's crust (% wt)
Si	28.0	3.31	25.7
Fe	10.7	10.7	4.7
Mg	0.66	4.8	2.0
Ca	1.0	35.6	3.4
Al	9.74	1.6	7.5
Ti	0.54	10^{-5}--0^{-4}	0.5
K	1.1	0.2	2.4
Na	0.3	2.3	2.6
S	0.28	-	-

Conclusion

Lithium metaborate fusion followed by acid digestion of Nigerian coal-ash provides a homogeneous solution which, with lanthanum chloride as a releasing agent, resulted in good precision using the flame methods of analysis to determine the constituent oxides. The values of the constituent oxides of Nigerian Lafia, Enugu and Okaba coal ash are similar but Lafia coal ash is higher in iron oxide than the others, while Enugu coal ash is higher in alumina than the others and the coal ash of Okaba has higher oxide of sulphur than the others. Also from Table 42 it is observed that Nigerian Lafia coal ash has lower values of the alkali oxides than does British Allerton Bywater washed smalls coal ash but both coals contain elemental oxides that are within the range described in the literature.

4.8. DETERMINATION OF TRACE ELEMENTS IN NIGERIAN COALS

Summary of Method

Whole coal was ashed, dissolved by mineral acids and the individual elements determined on Inductively Coupled Plasma (ICP) and the results confirmed by the Atomic Absorption Spectrophotometer [136].

Apparatus

1) Atomic Absorption Spectrophotometer, Perkin elmer 5000 and 270 models.
2) Inductively coupled plasma of the same make model 5500 was also used for trace element determination.
3) Muffle furnace with temperature control.
4) Polyethylene bottles, 125cm^3 with screw cap lids, capable of withstanding 130°C.
5) Volumetric flask, 100cm^3 capacity.
6) Steam bath.
7) Analytical Balance, capable of weighing to 0.1mg.
8) Crucible, 50cm^3 quartz or high silica.

Reagents

1) Microanalytical reagent grade of chemicals were used in all the chemistry work.
2) Saturated boric acid solution - 60 grams of H_3BO_3 dissolved in 1 litre of deionised water.
3) Aqua regia solution - a mixture of 1 part HNO_3, 3 parts HCl and 1 part H_2O.
4) Standard stock solution - prepared from high purity metals, avoiding H_2SO_4, which has adverse effects on the flame and gives some sulphate of low solubility. Stock solutions of at least 100ppm for each metal were prepared and dilution for standards in the range of 10 to 0.1ppm was made depending on the sensitivity of the element.

Calibration and Standardisation

Calibration standards were prepared from stock solutions appropriately diluted so that it finally contained 1.5% v/v aqua regia solution, 3% V/V HF, and 3% v/v H_3BO_3. The standards were made (Table 44) to closely approximate sample solution so that errors due to matrix differences were minimal with matrix containing 500ppm ($0.05g/100cm^3$), 1500ppm Al ($0.15g/100cm^3$), 1000ppm Ca ($0.10g/100cm^3$ 0.25g $CaCO_3$).

These were pipetted into 100cm^3 flask containing 5cm^3 6% H_3BO_3 and 5cm^3 HCl and then made to the mark with deionised water. A matrix minus the standards was prepared for a blank leaving out Scandium standard.

Sampling

A representative top size of 150μm air-dried whole coal was taken.

Procedure

6 grams sample of the coal was ashed in 50cm^3 quartz crucible. The sample was placed in a cold muffle furnace set to attain 300°C in about 1 hour and 500°C in 2 hours. The sample was stirred once each hour until no carbonaceous materials remain. After cooling and weighing the ash was ground in an agate mortar and then re-ignited at 500°C for 1 hour, cooled, and re-weighed, in order to calculate percent ash.

0.5g of the ash samples were placed in 125cm^3 polyethylene bottles with screw caps. 3cm^3 of aqua regia solution and 5cm^3 HF were added to the samples, screw caps tightened, and the bottles placed on a steam bath for at least 2 hours.

$50cm^3$ of saturated H_3BO_3 solution were added to the resultant solution. The use of H_3BO_3 solution not only complexes fluorine but it also has been shown to have good flame properties and acted as a flame buffer. (when a residue remained the mixture was reheated for about an hour to aid dissolution after which time the trace elements were regarded as having quantitatively been leached.) The solutions were cooled to room temperature and their volume adjusted to $50cm^3$ by the addition of deionised water. Samples were stored in polythylene bottles.

Analysis

Using the Atomic Absorption Spectrophotometer the parameters applied are given in Table 46 while in using the ICP the parameters are as given in Table 45.

Table 44: Calibration Standards for the use of Inductively Coupled Plasma and Atomic Absorption Spectrophotometer to Determine Trace Elements in Nigerian Coals.

Parameters	Concentrations (ppm)
P_2O_5	100
Zn	10
Ni	5
Cr	5
V	10
Cu	5
Sc	4

Table 45: Operational conditions for the use of the inductively coupled plasma in the determination of trace elements in Nigerian coals

Element File	Wavelength (nm)	Standards (ppm)	Integration time (secs)
P GO1	213.62	100 P_2O_5	2
Zn TE1	213.86	10	2
Ni TE1	231.60	5	2
Ca TE1	267.72	5	2
V TE1	292.4	10	2
Cu TE1	324.75	5	2
Sc TE1	357.25	4	2

Table 46: Operational Conditions for the Use of the Atomic Absorption Spectrophotometer for Trace Element Determination in Nigerian Coals

Element	Wavelength (nm)	Oxidant/Fuel
Be	234.9	N_2O/C_2H_2
Cd	228.8	Air/C_2H_2
Co	240.7	N_2O/C_2H_2
Cr	357.9	Air/C_2H_2
Cu	324.8	Air/C_2H_2
Mn	279.5	N_2O/C_2H_2
Mo	313.3	Air/C_2H_2
Ni	232.0	Air/C_2H_2
Pb	283.3	N_2O/C_2H_2
V	318.0	N_2O/C_2H_2
Zn	213.9	Air/C_2H_2

Further setting for the ICP was:-

Power	1250 watts
Viewing height	16mm
Nebulising pressure	28 psi
Pump max	(1000)
Flows	Argon
Plasma	$15dm^3/min$
Auxillary	$0.2dm^3/min$
Nebuliser	$0.5dm^3/min$

4.8.1 Results and Discussions

The results of trace element analysis in Nigerian coals by ICP are given in Table 47. The values of the trace elements in Nigerian coals were compared with those of trace elements in coals [137,138] from other countries such as Norway, England, U.S.A., W.Germany, Poland, Soviet Union and Australia, as contained in Table 48. From these results generally, the abundance of the elements determined in Nigerian Coals are as follows:

Lafia Coal:- F>Zn>Mn>>V≈Cr>Ni>Cu>B>Pb>Co>Be>Mo>Cd>Hg

Enugu Coal:- B>Mn>V≈F>Cr>Zn>Cu>Pb≈Ni>Co>Be>Mo>Hg

Okaba Coal:- Mn>>B>>V>Cr>Cu>Pb>F=Ni>=Zn≈Co>Be>Mo>Hg

The relative abundance of the elements in each coal depends on the biogeochemistry of the area of depsoit.

Table 47: Concentrations of Trace Elements (ppm) in Nigerian Coals

Parameter	Lafia Coal	Enugu Coal	Okaba Coal
Mo	1	1	1
Zn	166	16	5
Co	8	7	4
Cd	0.5	0.1	0.1
Ni	20	10	5
Mn	118	34	168
Cr	35	22	13
V	36	31	16
Be	1.3	1.9	1.5
Cu	18	13	9
Pb	14	11	7
F	180	30	5
B	16	57	85
Hg	0.06	0.14	0.10

TABLE 48: RESULTS OF ANALYSIS OF COAL SAMPLES IN ug/g (EXCEPT FOR S)

	Norway	England	U S A		West Germany		Poland		Soviet Union			Australia
			A	B	HA	W	A	B	A	B	C	
As	2.0	14.9	6.3	0.8	2.9	0.7	1.9	0.7	1.6	3.1	1.2	2.5
Ba	337	122	92	76	96	106	148	173	290	155	254	329
Be	5	5	5	13	5	5	5	5	5	5	5	15
Br	2	99	23	12	16	8	13	10	14	5	5	2
Ca	2200	1500	1500	500	1000	2100	4000	5400	5200	2800	450	240
Cd	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.3
Cl	30	3680	1580	580	490	800	1200	1300	1230	150	30	80
Co	0.6	6.5	2.7	13.2	5.0	6.5	3.0	4.1	4.1	6.6	3.5	18.4
Cr	1.8	12.5	11.4	3.9	10.3	15.4	6.5	8.6	7.7	17.4	7.7	6.5
Cu	5	22	14	15	12	21	14	13	24	8	6	15
F	54	75	39	17	45	42	72	32	58	53	83	64
Fe	6200	6100	9100	1370	6600	2300	9000	9600	6000	26200	14500	2000
Hg	0.066	0.120	0.066	0.015	0.124	0.115	0.050	0.070	0.177	0.111	0.074	0.015
In	0.009	0.028	0.013	0.005	0.019	0.028	0.005	0.020	0.023	0.022	0.021	0.074

TABLE 48: (Contd.)

	Norway	England	U S A		West Germany		Poland		Sovjet Union			Australia
			A	B	HA	W	A	B	A	B	C	
Mn	10	47	16	10	224	24	127	134	151	36	77	21
Mo	0.26	2.68	0.70	0.99	1.69	1.49	0.61	0.39	1.31	1.51	1.01	2.23
Na	1340	1820	240	410	310	550	970	950	1160	860	340	60
Ni	50	50	50	50	50	50	50	50	50	50	50	50
Pb	10	10	10	10	10	10	10	10	10	10	10	60
S	0.78%	1.12%	1.02%	0.61%	0.85%	0.79%	0.60%	0.76%	0.70%	0.50%	0.40%	0.47%
Sb	0.02	1.54	0.28	0.08	1.47	0.92	0.78	0.82	0.97	0.47	0.14	2.47
Se	1.5	1.4	3.7	1.8	1.0	1.8	0.9	1.8	1.5	1.2	0.6	0.5
Sr	286	15	60	87	57	56	50	37	163	128	105	180
Ti	140	810	800	100	200	360	340	350	920	1050	610	1120
Tl	20	20	20	20	20	20	20	20	20	20	20	20
V	5.5	39.5	16.6	5.8	23.8	34.0	8.7	14.5	24.2	26.3	12.7	20.5
Zn	0.3	15.1	3.1	1.1	9.6	13.3	24.4	34.0	16.7	23.7	17.3	116

Whereas Fluorine is the most abundant element in Lafia Coal, Boron is the most abundant element Enugu Coal and Manganese is the most abundant element in Okaba Coal. However, mercury is the least abundant element in all the three coal types studied. F(180ppm), Zn(166ppm) and Mn(118ppm) are the elements with concentrations 100ppm and above in Lafia Coal. No element in Enugu Coal has a concentration as high as 100ppm, while only Mn(168ppm) is the only element with concentration above 100ppm in Okaba Coal.

The levels of concentrations of the heavy metals Pb, Hg, Cd, and Cn in Nigerian coals are similar to their concentrations in the coals from the countries compared. The elements Zn, Cr, Ni, and V in Lafia coal are of higher concentration than in Enugu and Okaba coals from Nigeria and coals from other countries. Be is lower in Nigerian coals than in coals from other countries.

Conclusion

Since Lafia coal has the elements determined at concentrations that may be ecologically significant, combustion of Lafia coal will no doubt result in atmospheric emissions of these elements. This could cause environmental pollution of land and water by increasing metallic burden in aquatic ecosystems and bilogical samples, which could cause deleterious health effects in man and other living sources.

CHAPTER FIVE

WASHABILITY TEST ON NIGERIAN LAFIA COAL

5.1. INTRODUCTION

Washability test was carried out on Nigerian Lafia coal with the aim of determining the prospects of up-grading it. From the characterization of its physico-chemical properties (discussed in chapter four) its high ash and high sulphur contents were established. Its cokeability was also brought to light from the crucible swelling number (CSN) of 5 to 6 for the run-of-mine (ROM) coal described in section 4.6. To be usable for metallurgical coke preparation its ash has to be brought down below 10% and its sulphur below 1.0%.

5.2. Apparatus

Simple laboratory wares were used including Nicolson hydrometer for adjusting the specific gravity of the liquid medium. Carbon tetrachloride, benzene, bromoform, zinc chloride solution, sulphuric acid and water were used in the test to prepare media of varying densities. Air-oven was used for drying off the moisture as well as remains of organic liquids that adsorbed on to coal particles.

Muffle furnace was used to ash the coal to determine the ash component. A chemical balance sensitive enough to read to four decimal places was used in weighing the reagents. Glass beakers, plastic beakers, and gas jars and glass stirring rods were other apparatus used for the practical work (Figure 29).

5.3. Analytical Procedure for Washability test on Lafia Coal

5.3.1. Grinding and Screening of Coal Sample

For the float-and-sink tests the coal was ground manually to pass 100 percent through minus 60 mesh sieve. Lafia coal belonging to the high volatile bituminous A rank was found to be rather easy to grind. We started with sizes of eggs and nuts and these were crushed with the pestle in the mortar. To avoid generating too much fines during the grinding operation the sample was ground in small batches. After about five to ten impacts of the pestle against the coal in the mortar the coal was transferred onto the sieve and the portion of it under minus 60 mesh grain size passed through and was collected. The remaining portion that could not pass through the sieve aperture was returned to the mortar for more pestle impacts after which it was again sieved.

The sequence of crushing, grinding and sieving was repeated until all the sample passed through the desired sieve. The coal sieved batches were put together and stirred using a mechanical stirrer in an enclosure to prevent air from blowing off the coal fines and dust. After thoroughly mixing the sample it was then riffled using a mechanical riffler. Sub-samples of 300g each were prepared for each float-and-sink test run.

5.3.2. Preparation of float-and-sink bath medium

Carbon tetrachloride (SG. 1.584), benezene (SG. 0.879) and bromoform (S.G. 2.89) were the only organic media readily available and used during the course of this study. Zinc chloride solution was also used. In few cases sulphuric acid was used to prepare the required aqueous media density. Care was exercised to minimize inhalation of the vapours of the organic liquid media being aware of their toxicity and carcinogenic cumulative physical effects on the human body. Such caution exercised consisted in wearing moist satin nose-hood and facial mask with eye goggles since such vapours could diffuse into the blood stream through thin epithelial linings of the eye lids, nose, and lips.

Gloves were also worn on the hands out of caution. The mixing of liquids and solutions were done in a fume-cupboard with extractor fan on. The baths of the specific gravity series 1.200 to 1.800 (and in some cases higher than 1.800) were lined up in the fume chamber and the experiments ran in there. In places and on occasions where the fume chamber was either not available or its extractor fan not working well the experiments were run in the open air so that natural air currents would dilute the media chemical vapours. Some experiments were run in well aerated spacious laboratory rooms and some in the open air within the laboratory premises. Furthermore, to prevent environmental pollution the spent media liquids and chemicals were stocked in large labelled glass or polypropylene caskets for re-use on future occasions, rather than throwing same on to the soil which would allow percolation and pollution of soil, ground and surface water to the detriment of life plants and the food chain with man at its apex.

The appropriate volumes of the media liquids were taken and mixed to produce media bath of the desired specific gravities. The formula applied to determine the volume of each liquid to go into the mixture to produce medium bath was:

$$V_1d_1 = V_2d_2 + V_3d_3$$

where V_1 was the volume of the bath medium

d_1 was the density of the bath medium

V_2 was the volume of one of the bath liquids to be mixed

d_2 was the density of the bath liquid whose volume was V_2

V_3 was the volume of the second bath liquid to be mixed

d_3 was the density of the second bath liquid whose volume was V_3

For example, to prepare the bath medium whose volume was 500cm^3 and specific gravity was 1.202 using carbon tetrachloride and benzene whose respective specific gravities were 1.584 and 0.879 the calculation ran as follows

$$V_1d_1 = V_2d_2 + V_3d_3$$

(bath medium) (carbon tetrachloride (benzene)

Assume we take 300cm^3 of carbon tetrachloride

$$\text{then} \quad 500 \times 1.202 = (300 \times 1.584) + (0.879V_3)$$

$$601 = 475.2 + 0.879V_3$$

$$V_3 = 143.12\text{cm}^3.$$

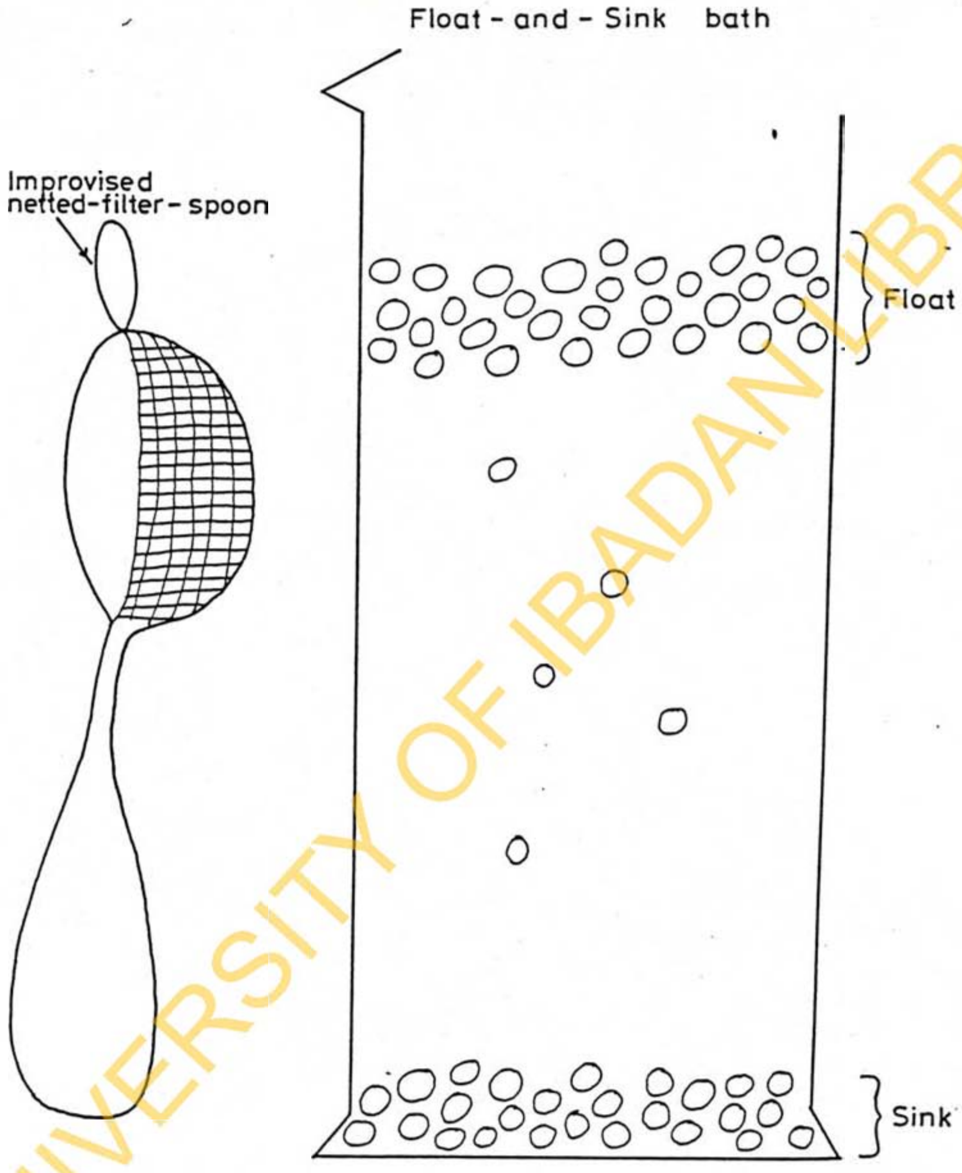


Fig. 29: Instrumentation for Washability Test

143.12cm^3 of benzene was added to the 300cm^3 of carbon tetrachloride in a clean tall glass beaker to produce the 500cm^3 of the bath medium whose specific gravity was 1.202. Nicholson hydrometer was used to ascertain that the desired specific gravtiy was achieved. If it was required to lower the specific gravity by 0.1, or less, micro-pipette was used to introduce tiny drops of benzene to the mixture. The mixture was stirred with a glass rod and the specific gravity re-measured and the process was repeated until the desired specific gravity was obtained. If it was required to raise the specific gravity of the bath medium produced from the mixture of several bath liquids, then,rather than add benzene, it was carbon tetrachloride that was added, in micro-droplets, and the hydrometer used to regulate the specific gravity to the desired value.

5.3.3. Procedure for Washability Test

The liquids of prepared specific gravities were arranged in descending order of their densities in 2000cm^3 tall glass or polypropylene measuring cylinders or beakers. The coal was introduced into the baths carefully in a place with still air to prevent the fine dust part of the sample from being blown off by air currents.

A glass stirrer was then used to stir the sample to get it well wetted with the bath medium. The coal slurry was then left for a total of fifteen minutes resident time in the bath. Thereafter the floated coal was skimmed off using a fine filter and transferred directly into the bath of the next lower specific gravity. Again the bath was stirred using glass stirring rod to get the coal properly wetted by the bath medium and also to counter the surface tension forces which otherwise would reduce the effect of the gravimetric force which was responsible for the sinking particles. Again after fifteen minutes resident time the floated coal fraction was scooped off using the improved sieve-like netted-filter-spoon. The scoop was again transferred directly and immediately into the bath of the next lower specific gravity.

While waiting for the fifteen minutes resident time of each coal float fraction the sink was dewatered, filtered and drained dry on a white satin cloth giving a coal-sink-cake. The cake was spread thinly on the satin cloth and put on a shelf in an air-oven at 30°C temperature for 24 hours to dry. When dry the sink fraction was weighed, then prepared for laboratory chemical analysis by grinding it to pass 100% through minus 72 mesh sieve.

The sink fraction was analysed for every medium bath. Only in the last bath that both the sink and the float were analysed.

Different particle sizes of coal were treated in different density media to see which particle size of Lafia coal would wash best if later the coal is being washed at the commercial scale.

For the different sizes and specific gravities the data obtained were tabulated and later plotted as curves. The different curves to be plotted and the interpretations derivable from them have been explained in the literature review given in Chapter 2.

5.4. Results and Discussions

The results of the washability test of Nigerian Lafia coal are depicted in Tables 49, 50, 51, 52, 53 and 54. Each result's Table is the mean of three determinations that were averaged to establish the data in Table 50. The data in Table 49 were plotted into family of curves in figure 30 while the data in Table 50 were plotted into the curves in figure 31. The calculations that generated the data of the washability test results were worked out as explained in section 2.5.1.2. The family of curves also were developed in line with the description in section 2.5.1.2 and their interpretation will follow the sample given in section 2.5.1.4.

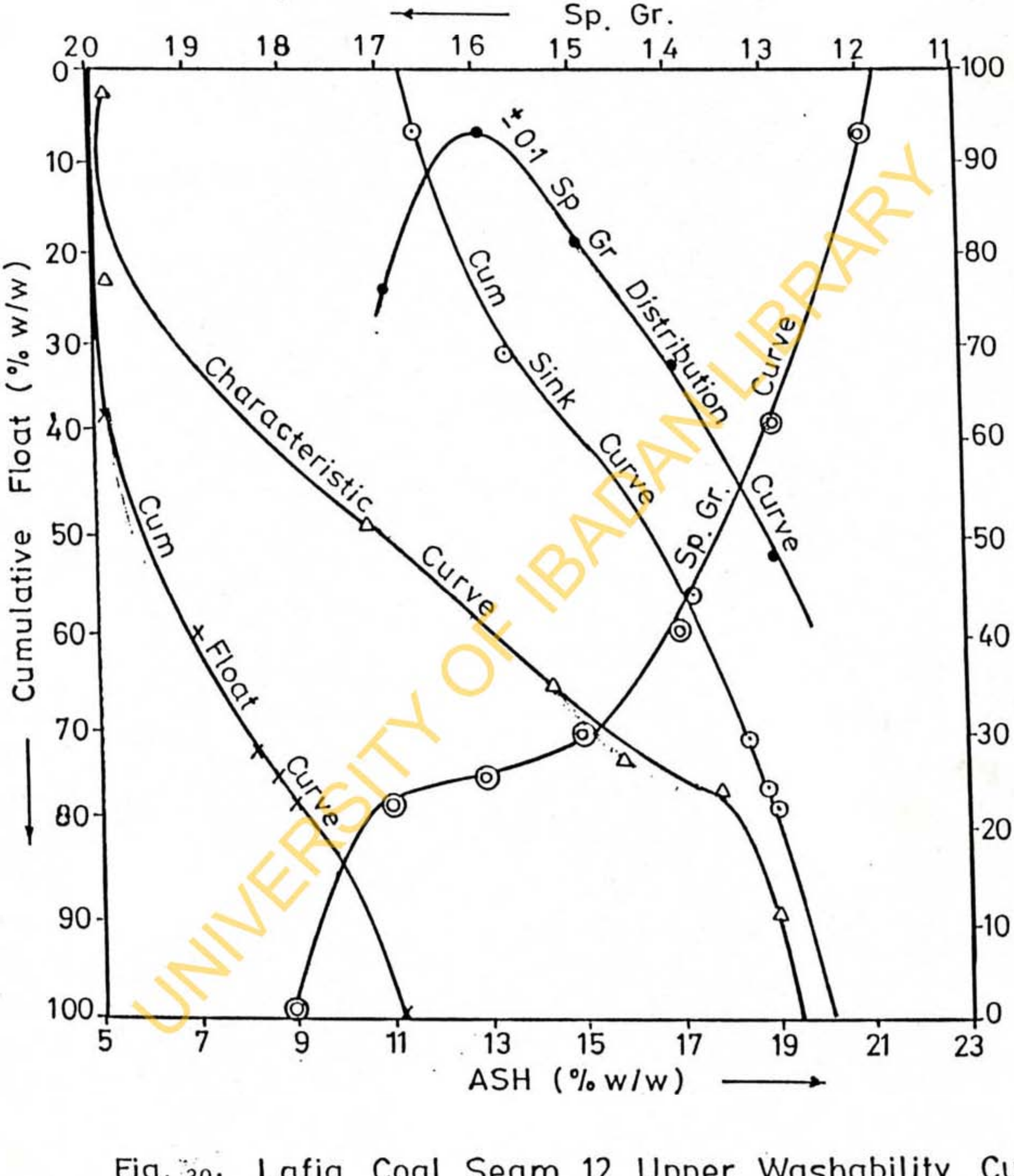


Fig. 30: Lafia Coal Seam 12 Upper Washability Curves using Benzene and Carbon Tetrachloride Media for minus 60 Mesh by 0 (Single Run).

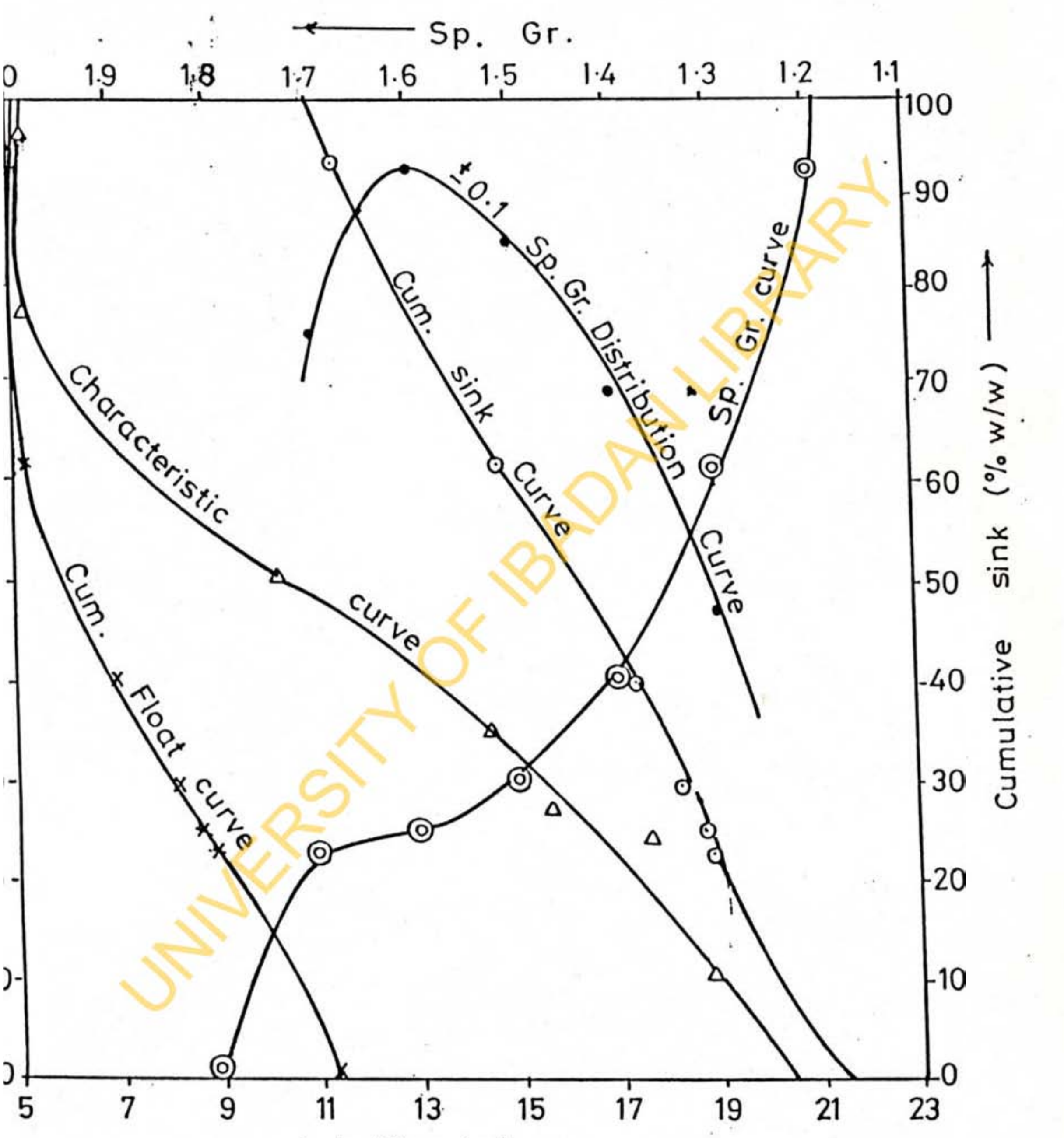


Fig. 31: Lafia Coal Seam 12 Upper Washability Curves using Benzene and Carbon Tetrachloride Media for minus 60 Mesh by 0 (Average of Nine Determinations).

TABLE 49:

RESULTS OF WASHABILITY TEST OF LAFIA COAL SEAM 12 SHAFT 2 PARTICLE SIZE MINUS 60 MESH
(One run out of nine)

S/NO	Specific gravity	Float Fractions (% wt)	Float Fractional Ash (% wt)	Float Cum Fractions (% wt)	Float Cum Ash of Fractions (% wt)	Sink Cum Fractions (% wt)	Sink Cum Ash of Fractions (% wt)	Mid-Point of sort (% wt)	S.G. Distribution (± 0.1)
1	1.202	7.03	5.22	7.03	5.22	92.97	11.68	3.52	x
2	1.302	31.43	5.17	38.46	5.18	68.57	13.47	22.75	52.53
3	1.402	21.10	10.61	59.56	7.10	40.44	17.30	49.01	31.92
4	1.502	10.82	14.32	70.38	8.21	29.62	18.39	64.97	15.70
5	1.602	4.88	15.92	75.26	8.71	24.74	18.88	72.82	7.12
6	1.702	2.24	17.84	77.50	8.98	22.50	18.98	76.38	24.75
7	1.800	22.51	18.97	100.01	11.22	0	x	88.76	x

TABLE 50:

RESULTS OF WASHABILITY TEST OF LAFIA COAL SEAM 12 SHAFT 2 PARTICLE SIZE MINUS 60 MESH (MEAN 9 TRIALS)

Float fraction number of starting coal	Specific Gravity	Float fractions (g)	Float fractions (% w/w)	Float fractional ash (% w/w)	Float cumulative fractions (% w/w)	Float cumulative ash of fractions (% w/w)	Sink cumulative fractions (% w/w)	Sink cumulative ash of fractions	Mid point of sort (% wt)	Specific gravity distribution (% w/w)((± 0.1)
1	2	0	3	4	5	6	7	8	9	10
1	1.201	18.42	6.90	5.18	6.91	5.18	93.09	11.54	3.46	x
2	1.302	84.16	31.56	5.24	38.46	5.23	61.54	14.63	22.69	52.58
3	1.402	56.07	21.03	10.24	59.49	7.00	40.51	17.17	48.98	31.71
4	1.503	28.50	10.69	14.35	70.18	8.17	29.82	18.18	64.83	15.59
5	1.601	13.09	4.91	15.65	75.09	8.64	24.92	18.67	72.63	7.23
6	1.704	6.20	2.32	17.53	77.41	8.88	22.59	18.79	76.25	24.91
7	1.801	60.24	22.59	8.79	100.00	11.12	0	x	88.71	x

Legend

(-) - float at or lighter than

(+) - sink at or heavier than

(x) - reference opoint

TABLE 51

RESULTS OF WASHABILITY TEST OF LAFIA COAL SEAM 12 SHAFT 2, PARTICLE SIZE 1-12mm

Float fraction number of starting coal	Specific gravity of medium bath 2	Float fractions (% wt)	Float fractional Ash	Float cum fractions (% wt)	Float cum Ash of fractions (% wt)	Sink cum fractions (% wt)	Sink cum Ash of fractions (% wt)	Mid-point of sort (% wt)	S.G. Distribution (± 0.1)
1	-1.3	3.95	11.2	3.95	11.2	96.05	26.12	1.98	x
2	+1.3 -1.4	44.76	14.4	48.71	14.14	51.29	36.34	26.33	66.26
3	+1.4 -1.5	21.50	24.4	70.21	17.28	29.79	44.96	59.46	37.28
4	+1.5 -1.6	15.78	35.3	85.99	20.63	14.01	55.83	78.10	23.81
5	+1.6 -1.8	8.03	44.3	94.02	22.65	5.98	71.32	90.01	9.66
6	+1.8 -2.0	1.63	58.3	95.65	23.25	4.35	76.2	94.84	5.98
7	+2.0	4.35	76.2	100.00	25.56	0	x	97.83	x

Legend

(-)- float at or lighter than
(+)- sink at or heavier than
Cum- cumulative
(x)- reference point

For the experiments that produced the data of Table 51 the specific gravity of the bath media in which the coal was floated-and-sunk ranged between 1.3 and 2.0. Carbon tetrachloride (SG. 1.584) and benzene (S.G. 0.879) were mixed to get the bath media whose specific gravity were lower than 1.584. In the case of the bath media whose specific gravity were higher than 1.584 a mixture of bromoform (S.G. 2.89) and carbon tetrachloride was used. The volumes of the bath liquids to take to mix with another one in order to produce the desired bath medium of known volume and specific gravity were calculated as illustrated in section 5.3.2.

From Table 51 it is seen that the float fractional ash of 11.2% was obtained from the float fractions of 3.95 percent which came from the medium bath of the lowest specific gravity 1.3. The highest yield of float fractions of 44.76 percent was obtained from the bath medium with specific gravity 1.3 and it had float fractional ash of 14.4 percent. The float cumulative of fractions yield 85.99 percent obtained from the bath medium of specific gravity of 1.5 had float cumulative ash of fractions of 20.63 percent. Now, in industrial washing the cut-off point for clean coal is commonly about the specific gravity of 1.5.

For metallurgical coal the ash content should not be higher than 10 percent. The product of this washability test at the specific gravity of 1.5 which had float cumulative ash of fractions of 20.63 has not met the requirements for metallurgical coal. Therefore, the particle size 1-12mm was too coarse to wash Lafia coal for metallurgical coal use. Following this observation it was decided to crush Lafia coal (seam 12) to minus 6.7 mesh by 0 and to further classify the sample into sizes minus 6.7 plus 3.35mm, minus 3.35 plus 1mm, minus 1mm plus 0.5mm, minus 0.5mm by 0. The size minus 6.7 mesh by 0 was itself treated as a sample. The results are given in Table 52.

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Table 52.[illegible]: WASHABILITY TEST Particle size minus 6.7mm by 0

SUB-FRACTION		FLOTATION FRACTIONS						CUMULATIVE FLOATS					CUMULATIVE SINKS				
	% TOTAL	SPECIFIC GRAVITY	% SUB-SEC	% TOTAL	ANALYSIS % ASH	TOT SUL	CL	% SUB-SEC	% TOTAL	ASH	TOT SUL	CL	SUB-SEC	TOTAL	ASH	TOT SUL	CL
-6.7+3.35mm	31.6	FLOATS @ 1.30	0.90	0.28	6.5	1.37	0.00	0.9	0.3	6.5	1.37	0.00	100.0	31.6	27.5	1.54	0.00
		1.30- 1.40	29.80	9.42	14.1	1.22	0.00	30.7	9.7	13.9	1.22	0.00	99.1	31.3	27.5	1.54	0.00
		1.40- 1.50	41.20	13.02	21.6	1.02	0.00	71.9	22.7	18.3	1.11	0.00	69.3	21.9	33.5	1.68	0.00
		1.50- 1.60	9.70	3.07	30.7	1.38	0.00	81.6	25.8	19.8	1.14	0.00	28.1	8.9	50.9	2.65	0.00
		1.60- 1.70	6.90	2.18	40.3	2.36	0.00	88.5	28.0	21.4	1.23	0.00	18.4	5.8	61.6	3.32	0.00
		1.70- 1.80	1.30	0.41	47.0	3.10	0.00	89.8	28.4	21.7	1.26	0.00	11.5	3.6	74.4	3.90	0.00
		SINKS @ 1.80	10.20	3.22	77.9	4.00	0.00	100.0	31.6	27.5	1.54	0.00	10.2	3.2	77.9	4.00	0.00
-3.35+1mm	26.4	FLOATS @ 1.30	1.80	0.48	5.5	1.30	0.00	1.8	0.5	5.5	1.30	0.00	100.0	26.4	27.9	1.57	0.00
		1.30- 1.40	25.90	6.84	13.0	1.32	0.00	27.7	7.3	12.5	1.32	0.00	98.2	25.9	28.3	1.58	0.00
		1.40- 1.50	35.50	9.37	20.5	1.08	0.00	63.2	16.7	17.0	1.18	0.00	72.3	19.1	33.8	1.67	0.00
		1.50- 1.60	15.60	4.12	29.2	1.17	0.00	78.8	20.8	19.4	1.18	0.00	36.8	9.7	46.7	2.24	0.00
		1.60- 1.70	7.30	1.93	38.8	2.04	0.00	86.1	22.7	21.1	1.25	0.00	21.2	5.6	59.5	3.02	0.00
		1.70- 1.80	2.70	0.71	46.0	2.49	0.00	88.8	23.4	21.8	1.29	0.00	13.2	3.7	70.4	3.54	0.00
		SINKS @ 1.80	11.20	2.96	76.3	3.79	0.00	100.0	26.4	27.9	1.57	0.00	11.2	3.0	76.3	3.79	0.00
-1.0+0.5mm	12.1	FLOATS @ 1.30	3.51	0.42	5.9	1.36	0.00	3.5	0.4	5.9	1.36	0.00	100.0	12.1	30.7	1.60	0.00
		1.30- 1.40	12.63	1.53	13.6	1.26	0.00	16.1	2.0	11.9	1.28	0.00	96.5	11.7	31.6	1.60	0.00
		1.40- 1.50	37.07	4.49	20.6	1.07	0.00	53.2	6.4	18.0	1.13	0.00	83.9	10.1	34.3	1.66	0.00
		1.50- 1.60	17.54	2.12	23.3	1.09	0.00	70.7	8.6	20.3	1.12	0.00	46.8	5.7	45.2	2.12	0.00
		1.60- 1.70	10.92	1.32	37.6	1.62	0.00	81.7	9.9	22.8	1.19	0.00	29.3	3.5	55.4	2.74	0.00
		1.70- 1.80	4.01	0.48	13.3	2.17	0.00	85.7	10.4	23.8	1.24	0.00	18.3	2.2	60.0	3.40	0.00
		SINKS @ 1.80	14.33	1.73	72.3	3.75	0.00	100.0	12.1	30.7	1.60	0.00	14.3	1.7	72.3	3.75	0.00
-0.5mm	29.9	FLOATS @ 1.30	1.80	0.54	3.6	1.33	0.00	1.8	0.5	3.6	1.33	0.00	100.0	29.9	28.5	1.67	0.00
		1.30- 1.40	20.80	6.22	9.1	1.33	0.00	22.6	6.8	8.7	1.33	0.00	98.2	29.4	28.9	1.67	0.00
		1.40- 1.50	20.50	8.52	16.9	1.20	0.00	51.1	15.3	13.3	1.26	0.00	77.4	23.1	34.3	1.76	0.00
		1.50- 1.60	15.40	4.60	24.8	1.18	0.00	66.5	19.9	15.9	1.24	0.00	48.9	14.6	44.4	2.09	0.00
		1.60- 1.70	11.40	3.32	31.7	1.43	0.00	77.8	23.3	18.2	1.27	0.00	33.5	10.0	53.4	2.51	0.00
		1.70- 1.80	1.80	0.54	33.4	1.71	0.00	79.6	23.8	18.6	1.28	0.00	22.2	6.6	64.5	3.06	0.00
		SINKS @ 1.80	20.40	6.10	67.2	3.18	0.00	100.00	29.9	28.3	1.67	0.00	20.4	6.1	67.2	3.18	0.00
-6.7+1.0mm	55.0	FLOATS @ 1.30	1.31	0.76	5.9	1.33	0.00	1.3	0.8	5.9	1.33	0.00	100.0	58.0	27.7	1.56	0.00
		1.30- 1.40	28.02	16.25	13.6	1.26	0.00	29.3	17.0	13.3	1.26	0.00	98.7	57.2	28.0	1.56	0.00
		1.40- 1.50	38.41	22.39	21.1	1.05	0.00	67.3	39.4	17.8	1.14	0.00	70.7	41.0	33.6	1.68	0.00
		1.50- 1.60	12.39	7.18	29.8	1.26	0.00	80.3	46.6	19.6	1.16	0.00	32.1	18.6	48.7	2.43	0.00
		1.60- 1.70	7.08	4.11	39.6	2.21	0.00	87.4	50.7	21.2	1.24	0.00	19.7	11.4	60.6	3.17	0.00
		1.70- 1.80	1.94	1.12	46.4	2.71	0.00	89.3	51.8	21.8	1.28	0.00	12.6	7.3	72.4	3.72	0.00
		SINKS @ 1.80	10.66	6.18	77.1	3.90	0.00	100.0	58.0	27.7	1.56	0.00	10.7	6.2	77.1	3.90	0.00

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Table 52: (contd.) WASHABILITY TEST

SUB-FRACTION		FLOTATION FRACTIONS						CUMULATIVE FLOATS					CUMULATIVE SINKS				
					ANALYSIS												
	% TOTAL	SPECIFIC GRAVITY	% SUB-SEC	% TOTAL	ASH	TOT SUL	CL	% SUB-SEC	% TOTAL	ASH	TOT SUL	CL	SUB-SEC	TOTAL	ASH	TOT SUL	CL
-6.7 x0mm	100.0	FLOATS@ 1.30	1.72	1.72	5.2	1.34	0.00	1.7	1.7	5.2	1.34	0.00	100.0	100.0	28.3	1.59	0.00
		1.30- 1.40	24.00	24.00	12.5	1.28	0.00	25.7	25.7	12.0	1.28	0.00	98.3	98.3	28.7	1.60	0.00
		1.40- 1.50	35.40	35.40	20.1	1.09	0.00	61.1	61.1	16.7	1.17	0.00	74.3	74.3	33.9	1.70	0.00
		1.50- 1.60	13.91	13.91	27.9	1.21	0.00	75.0	75.0	18.7	1.18	0.00	38.9	38.9	46.6	2.26	0.00
		1.60- 1.70	8.81	8.81	36.3	1.82	0.00	83.8	83.8	20.6	1.24	0.00	25.0	25.0	57.0	2.85	0.00
		1.70- 1.80	2.15	2.15	42.4	2.34	0.00	86.0	86.0	21.1	1.27	0.00	16.2	16.2	68.3	3.40	0.00
		SINKS @ 1.80	14.01	14.01	72.2	3.57	0.00	100.0	100.0	28.3	1.59	0.00	14.0	14.0	72.2	3.57	0.00
-3.35 +0.5mm	38.5	FLOATS@ 1.30	2.34	0.90	5.7	1.33	0.00	2.3	0.9	5.7	1.33	0.00	100.0	38.5	28.8	1.58	0.00
		1.30- 1.40	21.73	8.37	13.1	1.31	0.00	24.1	9.3	12.4	1.31	0.00	97.7	37.6	29.4	1.59	0.00
		1.40- 1.50	35.99	13.86	20.5	1.08	0.00	60.1	23.1	17.3	1.17	0.00	75.9	29.2	34.0	1.66	0.00
		1.50- 1.60	16.21	6.24	28.9	1.14	0.00	76.3	29.4	19.7	1.16	0.00	39.9	15.4	46.1	2.19	0.00
		1.60- 1.70	8.44	3.25	38.3	1.87	0.00	84.7	32.6	21.6	1.23	0.00	23.7	9.1	57.9	2.91	0.00
		1.70- 1.80	3.11	1.20	44.9	2.36	0.00	87.8	33.8	22.4	1.27	0.00	15.3	5.9	68.7	3.49	0.00
		SINKS @ 1.80	12.18	4.69	74.8	3.78	0.00	100.0	38.5	28.8	1.58	0.00	12.2	4.7	74.8	3.78	0.00
-1.0 x0mm	42.0	FLOATS@ 1.30	2.29	0.96	4.6	1.34	0.00	2.3	1.0	4.6	1.34	0.00	100.0	42.0	29.1	1.65	0.00
		1.30- 1.40	18.44	7.75	10.0	1.32	0.00	20.7	8.7	9.4	1.32	0.00	97.7	41.0	29.7	1.65	0.00
		1.40- 1.50	30.97	13.01	18.2	1.16	0.00	51.7	21.7	14.7	1.22	0.00	79.3	33.3	34.3	1.73	0.00
		1.50- 1.60	16.02	6.73	25.9	1.15	0.00	67.7	28.4	17.3	1.20	0.00	48.3	20.3	44.6	2.10	0.00
		1.60- 1.70	11.19	4.70	33.4	1.48	0.00	78.9	33.1	19.6	1.24	0.00	32.3	13.6	53.9	2.57	0.00
		1.70- 1.80	2.44	1.02	38.1	1.93	0.00	81.3	34,2	20.1	1.26	0.00	21.1	8.9	64.8	3.15	0.00
		SINKS @ 1.80	18.65	7.83	68.3	3.32	0.00	100.0	42.0	29.1	1.65	0.00	18.7	7.8	68.3	3.31	0.00

TABLE 53:

RESULTS OF WASHABILITY TEST OF LAFIA COAL SEAM 12 SHAFT 2 PARTICLE SIZE 0-1mm

Float fraction number of the starting coal	Specific gravity	Float fractions (% wt)	Float fractional Ash (% wt)	Float cumulative fractions (% wt)	Float cumulative ash of fractions (% wt)	Sink cumelative fractions (% wt)	Sink cumulative ash of fractions (% wt)	Mid-point of sort (% wt)	Specific distribution ($\pm$ 0.1)
1	-1.40	40.08	11.6	40.08	11.60	59.92	37.92	20.04	x
2	+1.40 -1.50	18.32	19.8	58.40	14.17	41.60	45.24	49.24	18.32
3	+1.50 -1.80	25.57	36.5	83.97	20.97	16.03	60.90	71.19	25.57
4	+1.80	16.03	60.9	100.00	27.37	0	x	91.99	16.03
									x

Legend

(-) - float at or lighter than

(+) - sink at or heavier than

cum - cumulative

(x) - reference point

At a cut-off point of specific gravity between 1.50 and 1.60 and for the particle size minus 6.7 plus 3.35mm sulphur in the original coal of 2.8% went down to 1.14 subsequent to the washing. Desulphurization achieved was thus 59.3%. Ash reduction achieved was 39.1% or from 32.5% ash content in unwashed coal to 19.8% after the coal had been washed. The sub-section (minus 6.7 by plus 3.35mm) yield of coal concentrate was 81.6% which was 25.8% of the total crushed coal of minus 6.7mm by 0.

At the cut-off point of specific gravity between 1.50 and 1.60 and for the particle size minus 3.35 plus 1mm the sub-section yield was 78.8%, the ash removal was 40.3% while desulphurization achieved was 57.86%.

Again considering specific gravity of between 1.5 and 1.6 as the cut-off point and for particle size minus 1 plus 0.5mm the sub-section yield was 70.0%, the ash removal was 37.54% and the desulphurization achieved was 60%.

From the result Table it is seen that at a cut-off point of specific gravity between 1.50 and 1.60 for the minus 6.7mm by 0 75.0% was the sub-sectional yield, which was also 75.0% yield of the total coal sample. Ash removed was 42.46% and desulphurization achieved was 57.86%.

The specific gravity of between 1.50 and 1.60 has been chosen for consideration of cumulative float product because it is the specific gravity range used in industrial coal washery which is the ultimate goal of this study. Desulphurization achieved was in each case higher than 55% but ash removal was only a mean of about 40%. The ash content of the washed coal product was about 19.0% on the average. The ash remaining in the coal after washing was thus too high for metallurgical coal. It therefore, was necessary to further crush Lafia coal down to finer-sized particles for good ash reduction and desulphurization.

Another batch of Lafia coal sample was crushed down to minus 1mm by 0 particle size and then tested for float-and-sink. The results are contained in Table 53. The lowest float cumulative ash of fractions was 11.60% and was obtained against float cumulative fractions of 40.08%. The next higher float cumulative ash of fractions was 14.17% obtained against the yield of 58.40% of float cumulative fractions. Again the ash is too high for metallurgical coal which should have ash content less than 10%.

Another batch of Lafia coal was crushed down to a top size of minus 60 mesh by 0 and tested for float-and-sink. The results are tabulated in Table 49.

The specific gravities of bath media of the work that produced Table 49 ranged from 1.202 to 1.800. It was note-worthy that the float cumulative ash of fractions of the sample went as far down as 8.98% against clean float cumulative fractions of 77.50% at the cut-off specific gravity of 1.702. The lowest float cumulative ash of fractions was 5.22% against float cumulative fractional yield of 7.03% at the cut-off specific gravity of 1.202. Seeing that at the particle size of minus 60 mesh by 0 the clean coal product had ash lower than 10% which is the upper end of the ash level that metallurgical process could tolerate in coal fuel the float-and-sink of this particle size was repeated nine times. Table 49 is one of these nine runs while Table 50 is the mean of the nine determinations.

Both Tables 49 and 50 were then plotted into curves in Figures 28 and 29.

The washability curves tell very much about the response of seam 12 upper to washing. Two families of washability curves (Figures 28 and 29) were drawn; the first for one of the nine experiments and the second for the average of the nine runs. The similarity of the curves is a confirmation of the reproducibility of the float-and-sink experiments. Figure 29 will be used to interprete the results obtained.

The characteristic curve Figure 29 shows that Lafia coal seam 12 upper could be washed between specific gravities 1.40 and 1.65 judging from the fact that it is between these two specific gravity points that the curve is flatter than the remaining part of its curves. From the ±0.1 specific gravity distribution curve the percent distribution at specific gravities 1.45, 1.50, 1.55, 1.60 and 1.65 are respectively 21.0, 15.0, 10.0, 8.0, and 10.0. If we superimpose Bird's Table (Table 22, section 2.5.1.5) on the washability curves then we see that at specific gravities between 1.55 and 1.65 Lafia coal can be washed though with some but moderate difficulty and required efficient process to yield high tonnages under good operation. Its washing becomes progressively more difficult at specific gravities lower than 1.55 and higher than 1.65. At specific gravity of separation of 1.60 the yield of this minus 60 mesh size of Lafia coal will be 75.0 percent which will have cumulative ash of float of 8.5 percent and the dirtiest piece of coal in the yield will then assay 16.5 percent ash content. A cut-off at specific gravity 1.45 will yield 63 percent of clean coal with cumulative ash of float of 7.0 percent and the dirtiest piece of coal will assay 14.0 percent ash. At 1.50 specific gravity the yield will be 69 percent with over-all ash of 8.0 percent and dirtiest piece of coal having ash content of 15.0 percent.

At 1.55 specific gravity the yield of float will be 74.4 percent and such product would assay cumulative float ash of 8.3 percent and dirtiest piece of coal would have ash of 17.2 percent. The highest yield of 77.0 percent of float has ash content of 9.6 percent and dirtiest piece ash of 19.4 percent. These readings are summarized in the Table 54 below. At the yield of float of 75.0 percent corresponding to the sink or reject or 25.0 percent the sink will assay 19.94 percent ash from the cumulative sink curve. The reject is usable in coal-fired thermal power generation.

Since the ultimate aim of this project is to prepare metallurgical coke from Lafia coal it was included all along the research investigations to verify coking properties of the coal both as run-of-mine coal and as washed concentrate. In keeping with this approach the crucible swelling index of the floated fractions was determined using the swelling index furnace (Figure 27). The results obtained are depicted in Plates 19 and 20 as well as in Figure 32. In Plate 19 the determination was in progress. In Plate 20 the coke buttons obtained from the determination show improvements in the caking quality of the coal float fractions up to a value higher than crucible swelling number 8. In Figure 32 the correlation between the cleanness of the coal and its caking property was shown graphically.

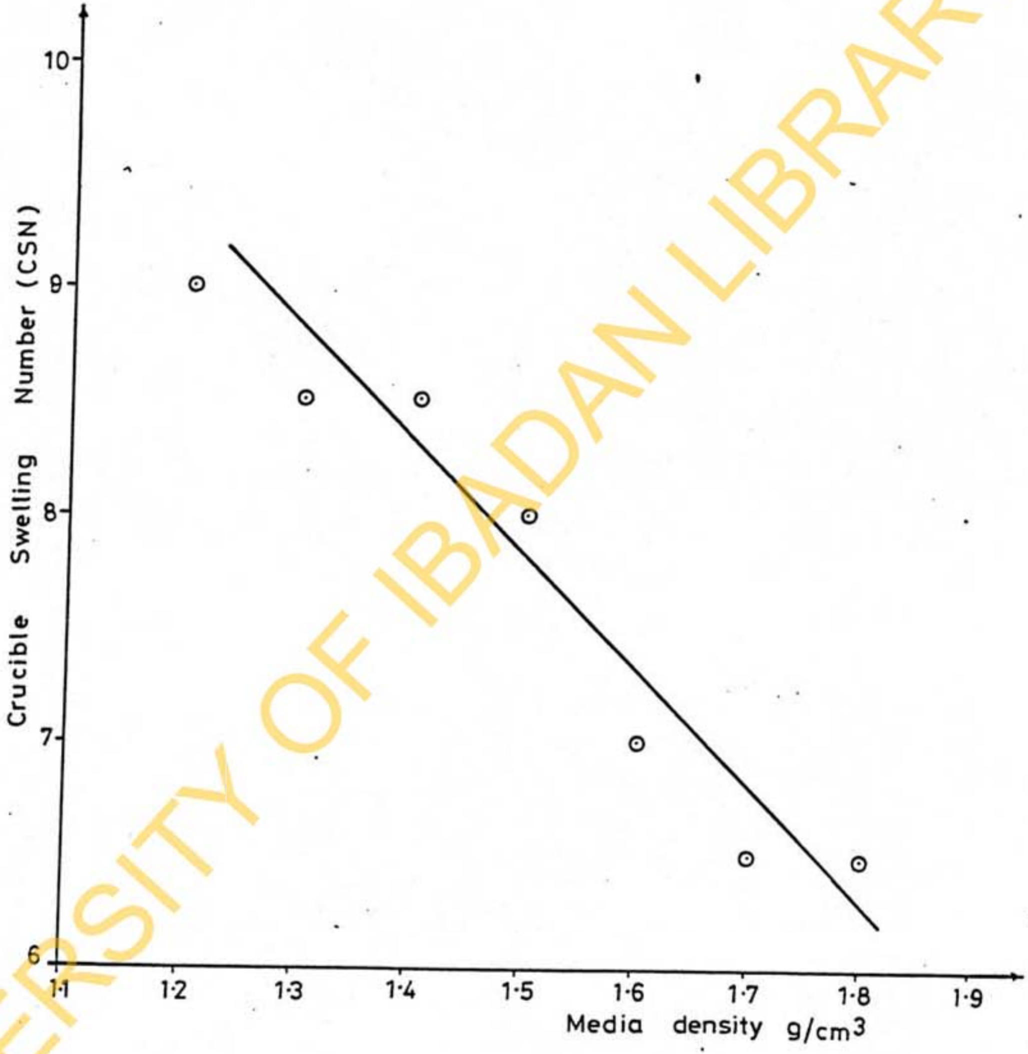

Fig. 32. Coking properties (CSN) of the float fraction seperated from Lafia/Obi coal using Heavy Media Seperation Method (HMS).

Table 54: Interpretation of experimental washability curves for Lafia Coal

Specific Gravity	% Distribution	of Yield of Float	% Ash of Cumulative Float	% Ash of Dirtiest Piece
1.45	21.0	63.0	7.0	14.0
1.50	15.0	69.0	8.0	15.0
1.55	10.0	74.4	8.30	17.2
1.60	8.0	75.0	8.5	16.5
1.65	10.0	77.0	9.60	19.4

Washing has the positive effect on the coking property as observed on the improved crucible swelling number 6 and 7 of the non-volatile residue left in the crucible after the determination of volatile matter content. The swelling number of untreated coal is below 6.

Sulphur reduction was low generally, through all the sizes treated, ranging from the unwashed coal total sulphur value of 2.80 percent to 1.16 or about 58.57 percent. This level of sulphur reduction compares well with results obtained for similar coals with difficult washability.

For the nine runs the average cumulative float yield was 74.6 percent. The yield is fair especially when we consider that the density (of 1.50 to 1.60) of separation applied in these experiments was the same as is used in industrial coal washery. The coal reject at this S.G. cut-off point is usable for thermal coal combustion.

5.5. CONCLUSION

1. From the results of the washability test it is now known that Nigerian Lafia coal can be washed using some physical means to achieve substantial desulphurization.

2. At the particle size minus 12mm by 1mm Lafia coal is too coarse for effective desulphurization and ash removal washery operation.

3. At the finer particle size minus 6.7mm by 0 and a cut-off specific gravity of between 1.50 and 1.60 the subsection as well as the yield of the total coal sample was 75.0% with 57.86% desulphurization and 42.46% ash removal in Nigerian Lafia coal.

4. At minus 60 mesh of grind and a cut-off specific gravity of 1.202 the float cumulative fractional yield of 7.03% contained the lowest float cumulative ash of fractions of 5.22%.

5. The characteristic curve (Figure 29) showed that Lafia coal seam 12 could be washed between specific gravities 1.40 and 1.65 judging from the fact that between these two specific gravity points that the curve is flatter than the remaining part of its course.

6. From the ±0.1 specific gravity distribution curve it was observed that at specific gravity 1.60 Lafia coal can be washed though with some but moderate difficulty and required efficiency process to yield high tonnages under good operation.

7. At 1.50 specific gravity Lafia coal yielded 69 percent with over-all ash of 8.0 percent in the float-and-sink test.

Since the wheel is not to be re-invented, a straight-forward approach was made to investigate four methods of desulphurization, namely,

(i) Washability test
(ii) Wet Concentrating table
(iii) Froth flotation
(iv) Heavy Media Separation

The washability test characteristic curves showed that Lafia seam 12 could be washed between specific gravities 1.40 and 1.65 judging from the fact that it is between these two specific gravity points that the curve is flatter than the remaining part of its course.

The ±0.1 specific gravity distribution curve showed that the percent distribution at specific gravities 1.45, 1.50, 1.55, 1.60 and 1.65 are respectively 21.0, 15.0, 10.0, 8.0 and 10.0. By referring to Bird's Table constants which has more or less universal acceptability and application on coal washability we obtain the interpretation that at specific gravity 1.60 Lafia coal can be washed though with some but moderate difficulty and requires efficient process to yield high tonnages under good operations.

At the cut off point specific gravity of between 1.50 and 1.60 for coal particle size minus 6.7 plus 3.35mm sulphur in the original coal of 2.8% went down to 1.14% subsequent to washing. Desulphurization achieved was thus 59.3%. Ash reduction achieved was 39.1% or from 32.5% ash content in unwashed coal to 19.8% after the coal had been washed. The sub-section (minus 6.7 by plus 3.35mm) yield of coal concentrate was 81.6% which was 25.8% of the total crushed coal of minus 6.7mm by 0.

At the cut off specific gravity of 1.60 the yield of coal particle size of minus 60 mesh (B.S.) will be 79.0 percent with float cumulative ash of 9.0 percent and the dirtiest piece of coal in the float will assay 18.58 percent ash content. If the cut off point is taken at 1.50 specific gravity the yield will be 69 percent with over-all ash of 7.6 percent and the dirtiest piece of coal will have ash content of 15.0 percent. The reject of 21 percent corresponding to clean coal yield of 79.0 percent has ash content of 19.94 percent as read from the cumulative sink curve. Such a reject is usable in coal-fired thermal power generation. At the same time the 79.0% clean coal yield which has ash of 9.0 percent could, on account of the ash now being low enough, be conveniently used in metallurgical coke preparation.

The wet concentrating tabling of Lafia coal achieved from 51% to 69% desulphurization and 50% ash reduction with 86% clean coal yield of coal particle size 6mm by 0.

The froth flotation method of desulphurization achieved a mean of 61% sulphur removal and 73% ash reduction with a mean clean coal yield of 48.4% of coal particle size minus 60 mesh.

Heavy media separation (HMS) of coal of particle size under 0.5mm did not show consistent appreciable gangue and sulphur rejection. Total sulphur of 3.02% reduced to 2.52% which was sulphur rejection of only 16.5% for seam 13 coal sample. In another run the sulphur rejection was 34.9% or from 2.84% to 1.85% for coal particle size minus 0.5mm by 0 sample.

However, in some other instances there was no sulphur rejection. Total sulphur in raw coal was 1.33 percent and the concentrate showed 2.16 percent of total sulphur which implied 62.4 percent worsened position of sulphur content in the particular experiment. From these results it can be concluded that the Heavy Media Separation (HMS) method did not work well on desulphurization of finely ground Lafia coal.

CHAPTER SIX

EXPERIMENTAL

DESULPHURIZATION STUDIES ON NIGERIAN LAFIA COAL, USING COAL PETROLOGY AND SCANNING ELECTRON MICROSCOPE (SEM) TECHNIQUES

6.1. Introduction

Reflected white light microscopy was used jointly with scanning electron microscopy (SEM) as analytical tools to follow the progress of coal desulphurization. Interestingly enough, while other methods of analyses usually give the average values for the whole of the coal represented in the sample, coal petrology makes it possible for one to see in details all the minutest structural particles, relationships and varieties. Judgements arrived at subsequent to microscopy studies were therefore well based on the sum total of the structural components and their properties.

Using coal petrography, the macroscopic (enlarged) view of the types of coal building block were examined before going on to the microscopic (eye-needing-aid-to-see) components.

Nigerian coals were seen as existing in banded or layered form. This is like a big loaf of sliced bread, for an analogy, where the slices vary in structure. Like other hard coals (1) Nigerian coals showed layers of bright bands (vitrain), semi-bright bands (clarain), dull bands (durain), and mineral charcoal (fusain).

To be able to see the microscopic components of the coal we had to prepare polished samples. Some samples were polished as the coal was without breaking it down. Another method used, however, required that the coal sample be broken down to particle sizes between 850 and 250 microns. Both methods were adopted to gain the advantages of seeing the coal components in situ and in their scattered but as fully liberated particulates. The methods of preparing polished coal samples for microscopic examination will now be outlined.

6.2. Preparation of Coal Polished Surface

The methods for both the crushed and ground sample as well as for lump piece of coal were handled are now described.

6.2.1. Method for Particulate Sample

A representative sample of the coal was taken. The coal was pulverized to 100 percent minus 200 mesh (British Standard) by stage-wise crushing and sieving to avoid generating too much fines below 100 microns. 5 grammes of the pulverized coal was abstracted and mixed with an embedding material which was an epoxy resin. Carnauba wax, araldite and Schneiderhohn mixture were, on separate occasions, used as embedding chemicals. Synthetic resins have proved in recent years to be particularly suitable because their polishing characteristics differ only slightly from those of coal and because they are inert to various immersion liquids. Two parts of coal to three of resin were mixed. Paper box of 25mm cube was used to cast the coal-resin mixture which was then left for twenty-four hours to solidify before grinding and polishing.

6.2.2. Method for Lump Coal Specimens

The coal blocks were impregnated with araldite, Schneiderhohn mixture or Carnauba wax, all of which had good penetration. They were left to harden for twenty four hours before taking them up for grinding and polishing.

In between one polishing grade of alumina and another, the coal block was thoroughly rinsed with water jet from the top.

A well polished coal block was relief-free and reflected very much like a mirror. The sample was then allowed to dry in an oven at about 30°C for two hours and then transferred to a desiccator to cool to room temperature, before it was studied under the microscope.

6.3. MICROSCOPY EXAMINATION OF LAFIA COAL

Macerals which by definition are microscopic constituents of coals analogous to minerals of rocks were identified and classified on the basis of their morphology, source material, colour or level of reflectivity, and nature of formation.

6.3.1. Instrumentation

E. Leitz MPV2 microscope equipped with vertical illuminator, oil immersion objective magnification x50 and eye piece x10 power was used for maceral analysis and reflectance measurement. The light source was a tunsten incandescent lamp with a power output of 100W.

The microscope was equipped with a rotating object stage and an attachable mechanical stage for point counting. The mechanical stage was capable of advancing the specimen laterally by equal steps of such length that only a negligibly small proportion of the particles examined received more than one count on the same particle. The step length is equal to half the maximum particle diameter, i.e. 0.5mm for samples with a standard top particle size of 1mm. The stage also permits a similar stepped advance in the perpendicular direction. The lateral movement was actuated by the counter mechanism, whereas the perpendicular movement was satisfactorily performed manually.

Swift point counter capable of registering the counts in each category and the grand total of petrographic components was used.

Sample-mounting equipment comprised glass slides, modelling clay, and levelling press.

Immersion oil that is non-drying, and non-corrosive with a refractive index of 1.5180 ± 0.0004 at 23°C at a wavelength of 546nm and with a temperature coefficient (-dn/dt) of less than 0.0005/k was used for both maceral analyses and measurement of maximum reflectance.

6.3.2. Procedure For Maceral Analysis

The microscope was well focused to give the best image of the object under study. This is technically known as Kohler illumination.

The particulate block was mounted on glass-slide using plasticine and a levelling press. It was then placed on the microscope stage held by two pins. Two drops of immersion oil were placed on the surface of the block. The sample was focused and the images of the different macerals (organic building and identifiable components) identified and their number and/or frequency of occurrence counted and recorded to give a statistical analysis of the macerals. During the counting of the macerals, the images of the medium used to mount the particulate coal or used to hold the cracked coal-piece together were ignored.

There were some cases in which materials lay in boundaries between macerals or between maceral and mounting medium. In such cases, examination was made on the material lying immediately adjacent to the cross-line intersection in the top right, bottom right, bottom left, and top left quadrants. The first of these not having a boundary in it was taken and counted.

The block was advanced by one step in the left right direction. As the specimen was traversed, counting of macerals and carbominerites continued. At the end of a traverse, the block was advanced by a step of at least equal length in the perpendicular direction to start the next parallel traverse. The step length chosen was 0.5mm and it was kept constant to ensure a uniform counting of points over the surface of the block. A total of 500 counts were made on each coal particulate block. The macerals and micrographs of Okaba and Enugu polished specimens are shown in plates 24 and 25 respectively.

6.4. METHOD OF DETERMINING MICROSCOPICALLY THE REFLECTANCE OF VITRINITE

6.4.1. Introduction

Reflectance may be defined as the percentage of the normal incident light reflected from a polished surface. Two types of reflectance are measured experimentally, namely, Maximum Reflectance and Random Reflectance. The Maximum Reflectance is the highest value of reflectance obtained when any polished section of a particle or lump of coal is rotated in its own plane in linearly polarized light.

Random Reflectance on the other hand is the reflectance of any polished section of a particle or a lump of coal when determined in unpolarized light without rotation of the specimen.

One property that can be used as an indicator of rank, independent of the petrographic composition, is the reflectance of the vitrinite in the coal, which increase progressively with increasing degree of coalification. The reflectance of the sub-macerals of vitrinite differ even in a single coal seam and therefore the value of the reflectance obtained depends on the choice of the sub-macerals used for measurement. Reflectance measurements are made on one or more of the sub-macerals of vitrinite and in reporting the results it is necessary to specify on which sub-macerals the measurements were made. Consequently, a vital step in the measurement of vitrinite reflectance is the identification of vitrinite and its various sub-macerals. The reflectance value obtained also depends on whether maximum or random reflectance measurements are made, so that the type of measurement has to be specified.

6.4.2. Principle

The light with a wavelength of 546nm, reflected at near normal incidence from a specified area of well-polished vitrinite, measured under oil immersion using a photomultiplier is compared with light reflected under identical conditions from a number of standards of known reflectance. Because different vitrinite particles within a single coal seam invariably differ slightly from one another in optical properties, enough readings on different particles are taken to ensure that the results are representative.

6.4.3. Instrumentation and Materials

Leitz MPV2 binocular polarizing reflected light microscope with photometer is described under methods of maceral analysis, was used for reflectance measurement.

Immersion oil, as described under maceral analysis experiment, was used.

Calibration standards used were of optical glasses with refractive index of 1.70-1.97 and of reflectance in immersion oil from 0.32 to 1.66 percent.

The other reflectance standard used was Yttrium aluminium garnet (YAG) of refractive index 1.84 and reflectance in oil of 0.92 percent.

6.4.4. Procedure

For measurement of maximum reflectance, the polarizer was set to 45° to use a Berek prism incorporated in the microscope. The microscope was adjusted for Kohler illumination. The microscope was next aligned. To do this, the image of the field stop was centred and focused. The objective was centred with respect to the axis of rotation of the stage and the centre of the measuring aperture was adjusted to be conincident with the cross lines.

The standard samples were used to calibrate the microscope settings. Immediately after the calibration, the levelled polished block was put on the mechanical stage. Oil was dropped on the polished surface. The specimen was brought into focus at one of its corners. The specimen was moved slightly using the mechanical stage until cross-lines are focused on a suitable area of vitrinite. It was ensured that the measuring area contained no cracks, polishing defects, mineral inclusions or relief effects and is away from maceral boundaries.

The light passed to the photomultiplier and the stage was rotated through 360° at a rate not exceeding 10 rpm. The highest relfectance reading obtained during the stage rotation was recorded. The specimen was moved a step of 0.5mm in the X and Y directions across the polished coal block surface to ensure that the measurements were evenly distributed.

At 15 minutes intervals the calibration of the apparatus was re-checked using the standard nearest in reflectance to that of the highest reflecting vitrinite in the specimen. Allowed difference in the reading of the standard was 0.02% and a higher value resulted in discarding the last set of readings. The instrument was then recalibrated using the full range of standards and the sample readings repeated. The results are given in Table 60.

6.5. DISCUSSIONS OF RESULTS

From their origin in the coal-forming plants, microscopic entities were identified by their form and appearance. Some of the macerals had their origin from woody and cortical tissues.

Examples of these are vitrinite, fusinite and semi-fusinite. Some other macerals had origins other than woody tissues and these are exemplified by the exinite macerals of sporinite, cutinite, resinite, alginite and sclerotinite. Yet there are some whose origin has not been traced to a specific vegetative tissue like micrinite. Some of these will now be further described in details as aids used to identify them.

Vitrinite

Vitrinite is the principal coal maceral and primary constituent of bright coal. A polished sample sometimes presents a more or less clear picture of the structure of the woody tissue and in that case it is called tellinite (Plate 8). When structureless, it is called collinite (Plate 9). Collinite is the colloidal medium which dominates in all bright coals. Viewed under reflected light, vitrinite is gray to yellowish white, depending on the concentration [6].

Fusinite (inertinite)

Fusinite is the maceral found in fossil charcoal (Plate 10). It always shows a cellular structure, namely carbonified cell-walls and hollow lumens. Sometimes the cell-walls are found to be crushed (and it is then described as "Bogenstruktur", see Plate 11). Polished fusinite shows strong reflectivity. In physical appearance, fusinite is hard but it is very friable.

Semi-Fusinite

Semi-fusinite is the transition stage from vitrinite to fusinite (Plate 12). Its cellular structure is not always easy to recognize. Its reflective power is intermediate between that of fusinite and vitrinite.

Sporinite

Sporinite is the fossil remains of spore exines, flattened parallel to the stratification. Normally the interior of the spore can be recognized only as a very thin band. In polished coal sample, sporinite has a lower reflectivity than vitrinite. In high rank coals, sporinite is hardly distinguishable from vitrinite, differentiation being possible only through use of polarized light.

Spores can be sub-divided into mega- or macro-spores (generally 200 - 500 microns) and microspores (smaller than 200 microns (Plate 13).

Cutinite

Cutinite is that type of exinite that has been formed from cuticles. It appears as more or less narrow bands frequently having one serrated edge. In reflected light, the colour is identical to that of sporinite and it too has low reflectivity (Plate 14).

Resinite

Resinite is the fossil remains of plant resins and waxes. This maceral occurs in shape of round and oval bodies and as inclusions in structureless vitrinite, collinite and as a filling of cell lumens in tellinite (Plate 15). It has low reflectivity.

Alginite

Alginite is the remains of algal bodies. It is not found in normal humic coals. In coals of low rank, alginite displays the weakest reflectance and the strongest fluorescence of all macerals. On polished surfaces, its colour is black to dark grey (Plate 16).

Sclerotinite

The term sclerotinite is used to describe all strongly reflecting fungal remains such as hyphae, mycelia, plectenchyme, spores and sclerotia which are common in tertiary coals (Plate 17).

Micrinite

Micrinite is structureless. It is a major component of dull coal. It possesses high reflectivity which comes between that of vitrinite and fusinite. Micrinite occurs in two varieties, namely, granular micrinite (Plate 18) (in dull coal and as inclusions in bright coal), and as massive micrinite (as the principal component of durain).

6.5.1. Identification of Mineral Inclusions in Nigerian Lafia Coal

Untreated Lafia coal piece was polished and then examined under the white light microscope (Plate 2). Pyrites were seen as brightly shining yellowish-white deposited in a netted form on the cell walls of cellular tissue of the coal-forming plant. Carbonate mineral (siderite) was seen in cleats and fissures of Lafia coal-piece (Plate 3).

In appearance the carbonate mineral inclusion in coal has a muddy dark greyish appearance and lacked plant-like appearance which characterised coal organic matter.

6.5.1.1. Coal-Mineral Association

Under the white light microscope mineral associations with coal macerals were distinguishable. Associations of coal and minerals having densities varying from 1.5 to 2.0g/ml are called carbominerites. Typical cases are now given in Table 55.

Table 55: Coal-mineral Association

Coal intergrown with a certain mineral or a mineral group	Composition
Carbargilite	Coal + 20-60% by volume of clay minerals
Carbopyrite	Coal + 5-20% by volume of sulphide minerals
Carbankerite	Coal + 20-60% by volume of carbonate minerals
Carbosilicite	Coal + 20-60% by volume of quartz
Carbopolyminerite	Coal + 20-60% by volume of various minerals

6.5.1.2. Measurement of Size of Pyrite Crystals in Lafia Coal

Using a graticule with its scale in microns the diameter of crystals, patches, massive and framboids (pine-apple shaped) of pyrites were measured (Tables 56 and 57).

As shown in Table 57, single crystals of pyrites predominate in the distribution of the different forms followed by the framboids and the massive. Patches were the fewest of the forms of pyrite. Furthermore, in Table 58 about 85 percent of the crystals of pyrite measure less than 75 microns (minus 200 mesh) while about 60 percent measure less than 45 microns.

From the measurement of the crystal sizes, it was deduced that for good pyrite liberation during coal desulphurization experiments, it would be necessary to crush the coal and grind it below minus 200 mesh particle size. In many of the experiments the fine size coal was used but in others coarser sizes were used for two or so reasons. Firstly, to see what desulphurization would be achieved using sizes coarser than minus 200 mesh size, and secondly because the cost of pelletizing or briquetting will be less if it became necessary to pelletize or briquette the desulphurized coal before carbonizing it.

Table 56: Distribution of Pyrite Among Various Forms in Lafia Coal

Sa/No	Single Crystal	Framboids	Patches	Massive
1	63.0	15.0	7.0	15.0
2	61.5	14.6	8.4	15.5
3	64.0	14.1	8.7	13.2
4	63.4	15.5	9.1	12.0
5	62.4	14.8	9.8	13.0
6	64.0	13.0	9.8	13.2
7	63.0	14.5	8.5	14.0
8	66.5	14.7	8.0	10.8
9	64.0	15.0	8.5	12.5
10	64.2	14.8	8.5	12.5

Table 57: Size of pyrite crystals in Lafia coal (as per cent)

Sal/No.	45 µm	75 µm
1	60	78
2	60	85
3	64	88
4	64	88
5	64	86
6	60	85
7	64	88
8	63	85
9	63	88
10	65	88

6.5.1.3. Monitoring the Progress of Lafia Coal Desulphurization Using Coal Petrology

Coal blocks were prepared and polished off, firstly, the untreated but finely crushed coal, secondly, of the first stage coal flotation and lastly of the (reverse froth floated) second stage clean coal concentrate. Plate 4 shows the appearance of the untreated run-of-mine Lafia coal under the microscope at 500 magnification. Pyrite crystals, nodules, framboids and various other forms of it show clearly along with clay and carbonate mineral inclusions in Lafia coal. Plate 5 shows the first stage coal flotation with much less pyrites than was seen in the raw coal. Already much of the pyrite had been floated off the coal. Plate 6 showed the clean coal concentrate which had but very few pyrite crystals left entrapped within the coal particles. Classical chemical analysis later confirmed the progress of desulphurization observed under the microscope.

Table 58: Petrographic Analyses of Nigerian Coals

Sample Number	Maceral groups and mineral matter (% volume composition)					
	Vitrinite	Exinite	Inertinite	Pyrite	Shale	Other minerals
1	76.0	Nil	2.8	2.0	11.3	8.0
2	87.0	Nil	4.4	2.0	4.0	3.0
3	88.0	Nil	2.0	1.0	2.0	2.0
4	55.0	16.0	21.0	Nil	3.0	6.0
5	68.0	9.0	15.0	Nil	7.0	1.0

Legend

Sample No. 1: Nigerian Lafia coal, original feed of minus 30 mesh.

Sample No. 2: First stage froth flotation using 1.3 kg/tonne centrifroth "M" concentrate, Nigerian Lafia coal minus 30 mesh.

Sample No. 3: Second stage froth flotation concentrate coal using 150 g/tonne Aenodepressant 633, 200 g/tonne isopropyl xanthate, no frother, minus 30 mesh feed.

Sample No. 4: Okaba coal.

Sample No. 5: Enugu Onyeama coal.

6.5.1.4. Maceral Analysis

Table 58 shows the result of maceral analysis of Nigerian coals. The coal organic macerals as well as the mineral inclusions were all identified and their occurrence counted giving the data in Table 58. Sample No.1 which was the untreated coal-feed had the highest values of mineral matter, shale and pyrites which were on volume basis 8.0%, 11.3% and 2.0% respectively. Sample No.2 which represented the first stage froth flotation of Lafia coal the concentrate showed an increased number of vitrinites from 76.0% in the raw coal to 87.0% in the float. The value of inertinite maceral went up from 2.8% in the raw coal to 4.4% in the first float. The counts obtained of the shale and other mineral matter went down respectively from 11.3% and 8.0% to 4.0% and 3.0%. Pyrite mineral did not reduce in the first float with its value remaining as 2.0% as it was in the raw (untreated) coal. Sample No.3 was the second (reversed) float coal concentrate and it showed improved value of 88.0% for the main coal maceral vitrinite. The value of inertinite in the second float went down to 2.0% from 4.4% in the first float. Shale also reduced to 2.0% in the second float.

Pyrite mineral reduced to 1.0% in the second (reversed) float from its initial value of 2.02% thus indicating roughly a 50% desulphurization achieved. Other mineral matter registered 2.0% in the final float.

Sample No.4 which was Okaba coal had low value for vitrinite (55.0%). Plates 24 and 25 are photomicrographs of Okaba and Enugu polished, particulate coal blocks.

6.5.1.5. Reflectance Measurements

The vitrinite reflectance distribution of Nigerian Lafia, Enugu and Okaba coals are given in Table 59. The mean maximum reflectance (in oil) of Lafia coal was found to be 1.14 percent which value places the coal in between the class of High Volatile A bituminous coal and the Medium Volatile bituminous coking coal. The mean maximum reflectance in oil of Enugu (Onyeama) and Okaba coals were found to be 0.5 and 0.44 percent respectively. These values place the coals in High Volatile B and C bituminous coals for Enugu and Okaba respectively.

Table 59: Reflectance Analysis of Nigerian Coals

Distribution/ Sample	Reflectance Class																				Av.Refl.(oil) maximum random	Ro max.
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20		
fia/Obi										19	73	8										1.14
ugu			4	47	46	3																0.50
aba			17	81	2																	0.44

N.B.: A reflectance class covers a range of 0.1% reflectance e.g. class 8 covers 0.80% and 0.89% reflectance.

6.5.1.6. Scanning Electron Microscopy (SEM)

Under the Scanning Electron Microscope, a larger magnification of the coal structure was observed.

Higher magnifications made both organic coal structure and quartz inclusions visible. The microscope detected silicon in coal (Figure 33).

6.6. CONCLUSION

It has been possible to follow up the progress of desulphurization of Lafia coal using the white light microscope. The distribution of sulphur in the forms of crystals, massive pyrites, framboids, were identified. The measurement of the sizes of the various pyrite form served as a good guide on how fine Lafia coal need be ground if the pyrites mineral was to be liberated for its effective removal.

Other inorganic mineral impurity varieties in Lafia coal were indentified. among them were carbonate and silicate minerals. Such identification served to make one aware of the need to wash off the alkali metals which have deleterious effect on crude steel product of the blast furnace.

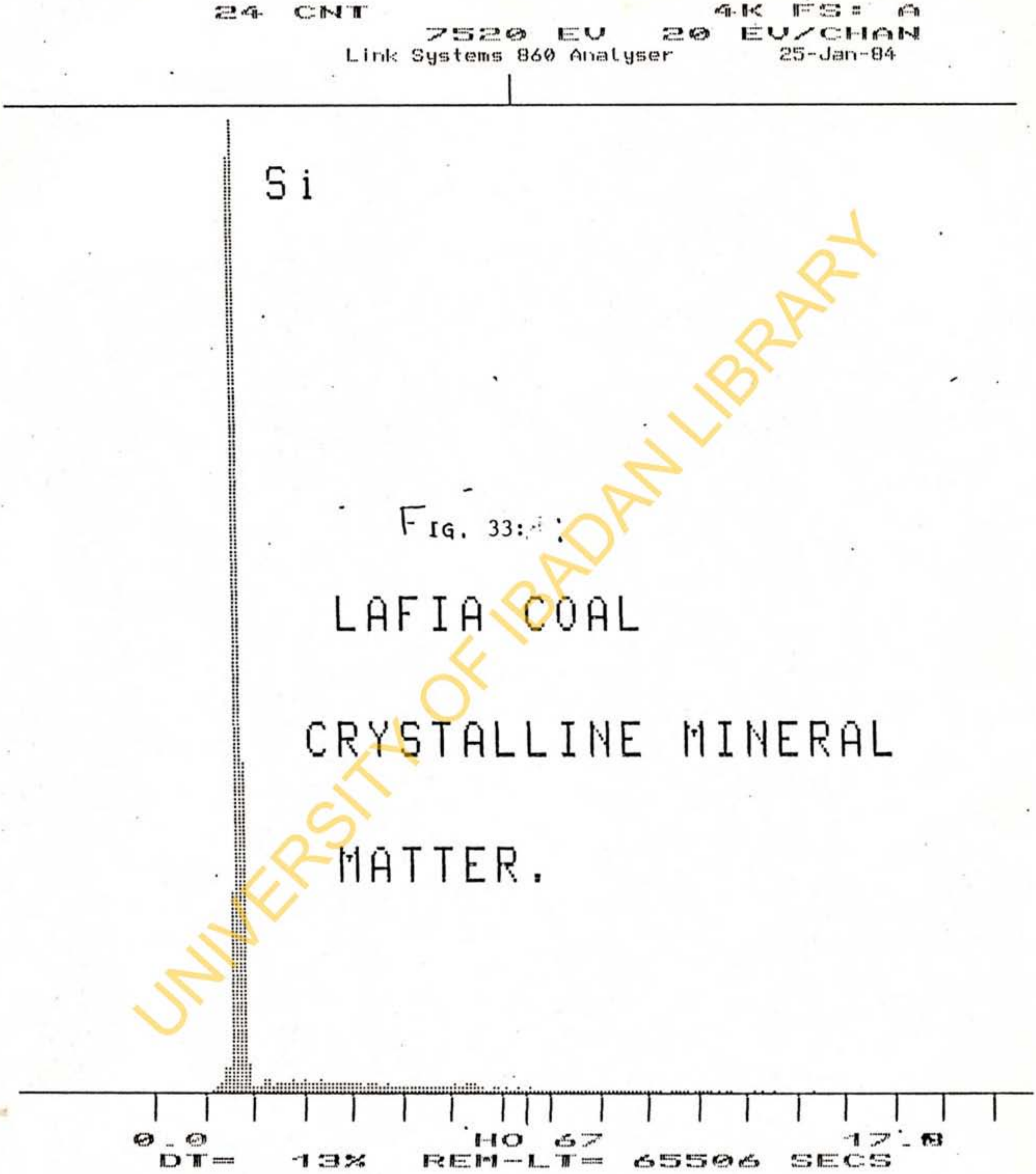

FIG. 33:4 :

LAFIA COAL

CRYSTALLINE MINERAL

MATTER.

QUARTZ

The study of the rock structure of the coal by which the macerals were identified is helpful in determining the quantity of the coals which could go into a coking blend. The measurement of reflectivity has made it possible in a standard way to follow the maturation of the coal deposits. That is, it became clear from measurements of the reflectance of vitrinite maceral that Lafia coal is much more mature than either Enugu or Okaba coals while Enugu is also more mature than Okaba coal.

CHAPTER SEVEN

DESULPHURIZATION OF LAFIA COAL BY FROTH FLOTATION

7.1. INTRODUCTION

Froth flotation which has found wide application in concentrating sulphides like cinnabar(HgS), Zinc blende (ZnS) has been resorted to in another effort at investigating feasible methods of desulphurizing Nigerian Lafia coal since the inorganic sulphur has been established to occur mainly in the form of pyrites, (FeS_2). Flotation agents such as Xanthates have been employed as pyrite concentrators because of their popularity as effective sulphide concentrators and because they are available readily and can be synthesized from local raw materials. Frothers which are also flotation agents include some form of alcohols and these could also be got locally. So, in effect of the trials reported in this study giving positive results which would recommend commercial use of flotation method for Lafia coal desulphurization the reagents required would be available readily from local sources.

7.2. Procedure

This procedure involves first stage and second stage floatation operations respectively.

EXPERIMENTAL

In the first flotation which was a normal one, the desired coal concentrate was froth-floated. Based on available literature information 310 g of coal ground to pass 100 percent through a known mesh sieve size was introduced into water from the tap in a flotation cell plate 22. The water in the flotation cell was 5000cm^3. This slurry thus produced was 6.2% coal-in-water slurry. 0.25g of centifroth "M" (equivalent to 822g of the frother to one tonne of coal) was pipetted using the microsyringe and dropped into the coal-water slurry. The flotation cell used in this first flotation was that manufactured by Denver of Great Britain. The volume of the cell was 5430cm^3. However, it was 5000cm^3 of its internal capacity that was filled with the slurry. The slurry was then conditioned by stirring it mechanically for 15 minutes at 1500 r.p.m to thoroughly wet the coal.

The froth floats of coal was then scraped into a receiving pan using a fine sieve scoop. Collection of froth floats was done for three minutes. This gave clean coal 1.

The underflow of shaly pyritised coal was filtered on a Buchner funnel with no 540 whatman filter paper using a water-pump and this was the reject 1.

The froth concentrate from the first stage flotation, that is clean coal 1,was re-pulped immediately as it was filtered on a white piece of satin cloth, without drying off the water. Water was added to make up the volume of 5000cm^3 in the cell. The coal-water slurry in the second flotation was thus of lower concentration than the 6.2% that was used in the first stage flotation.

The re-pulped slurry was dispersed by setting the impeller speed to 1500 rpm for two minutes. The amount of Aero Depressant 633 added was equivalent to 300g/tonne of coal. Also the xanthate collecter added was equivalent to 400g/tonne of coal. No frother was added to the second stage flotation which is called "reverse flotation" as the coal was depressed into the underflow and the unwanted pyrite contaminant was floated off. The air in-let was opened and frothing commenced.

In this second stage, reverse flotation, the liberated pyrite grains and coal-pyrite-associated particles (that is, carbo-pyrite) were froth-floated. Scrapping off and collection of the froth floats was done for three minutes. At the end of the three minutes floated material that splashed on the shaft of the impeller and on the upper edge inside of the flotation cell was scraped off and added to the froth-floated collection of pyrite and this constituted reject 2.

The shaft was rinsed thoroughly into the cell pan. The underflow in this second stage run was the clean coal No.2. Clean coal No.1 was re-pulped wet immediately after collection and so its weight and chemical characteristics were calculated from the analysis results of reject 2 and clean coal 2. The products of the second stage run were filtered using the same filtration equipments described above. They were then dried, weighed and analysed for ash, sulphur, forms of sulphur, volatile matter, calorific value and in some cases coking properties.

Several test runs were carried out for various slurry densities and different sieve mesh sizes, both for double normal flotation and normal/reverse flotation of Lafia coal. The results (average of 3 determinations) obtained are shown in Tables 59, 60, and 61.

7.2.1. Flotation Reagents Used

The flotation reagents used were those found most readily available.

The coal depressant used were Aero Depressant 633, which is a commercial reagent recommended for depressing carbonaceous gangue. It is a carbohydrate substance having a structure similar to the basic unit of the starch molecule.

The sulphydryl collector used to float the pyrite material was isopropyl xanthate. The five-carbon aliphatic group, potassium amyl xanthate which is generally used in those flotation operations requiring the highest degree of collecting power was not available at the time of the test.

The frother employed was the Centifroth 'M' which is a hydro-carbon oil that is devoid of any carcenogenic component. Centifroth "M" is a product of Century Oils Ltd, Hanley, Stoke-On-Trust, England. Methyl-siobutyl carbinol (MIBC) which is the widely used six carbon, branched chain aliphatic alcohol was not available at the time of the test runs.

Kerosine was also used as a frother and report of this is included.

Centifroth 'M' was compared with other frothers like Prism 'N' and Prism 'X' but the former was found to perform satisfactorily and hence its choice in this work.

The pH modifier chosen was sulphuric acid because of its relatively low cost and availability.

The Aero Depressant 633 and the isopropyl xanthate were added to the flotation pulp as 1-percent aqueous solutions, and the sulphuric acid was added as a 1-normal (1-N) solution. The Centifroth 'M' was used undiluted in the state the supplier (Century Oils Ltd) synthesized it.

7.2.2. Apparatus

The apparatus used in froth flotation were as follows:

1. Laboratory flotation machine type D12 (Serial Number BAG 17720) manufactured by Denver Equipment Company, England.
2. Microsyringe (SZR-515-32 OP) a product of Gallenkamp in Kloehn
3. Glass and plastic wares serving as containers
4. Measuring cylinder
5. Sieve scoop
6. Air oven for drying
7. Weighing balance

Table 60: Results of double normal flotation of Lafia coal borehole samples of seam 12
(Coal particle size minus 60 mesh)

Sa/No.	Borehole No.	Flotation Products (% wt.)		Chemical Analyses									
				Clean coal					Reject 1+2				
		Clean coal	Reject	M	Ash (db)	VM (daf)	Total sulphur	Pyritic sulphur	M	Ash (db)	VM (daf)	Total sulphur	Pyrit sulph
1.	8	58.5	41.5	2.22	10.2	27.8	1.66	0.96	3.61	27.3	28.1	1.20	0.65
2.	12	59.1	40.9	2.10	9.85	28.1	1.70	1.40	3.63	28.0	27.9	1.07	0.58
3.	26	60.2	39.8	1.98	10.5	28.2	1.60	1.17	3.46	27.8	28.0	1.00	0.42
4.	38	59.6	40.4	2.15	9.79	28.1	1.44	1.06	4.10	29.0	28.6	1.02	0.52
5.	42	59.8	40.2	1.98	10.20	28.1	0.94	0.44	3.62	27.5	28.1	1.78	1.12
6.	51	60.1	39.9	1.69	9.75	28.6	1.16	0.66	3.53	27.7	28.1	1.14	0.63
7.	66	58.4	41.6	2.36	10.3	27.2	1.56	0.96	3.09	28.2	28.3	1.12	0.54
8.	72	61.1	38.9	2.27	8.8	27.8	1.39	0.79	3.62	29.0	27.2	0.95	0.45
9.	106	60.8	39.2	2.42	10.1	27.7	1.28	0.72	3.79	28.5	27.3	0.80	0.35
10.	Composite (1-9)	61.3	38.7	2.11	10.2	28.4	1.30	0.75	3.50	30.0	28.4	2.20	1.50
11	Untreated composite	-	-	2.30	31.5	28.6	2.84	1.99					
Range, Mean	(1-9) (1-9)	58.4-61.3; 59.89		1.69-2.42; 2.13	9.79-10.30; 9.97	27.2-28.6; 28.0	0.94-1.70; 1.40	0.44-1.40; 0.90					

Table 61: Summary of sulphur removal and ash reduction in Lafia coal by the method of Froth Flotation

Test No	Clean coal total sulphur (% wt)	Coal feed total sulphur (% wt)	Clean coal yield (% wt)	Clean coal ash (% wt)	Coal feed ash (% wt)
1.	1.12	2.83	46.1	7.8	30.1
2.	1.09	2.83	55.5	7.14	30.1
3.	1.02	2.83	54.7	6.90	30.1
4.	1.12	2.82	44.7	8.4	29.9
5.	1.11	2.86	50.4	8.2	30.2
6.	1.02	2.81	62.1	7.5	29.8
7.	1.21	2.84	24.5	9.2	30.7
8.	1.20	2.85	57.0	9.2	32.5
9.	1.16	2.85	58.2	8.8	32.5
10.	1.13	2.80	54.9	8.0	32.5
11.	1.13	2.84	40.1	8.5	29.3
12.	1.12	2.84	41.0	8.2	29.3
13.	1.12	2.84	40.2	8.1	29.3
Mean	1.12	2.83	48.4	8.15	30.48
Desulphurization achieved = 60.9				Ash removal achieved = 73.3%	

TABLE 62: RESULTS OF TWO STAGE FLOTATION (NORMAL AND REVERSE) OF NIGERIAN LAFIA COAL PARTICLE SIZE, MINUS 240 MESH)

Test No	Experimental Conditions	Product: Feed, clean coal & reject	% wt	% Ash	% Total sulphur	% Pyritic sulphur	% Volatile matter
1.	<u>First Stage</u>: 822g/tonne	Clean coal 2	46.1	7.8	1.12	0.57	30.0
	centrifroth "M", slurry concentration 6.2% solid	Reject 2 Clean coal 1	19.9 66.0	17.0 23.1	2.03 1.85	1.48 1.19	28.6 29.6
	<u>Second Stage</u>: 300g/tonne						
	Aero depressant 633; 400g/tonne Isopropyl xanthate	Reject 1	34.0	44.2	1.81	1.23	32.8
	No Frother minus 240 mesh size	Feed	100.0	30.1	2.83	1.96	29.8
2.	<u>First Stage</u>: 822g/tonne	Clean coal 2	55.5	7.14	1.09	0.52	28.7
	centriforth "M", slurry concentration 6.2% solid						
	<u>Second Stage</u>: 500g/tonne Aero depressant 633;	Reject 2	9.6	18.8	1.92	1.28	28.4
	700g/tonne Isopropyl	Clean coal 1	65.1	22.3	1.83	1.14	28.7
	xanthate	Reject 1	34.9	45.0	1.78	1.20	32.2
	No frother, coal particle size minus 240 mesh	Feed	100.0	30.1	2.83	1.94	29.8
3.	<u>First Stage</u>: 822g/tonne	Clean coal 2	54.7	6.90	1.02	0.48	28.4
	centriforth "M", slurry concentration 6.2% solid	Reject 2	10.6	20.0	1.97	1.23	28.3
	<u>Second Stage</u>: 700g/tonne Aero depressant 633	Clean coal 1	65.3	22.5	1.85	1.12	28.4
	1kg/tonne isopropyl xanthate	Reject 1	34.7	43.4	1.81	1.21	32.0
	No frother, coal particle size, minus 240 mesh	Feed	100.0	30.1	1.83	1.94	29.8

TABLE 62 Contd.

Test No	Experimental Conditions	Product: Feed, Clean coal and reject	% wt	% Ash	% Total sulphur	% Pyrite sulphur	% volatile matter
4	First stage: 20g/tonne $Fecl_3$; 822g/tonne centrifroth "M"; slurry concentration 8.0 solids coal particle size minus 240 mesh Second stage: 200g/tonne Aero Depressant 633 375g/tonne Isopropyl xanthate	Clean coal 2	44.7	8.4	1.12	0.60	28.2
		Reject 2	21.4	17.5	2.03	1.09	28.5
		Clean coal 1	66.1	23.5	1.82	1.00	28.2
		Reject 1	33.9	42.6	1.82	0.75	31.8
		Feed	100.0	29.9	2.82	1.92	29.6
5	First stage: 40g/tonne $Fecl_3$ 822g/tonne centrifroth "M" slurry concentrations 8.0% solids; coal particle size minus 240 mesh Second stage: 375g/tonne Aero Depressant 633; 625g/tonne isopropyl xanthate	Clean coal 2	50.4	8.2	1.11	0.61	28.4
		Reject 2	13.3	18.1	2.00	1.04	28.5
		Clean coal 1	63.7	23.0	1.79	0.98	28.2
		Reject 1	36.3	42.9	1.72	0.92	32.1
		Feed	100.0	30.2	2.86	1.96	29.6
6	First stage: 60g/toone $Fecl_3$ 822g/tonne centrifroth "M"; slurry concentration 8.0% solids; coal particle size minus 240 mesh Second stage: 625g/toone Aero depressant 633; 875g/toone Isopropyl xanthate	Clean coal 2	62.1	7.5	1.02	0.52	28.2
		Reject 2	7.9	19.6	1.83	0.85	28.2
		Clean coal 1	70.0	23.3	1.83	0.99	28.4
		Reject 1	30.0	44.9	1.77	0.79	32.0
		Feed	100.0	29.8	2.81	1.93	29.8

TABLE 62 Contd.

Test No	Experimental conditions	Product: Feed Clean coal & reject	% wt	% Ash	% Total sulphur	% Pyrite sulphur	% Volatile matter
7	First stage: 3.2 kg/toone Kerosene (as frother)	Clean coal 2	24.5	9.2	1.21	0.70	28.2
		Reject 2	15.3	13.9	1.93	1.18	28.4
	Second stage: 150g/toone Aero Depressant 633 200g/toone Isopropyl xanthate; 1.6 kg/toone kerosene (as frother), coal particle size minus 60 mesh	Clean coal 1	39.8	18.3	1.78	1.09	28.2
		Reject 1	60.2	38.5	1.93	1.20	32.2
		Feed	100.0	30.7	2.84	1.94	29.8
8	First stage: 1.3 kg/tonne centrifroth "M" slurry concentration 9.0% solids	Clean coal 2	57.0	9.2	1.20	0.64	28.3
		Reject 2	2.2	19.6	2.18	1.56	28.2
	Second stage: 150g/toone Aero depressant 633 200g/tonne Isopropyl Xanthate; No frother PH 6.5-7 coal particle size minus 240 mesh	Clean coal 1	59.2	20.3	1.78	1.09	28.2
		Reject 1	40.8	48.6	1.68	1.02	32.1
		Feed	100.0	32.5	2.85	1.94	29.8
9	First stage: 1.3kg/tonne centrifroth "M", slurry concentration 9.0% solids	Clean coal 2	58.2	8.8	1.16	0.58	28.4
		Reject 2	3.2	20.3	2.13	1.53	28.2
	Second stage: 350g/toone Aero Depressant 633 500g/tonne Isopropyl Xanthate; pH 6.5-7; No frother coal particle size minus 240 mesh	Clean coal 1	61.4	20.6	1.80	1.05	28.2
		Reject 1	38.6	49.6	1.70	1.10	32.1
		Feed 100	100.0	32.5	2.85	1.94	29.8

TABLE 62 contd.

Test No	Experimental conditions	Product: Feed, clean coal and reject	% wt	% Ash	% Total sulphur	% Pyrite sulphur	% Volatile matter
10	First stage: 1.3kg/tonne Aero froth 65, slurry concentration of 9.0%	Clean coal 2	54.9	8.0	1.13	0.54	28.2
		Reject 2	6.5	17.2	2.39	1.63	28.3
	Second stage: 350g/tonne Aero Depressant 633; 250g/tonne Isopropyl xanthate 420g/tonne Aero froth 65; slurry concentration of 5.0% particle size/240 mesh	Clean coal 1	61.4	20.6	1.80	1.80	28.2
		Reject 1	38.6	51.4	1.80	1.20	32.1
		Feed	100.0	32.5	2.80	1.91	29.8
11	First stage: 700g/tonne centrifroth"M",slurry concentration 8.0% solids	Clean coal 2	40.1	8.5	1.13	0.51	28.3
	Second stage: 150g/tonne Aero Depressant 633; 200g/tonne Isopropyl Xanthate No frother coal particle size minus 100 mesh	Reject 2	10.1	17.1	1.95	1.23	28.2
		Clean coal 1	50.2	22.1	1.81	1.18	28.2
		Reject 1	49.8	37.0	1.83	1.20	32.1
		Feed	100.0	29.3	2.84	1.92	29.6
12	First stage: 700g/tonne centifroth "M", slurry concentration 8.0% solids	Clean coal 2	41.0	8.2	1.12	0.55	28.2
	Second stage: 250g/tonne Aero Depressant 633; 350g/toone Isopropyl Xanthate, No frother coal particle size minus 100 mesh	Reject 2	8.4	16.3	1.99	1.28	28.2
		Clean coal 1	49.4	21.2	1.79	1.14	28.3
		Feed	100.0	29.3	2.84	1.92	29.8

TABLE 62 contd.

13	First stage: 700g/tonne centrifroth "M", slurry concentration; 8.0% solids	Clean coal 2	40.2	8.1	1.12	0.88	28.2
		Reject 2	9.6	16.7	2.04	1.20	28.2
	Second stage: 250g/tonne Aero Depressant 633; 500g/tonne Isopropyl xanthate; No frother	Clean coal 1	49.8	21.4	1.82	0.94	28.3
		Reject 1	50.2	37.9	1.86	0.90	32.0
	coal particle size minus 100 mesh	Feed	100.0	29.3	2.84	1.92	29.6

Table 63: Summary of sulphur removal and ash reduction in Lafia coal by the method of Froth Flotation

Test No	Clean coal total sulphur (% wt)	Coal feed total sulphur (% wt)	Clean coal yield (% wt)	Clean coal ash (% wt)	Coal feed ash (% wt)
1.	1.12	2.83	46.1	7.8	30.1
2.	1.09	2.83	55.5	7.14	30.1
3.	1.02	2.83	54.7	6.90	30.1
4.	1.12	2.82	44.7	8.4	29.9
5.	1.11	2.86	50.4	8.2	30.2
6.	1.02	2.81	62.1	7.5	29.8
7.	1.21	2.84	24.5	9.2	30.7
8.	1.20	2.85	57.0	9.2	32.5
9.	1.16	2.85	58.2	8.8	32.5
10.	1.13	2.80	54.9	8.0	32.5
11.	1.13	2.84	40.1	8.5	29.3
12.	1.12	2.84	41.0	8.2	29.3
13.	1.12	2.84	40.2	8.1	29.3
Mean	1.12	2.83	48.4	8.15	30.48
Desulphurization achieved = 60.9				Ash removal achieved = 73.3%	

7.2.3. Results and Discussions

The results of double normal froth flotation as contained in Table 60 showed that the yield of clean coal flotation product varied from 58.4 to 61.3 percent. The total sulphur content of between 0.94 and 1.70 percent of the clean coal was achieved from an untreated coal composite-sample sulphur level of 2.84 percent. This represents a sulphur reduction of between 40 and 67 percent. The ash content was reduced considerably from an untreated coal composite-sample ash content of 31.5% to an average of about 10 percent.

Table 61 gives a summary of sulphur removal and ash reduction in Lafia coal by the method of froth flotation. Table 62 showed results of two stage froth flotation (normal and reverse) of varying coal-particle-sizes using different amounts of flotation agents, varying pulp densities and depressants.

Tests 1 to 3 were carried out on 6.2 percent coal solids in aqueous slurry using 833g/tonne of the frother centifroth "M" in the first stage while varying the coal depressant and the sulphide collector Aero depressant 633 and isopropyl xanthate respectively and using no frother in the second stage.

The coal depressant and sulphide collector were increased from tests 1 to 3 with the objective of producing progressively cleaner coal froth float concentrate. The objective was achieved as the coal flotation products obtained were progressively cleaner with regards to the left over of the ash and sulphur gangues. As the flotation agents increased the ash value of the products decreased giving percentages of 7.8, 7.14 and 6.90. Similarly, the total sulphur content decreased with increasing flotation agents. Values obtained were 1.12, 1.09 and 1.02 percent total sulphur as the coal depressant increased in the order 300g, 500g and 700g per ton of coal and as the xanthate sulphur collector increased in the order 400g, 700g and 1kg per ton of coal respectively.

The first stage normal froth flotation process alone produced in tests 1 to 3 reduction of 35% in total sulphur and 42% pyritic sulphur. The second stage, reverse froth flotation produced a further reduction of sulphur in the first stage, clean coal 1, of 41% total sulphur and 55% pyritic sulphur respectively. The combined normal and reverse flotation processes in tests 1 and 3 produced a 62% total sulphur and 74% pyritic sulphur reduction respectively.

This result compares favourably with pyritic sulphur removal of 68 - 73% froth flotation reported by the Energy Department Laboratory, U.S.A.

In tests 4, 5 and 6 iron III chloride solution was added in increasing amounts in the first stage of the experiments to depress pyrite gangue when coal was floated. In the second stage coal depressant (Aero depressant) and pyrite collector (isopropyl xanthate) were added in increasing amounts in an effort to produce progressively cleaner froth floats (i.e. coal concentrate). The yield of clean coal increased with flotation agents added and came up progressively cleaner. The yields were 44.7, 50.4 and 62.1 percent respectively. Total sulphur contained in the clean coal products were 1.12, 1.11 and 1.02 percent. The use of iron III chloride in the first stage of the double flotation coal cleaning technique gave cleaner coal yield. The sulphur content of the cleaned coal was within the limits that metallurgical coal could tolerate, that is, less than 1% total sulphur.

In test 7 kerosene was employed as the frother in both the first and the second stage of the flotation.

The intention was to see if kerosene would be an effective frother to enable its use since it is relatively easily available locally. From the results it is observed that using kerosene as a frother the coal concentrate produced was relatively clean with 9.2 and 1.21 percent ash and sulphur contents respectively. The clean coal yield of 24.5 percent is, however, too low to recommend the flotation procedure using kerosene as frother, as this would not be cost-effective.

The pH of the aqueous coal slurry (i.e. coal pulp in water) was maintained at between 6.5 and 7. The froth floats yield of 57 and 58.2 percent in tests 8 and 9 contained 9.2 and 8.8 ash and 1.20 and 1.16 percent total sulphur respectively. The pH control has thus proved beneficial towards the desulphurization process.

From test No.10 it was observed that when Aero-froth 65 which was another flotation agent was used it produced clean coal with ash 8.0 and total sulphur of 1.13 percent. The clean coal yield was 54.9 percent.

The results of tests 11, 12 and 13 indicate that coal particle size minus 100 mesh will give clean coal yield of about 40 percent containing ash and total sulphur of 8.2 and 1.12 percent respectively.

From the results it is apparent that double froth flotation of finely crushed coal particles (below minus 100 mesh) yields clean coal of ash content about 8 percent and total sulphur of about 1.12 percent. For example, in test 13 ash reduction was from 29.3 to 8.1 percent or 72.4 percent reduction and total sulphur reduction was from 2.84 to 1.12 or 60.6 percent.

Commercial production of Lafia coal for metallurgical coke preparation can become a reality using double flotation as a means of desulphurization.

7.3. CONCLUSION

The Froth flotation method has achieved 60.9% desulphurization and 73.3% ash removal alongside with 48.4% clean coal product yield of Lafia coal seam 12. Double normal flotation as well as double flotation with reverse in the second stage gave clean coal with over-all sulphur content of 1.12% and ash content 8.15%. It is known from the results of the analytical characterization of Lafia coal described in Chapter Four that organic sulphur in the coal reaches 0.8 to 0.9% and organic sulphur is not removed from coal organic matrix by physical desulphurization.

It is also known from microscopic examination of the coal improvement process described in Chapter Six that some pyritic sulphur are so firmly embedded in the organic coal matrix that the pyrite crystals cannot all be removed. Thus the resultant 60.9% desulphurization achieved (Table 63) is within the range achieved in washing coals of other lands that have similarly high sulphur.

CHAPTER EIGHT

WET TABLE CONCENTRATION OF LAFIA COAL

8.1. Introduction

This section outlines the theory of tabling and some of the factors that enter into tabling action, a description of the Deister Super-Duty diagonal-deck coal table and a discussion of the results of tabling various fine sizes of coals.

Although tables have been used for cleaning coal as coarse as 50 mm and favourable results reported, this research work experiments will be limited to consideration of cleaning coal fines from minus 25 mm down. This limit of minus 25 mm size coal is advised from results of the washability tests reported in Chapter 5 where the fine size dissemination of pyrite and shales in Lafia coal is apparent and also from the limitation imposed by the laboratory size table available.

Although it is well known that the performance of tables is even superior under classified or closely sized feed conditions, this work is concerned with unclassified feeds since in commercial practice where large quantities of coal are treated, the advantage gained in quick operation counterbalances few percentages in yield desirable from coal sizing before cleaning.

8.1.1. Explanation of Tabling Process

Basically the separation of different minerals on a concentrating table are the result of the following factors:

(1) specific gravity of the particles
(2) size of particles
(3) shape of particles
(4) stratification (or consolidation trickling)
and
(5) hindered settling

The effective use of these factors may be controlled and aided by the following table adjustments.

(1) differential action of the drive or head motion
(2) speed of motion
(3) length of stroke
(4) side tilt
(5) end elevation
(6) size of deck area

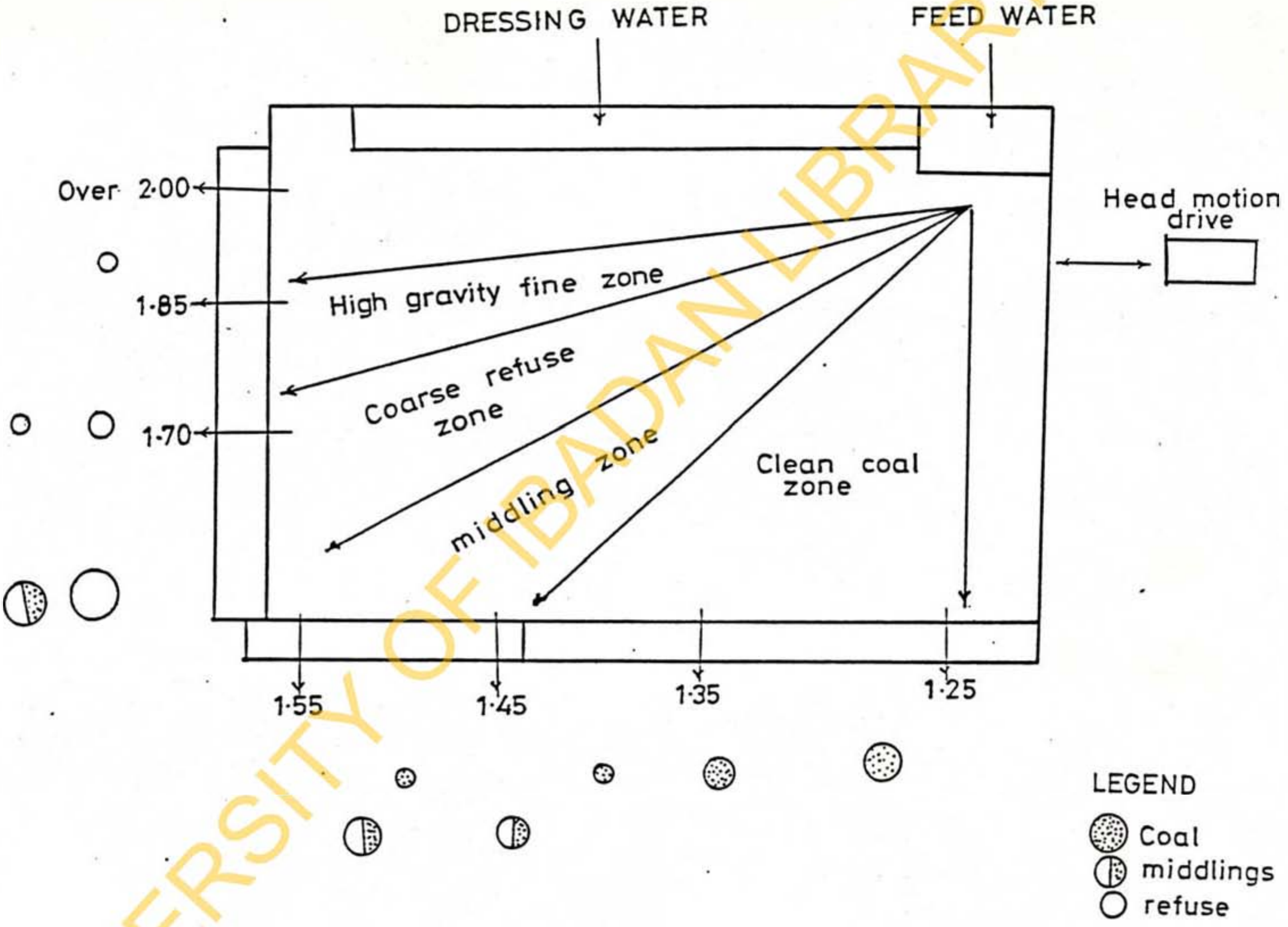

Fig. 34. Distribution of clean Coal Middlings and Refuse on the wet concentrating table according to specif gravity.

(7) shape of deck

(8) character of deck surface

(9) riffle design

(10) amount and distribution of wash water

(11) feed water-solids ratio

(12) feed rate, and

(13) feed uniformity.

Concentrating tables are essentially a rectangular or rhombohedral shaped deck (Figure 34) supported by suitable frame and bearings, generally sloping in the direction of the short axis and usually tilted upwards at the refuse end of the long axis and differentially reciprocated in the direction of the long axis.

This action of head motion tends to move the particles longitudinally towards the end of the table, while at right angles to this movement wash water is distributed along the high side of the table which tends to move particles down the slope parallel to the short axis of the table. Hence, the natural path of a particle on a concentrating table would be from the feed-box corner to the diagonally opposite corner of the deck as shown in Figure 34.

There is a natural tendency for heavier particles to sink to the deck of the table and for lighter particles to stay nearer the surface. As a consequence, the heavier particles are carried further along the longitudinal axis since, as they are on or near the surface of the deck, they are more affected by the reciprocating action of the table. The lighter particles as they are at or near the top of the moving bed or particles, travel slower in a longitudinal direction and thus become separated from the heavy particles.

Wash water and the diluting water in the feed cause the movement of the lighter gravity particles down the short axis of the table. Thus the feed material fans out from the feed box, where it enters on to the table with heavy gravity particles moving in one direction and lighter particles moving in another. By proper adjustments of all the factors previously mentioned, a clear-cut separation is possible between high and low gravity particles or between refuse and coal.

Riffles are of great importance to tabling and are responsible for their considerably greater capacity over smooth decks and for greater accuracy of separation.

Behind the riffles, particles are delayed in their downward movement along the slope of the deck. Hindered settling, consolidation trickling, and other phenomena occur as the particles tumble over the riffles and stratification ensues. The heavier particles are settled to the deck of the table and trapped behind the riffles thus ensuring their positive movement in the direction of the end of the table.

Since stratification and separation of particles are not complete as a result of any one riffle, a series of riffles are used, repeating the cycle of stratification and hindered settling from riffle to riffle, obtaining purer products as the particles fan out and progress forward and downward over the table.

The smallest heaviest particles stratify themsevles downward to the surface of the deck more quickly and are carried out by table movements toward the refuse end at a faster rate than the coarse heavy particles. Light gravity larger pieces ride on the top layer of particles and flow on down the slope of the deck as a result of the cross flow of wash water at right angles to the shaking movement of the table.

Heavy large particles settle down next to the fine heavy particles and are carried behind the riffles towards the longitudinal end but do not ride as high up the table as the heavy fine particles.

There is a tendency for the fine or small size light gravity particles to lie between the coarse heavy particles and work over into the zone with the coarse refuse and, if the only separation action on a table were that of straight stratification, this would result in fine coal getting over into the refuse. Fortunately, however, hydraulic classification occurs under hindered settling conditions which tends to counteract this and by proper use of water dilution and wash water as well as riffle design the small light particles of coal are washed off with coarse light particles or coarse coal.

When there is a definite clear cut gravity difference between coal and refuse, a wide size range or an unclassified feed may be treated very efficiently. When there is a large proportion of material intermediate in gravity between coal and refuse, the separation of an unsized feed will become increasingly difficult.

Figure 34 shows zones where the different material types would fall when a tabling exercise is completed.

In only rare instances, on coals of very difficult cleaning characteristics, it is necessary to cut a middling or a third product from a table. When it is done, these middlings are normally disposed of as a secondary fuel since little can be accomplished by retabling such material.

The advantage of a classified feed where a large proportion of middlings or near gravity material exists is that the danger of having small light gravity particles of coal get over into the refuse or middling zone vanishes. Sharper lines of demarcation can be made, which result in very high efficiencies.

For most coals, however, it has been demonstrated that under proper conditions of operating an unclassified feed may be used for cleaning sizes ranging from 25mm to 0, and produce products containing relatively few misplaced particles. On the extreme fines, separation may be effective on particles as small as 200 mesh with reasonably efficient recoveries on the 100 x 200 mesh sizes.

Brief mention should be made of the effect of particle shape in tabling. Generally speaking, flat particles have a tendency to pass over with the refuse. Flat particles adhere to the table and have a tendency to move along with heavier gravity particles. Fortunately, in most instances, coal particles are cubical or splintery and many refuse particles are naturally flat in shape.

An unmistakable advantage of a concentrating table which should be mentioned is that the products being made are always visible. The operator can observe his refuse, clean coal, or middlings and can note quickly the effect of any change in feed conditions or table adjustments.

8.2. Experimental

Coal samples were prepared from the two sources of core and shaft samples. The core samples were used to provide a broader view of the coal deposit than would have been the case if only shaft samples had been worked on. Core samples were provided by the geologists that did the exploration and sample sourcing for laboratory investigation. The shaft samples were collected and prepared as described in the section of sample collection.

The samples for wet concentration tabling experiments were crushed using Denver roll crusher set to the desired particle size. The crushed sample was in each case wetted with water before being introduced on to the table. Slurries of samples were thus introduced on to the table via the feed point when the power was put on and the table had started its shaking action. Tap water was opened to allow inflow of dressing water on to the table surface. Tap water flowing at the rate of 5 litres a minute had been optimised and kept constant through all the tabling experiments. Also it was found that a time of 30 minutes was sufficient for tabling a sample and so the time was also kept unvaried for each tabling experiment. At the end of a tabling experiment, the products of concentrate, and refuse which had been collected separately were each filtered on white satin cloth. They were then spread thinly on the cloth and left in the laboratory air to dry for twenty-four hours. When dried the sample products were carefully gathered into a light aluminium measuring bowl in which they were weighed on an analytical balance.

From the knowledge of the coal feed weight and that of the product, the percentage product yield was calculated. Schematic diagrams showing the flowsheets for the wet concentrating table are given in Figures 35 and 36. The products were each then further re-ground to prepare them for chemical analysis using minus 72 mesh size (British standard) sieve. Each analysis sample was passed 100% through the sieve. Ash, moisture, volatile matter, total sulphur, pyritic sulphur were determined as described previously.

The results of wet concentration tabling experiments are given in Tables 63, 64, 65, and 66.

8.3. Results and Discussion

From the results tables it was observed that the finer the coal sample feeds were ground the better the pyrite liberation and rejection. It was also observed that minus 200 mesh size gave particles that were too fine for practical coal beneficiation. As such, plus 200 mesh and say minus 48 or minus 35 mesh size had to be used.

8.3.1. 6mm By 0 Coal Particle Size of Lafia Coal

Five series of tabling were done on the 6mm by 0. These were (1) 6mm by 0, (2) 6mm by 14 mesh, (3) 14 by 48 mesh, (4) 48 by 200 mesh and (5) 200 mesh by 0 (Table 64). The aim was to see which size range responded best to desulphurization.

When the 6mm by 0 was tabled pyritic sulphur reduced from a value of 3.09% in the feed coal to 0.62% in the clean coal table product. This represented 80% desulphurization achievement. The yield of clean coal itself was 86.3%. Total sulphur reduced from its value of 3.66% in the feed to 1.21% in the clean coal table product. That again was 67% desulphurization of the total sulphur species.

Taking the minus 6mm by 0 coal as a whole, the 6mm by 14 mesh portion of it was 41.6%. When the 6mm by 14 mesh coal was tabled the pyritic sulphur reduction achieved was from 2.9% in the feed coal to 0.56% in the clean coal table product giving 81% desulphurization of pyritic sulphur. Total sulphur in coal reduced from the coal feed value of 3.4% to 1.13% or 67% desulphurization of total sulphur species and clean coal yield was 87%.

TABLE 64:

EFFECT OF COARSE MESH SIZES ON LAFIA COAL DESULPHURIZATION BY A WET CONCENTRATING TABLE

Product	Percent					
	Weight	Ash	Pyritic sulphur	Total sulphur	Pyritic sulphur reduction	Total sulphur reduction
1.	6 mm by 0, 100.0 percent					
Clean coal	86.3	11.6	0.62	1.21	80	67
Refuse	13.7	45.4	18.48	18.84		
Feed	100.0	24.5	3.09	3.66		
2.	6 mm by 14 mesh, 41.6 percent					
Clean coal	87.3	13.5	0.56	1.13	81	67
Refuse	12.7	36.7	19.19	19.60		
Feed	100.0	23.4	2.92	3.47		
3.	14 by 48 mesh, 38.9 percent					
Clean coal	86.1	12.8	0.47	1.07	83	69
Refuse	13.9	39.7	17.13	17.59		
Feed	100.0	24.0	2.75	3.50		
4.	48 by 200 mesh, 15.2 percent					
Clean coal	83.8	11.2	0.55	1.18	84	69
Refuse	16.2	67.8	17.67	17.80		
Feed	100.0	25.3	3.34	3.82		
5.	200 mesh by 0, 4.3 percent					
Clean coal	86.1	22.1	2.48	3.14	59	51
Refuse	13.9	71.8	27.49	27.54		
Feed	100.0	35.0	5.99	6.47		

For the 14 by 48 mesh size portion which was 38.9 percent of the whole 6mm by 0 coal sample the pyritic sulphur reduction was from the coal feed value of 2.75% to 0.47% in the clean coal or 83% desulphurization of pyritic sulphur species. The total sulphur in this fraction reduced from 3.05% in the coal feed to 1.07% in the clean coal or 69% desulphurization with respect to the total sulphur. Yield of clean coal was 86%.

For the coal particle size 48 by 200 mesh which constituted 15.2 percent of the whole 6mm by 0 total sample, the pyritic sulphur reduction attained was from 3.34% in the feed coal to 0.55% in the tabling product. That was a desulphurization of 84% of the pyritic sulphur. Total sulphur in this size fraction reduced from 3.82% in the feed coal to 1.18% in the clean coal or 69% desulphurization of the total sulphur. The yield of clean coal was 84% of its fractional quantity.

4.3 percent of the whole 6mm by 0 coal fraction was of the size 200 mesh by 0. For this fraction the pyritic sulphur reduced from 5.99% in the feed coal to 2.48% in the clean coal. 59% pyritic sulphur removal was achieved on the fraction. Total sulphur in the fraction went down by 52% or from 6.47% in feed coal to 3.14 in the tabling product. Clean coal yield was 86%.

TABLE 65:

EFFECT OF FINE MESH SIZES ON LAFIA COAL DESULPHURISATION BY A WET CONCENTRATING TABLE

Product	Percent					
	Weight	Ash	Pyritic Sulphur	Total sulphur	Pyritic sulphur reduction	Total sulphur reduction
	35 mesh by 0, 100 percent					
Clean coal	77.0	12.0	0.84	1.55	56	39
Refuse	23.0	50.2	4.94	5.37		
Feed	100.0	23.9	1.93	2.54		
	35 by 100 mesh, 63.4 percent					
Clean coal	64.0	11.5	0.53	1.26	74	53
Refuse	36.0	53.2	4.73	5.16		
Feed	100.0	25.0	2.06	2.68		
	200 mesh by 0, 36.6 percent					
Clean coal	91.8	14.2	1.31	1.96	23	17
Refuse	8.2	27.4	6.50	7.06		
Feed	100.0	21.3	1.70	2.37		

8.3.2. 35 Mesh By 0 Coal

Particle Size of Lafia Coal

As shown in Table 65 the coal particle size of 35 mesh by 0 was tested in three series, namely,

(1) 35 mesh by 0, (2) minus 35 mesh by plus 200 mesh and

(3) 200 mesh by 0.

The yield of clean coal concentrate for the 35 mesh by 0 was 77 percent. Pyritic sulphur reduction of the fraction was from 1.93% in the feed coal to 0.84% in the clean coal tabling concentrate. That was equivalent to 56% desulphurization with respect to pyritic sulphur content. The total sulphur removal realised in the fraction was from 2.54% in the feed coal to 1.55% in the clean coal concentrate. Total sulphur species was thus reduced by 39%.

The yield of clean coal when feed coal size 35 by 200 mesh was tabled was 64.0%. At the same time pyritic sulphur reduced from 2.06% to 0.53% or 74% pyritic sulphur rejection. Total sulphur removal was from 2.68% in the feed coal to 1.26% in the clean coal tabling concentrate which was 53% total sulphur rejection.

The size 200 mesh by 0 yielded 92% clean coal concentrate.

TABLE 66:

RESULTS OF DOUBLE TABLING OF LAFIA COAL

Sa/No.	Feed Size	Table Feed total sulphur	Clean coal total sulphur	% total sulphur reduction	% clean coal recovery
1.	9 mm x 0	2.82	2.12	24.8	91.8
2.	30 mesh x 0	-	1.40	50.4	87.5
3.	9 mm x 0	3.34	1.76	47.3	84.9
4.	30 mesh x 0	-	1.07	68.0	81.4
5.	9 mm x 0	3.83	3.29	14.1	92.4
6.	30 mesh x 0	-	2.72	29.0	87.4
7.	9 mm x 0	4.38	2.57	41.3	94.4
8.	30 mesh x 0	-	1.65	62.3	90.7
9.	9 mm x 0	4.77	1.98	58.5	83.5
10.	30 mesh x 0	-	1.54	67.7	78.8

N.B. (-) means no analysis was carried out.

with the high fractional total sulphur content of 1.96%. The pyritic sulphur reduction achieved in this fraction was from 1.70% to 1.31% or 24%. The total sulphur was reduced from 2.37% to 1.96% or 17% total sulphur rejection.

8.3.3. DOUBLE TABLING OF LAFIA COAL

Table 66 shows the result of Lafia coal particle size 10mm by 0 tabled twice. The first tabling product (9mm by 0) was re-ground to finer particle size (30 mesh by 0). The total sulphur content of the feed coal, the first table product and the second table product were determined and the percentage clean coal yield calculated for each experiment.

The total sulphur rejection achieved in the first tabling of the coarse (9mm by 0) coal particle size ranged between 14 and 58% indicating that desulphurization of coarse size of Lafia coal does not give much sulphur rejection. However, the results obtained from the second tabling of re-ground fine (30 mesh by 0) coal feed indicated total desulphurization in the range of 29% to 68% with tabling product yield of between 78% and 91%.

From the results discussed, it might be concluded that 6mm by 0 size coal when tabled as a whole gave a high yield of 86 percent and pyritic and total sulphur removal of 80%

and 67% respectively. This value compares favourably with the literature values ranging between 50 to 80 per cent of desulphurization commonly achieved in table concentration of high pyritic sulphur coals. It might be further concluded that the size range of 6 mm by 0 responded so well to desulphurization and was close enough not to require any further size classification. Grinding the coal finer than minus 6 mm particle size would not achieve better desulphurization but would involve higher cost in grinding power consumption and wear and maintenance of the grinding machinery. The sample ground to minus 35 mesh by 0 which was discussed above did not produce as much high yield (77%) as the minus 6 mm by 0 (80%). Also the sulphur rejection achieved on the 35 mesh by 0 was much lower (56% pyritic, 39% total sulphur) than the 6 mm by 0 (80% pyritic and 67% total sulphur).

Double tabling gave desulphurization of total sulphur varying from 29% to 68% with coal tabling product yield of between 79% and 91% while consuming less grinding energy than was the case in tabled coals of size 6 mm by 0 or 35 mm by 0. Double tabling is an innovation of this research study as there is no previous similar work to compare it with but it has worked out well from the results.

TABLE 67:

RESULTS OF WET CONCENTRATION TABLING OF LAFIA COAL SEAMS 13 (SAMPLE NOS 1-5) AND 12 (SAMPLE NOS 6 - 10) AT THE PARTICLE SIZE LEVEL OF 19mm BY 0

Product	Percent							
	Weight	Ash	Moisture	Volatile matter (d.a.f.)	Total sulphur	Pyritic sulphur	Pyritic sulphur reduction	Total sulphur reduction
Clean coal (1)	33.9	19.4	1.5	31.8	1.30	0.44	71	38.0
Refuse (1)	66.1	30.7	2.13	34.1	1.25	0.87		
Feed (1)	100.0	27.4	2.45	32.2	2.10	1.50		
Clean coal (2)	31.2	16.9	1.53	27.0	1.33	0.35	80	45.0
Refuse (2)	68.8	29.9	2.04	31.8	1.39	0.86		
Feed (2)	100.0	27.4	2.50	31.9	2.42	1.73		
Clean coal (3)	43,9	19.5	1.72	27.9	1.35	0.32	83	48.0
Refuse (3)	56.1	31.7	2.24	28.6	1.29	0.90		
Feed (3)	100.0	26.9	2.30	32.0	2.70	1.90		
Clean coal (4)	57.8	18.8	1.74	28.5	1.37	0.75	60	42.4
Refuse (4)	42.2	30.8	2.21	28.5	1.24	0,44		
Feed (4)	100.0	27.2	2.40	31.8	2.38	1.89		
Clean coal (5)	66.5	19.4	1.68	28.2	1.29	0.45	75	51.7
Refuse (5)	33.5	35.2	2.41	28.9	1.23	0.95		
Feed (5)	100.0	26.6	2.33	31.5	2.67	1.77		

TABLE 67:Contd.

Product		Percent units							
		Weight	Ash	Moisture	Volatile matter (d.a.f)	Total sulphur	Pyritic sulphur	Pyritic sulphur reduction	Total sulphur reduction
Clean coal	(6)	84.4	22.6	2.10	27.2	1.21	0.82	56	55.9
Refuse	(6)	15.6	59.5	2.20	38.3	2.47	1.90		
Feed	(6)	100.0	26.4	2.15	32.0	2.74	1.85		
Clean coal	(7)	78.7	21.1	2.01	27.3	1.27	0.80	55	61.0
Refuse	(7)	21.3	47.8	2.29	33.6	1.49	0.91		
Feed	(7)	100.0	27.5	2.22	31.8	3.26	1.79		
Clean coal	(8)	80.9	21.6	1.90	27.4	1.26	0.79	65	63.7
Refuse	(8)	19.1	54.2	2.12	36.2	2.34	1.86		
Feed	(8)	100.0	26.4	2.26	32.2	3.47	2.25		
Clean coal	(9)	81.4	22.5	1.88	27.4	1.23	0.84	46	47.5
Refuse	(9)	18.6	58.9	1.94	38.4	2.15	1.65		
Feed	(9)	100.0	27.5	2.10	32.0	2.34	1.55		
Clean coal	(10)	85.9	22.3	2.09	27.5	1.22	0.75	69	60.5
Refuse	(10)	14.1	66.9	1.77	44.7	2.68	1.85		
Feed	(10)	100.0	27.4	2.20	31.8	3.09	2.44		

8.3.4. Coal Particle Size Level of 19mm By 0

Table 67 contains the results of wet concentrating table of Lafia coal seams 12 and 13. Total sulphur reduction achieved by tabling seam 12 ranged between 48% and 61%. Pyritic sulphur removal in seam 12 also varied from 46% to 65%. The yield of clean coal tabled concentrate was between 79% and 86%. For seam 13 pyritic sulphur removal was higher, varying between 60% and 80% and total sulphur desulphurization attained was only within the range of 38% to 52%. The coal table product concentrate was much lower (34% to 67%), than for seam 12 (79% to 86%). On the whole, for the size consist 19mm by 0 the yield of table concentrating washery was low, the sulphur rejection itself was low and the ash content of the concentrate was high. As such, therefore, the particle size 19mm by 0, being coarse, has no merit to recommend it for Lafia coal to be washed at that size. The smaller size range 6mm by 0 discussed earlier showed sufficiently good economy (51% to 69% total sulphur desulphurization and 50% de-ashed, 86% yield) and produced satisfactory level of sulphur reduction which makes one recommend it as the practical size for tabling washery for Lafia coal. Seam 12 coal also responded well enough to double tabling to make one recommend the method for practical use for desulphurization of the seam coals.

A tentative flow diagram for the wet concentration tabling to desulphurize Lafia coal is given below and it is based on either double tabling in which the coarse product of the first tabling is further ground finer and retabled or a once fine-ground tabling (Figures 35 and 36).

The Run-of-Mine (ROM) coal would be delivered from the bunker near the mine mouth by conveyor and would be screened at 9mm, the over-size crushed and recycled to the screen. The undersize from the screen would pass to an over-spill feed bunker from where the raw coal will be fed at a controlled rate to the first table. The refuse from the first table is channelled off to a refuse lagoon. The refuse lagoon requires to be treated with lime to neutralize its acidity. The clean coal product of the first table is filtered and the filter cake sent to a crusher which now grinds the coal to minus 30 mesh particle size. The re-ground coal is now channelled to the second wet concentration table. The refuse again is sent off to the refuse lagoon. The coal concentrate is dewatered using a centrifuge. The filter cake is the desulphurized clean coal which could now be stored in a storage bunker where it is

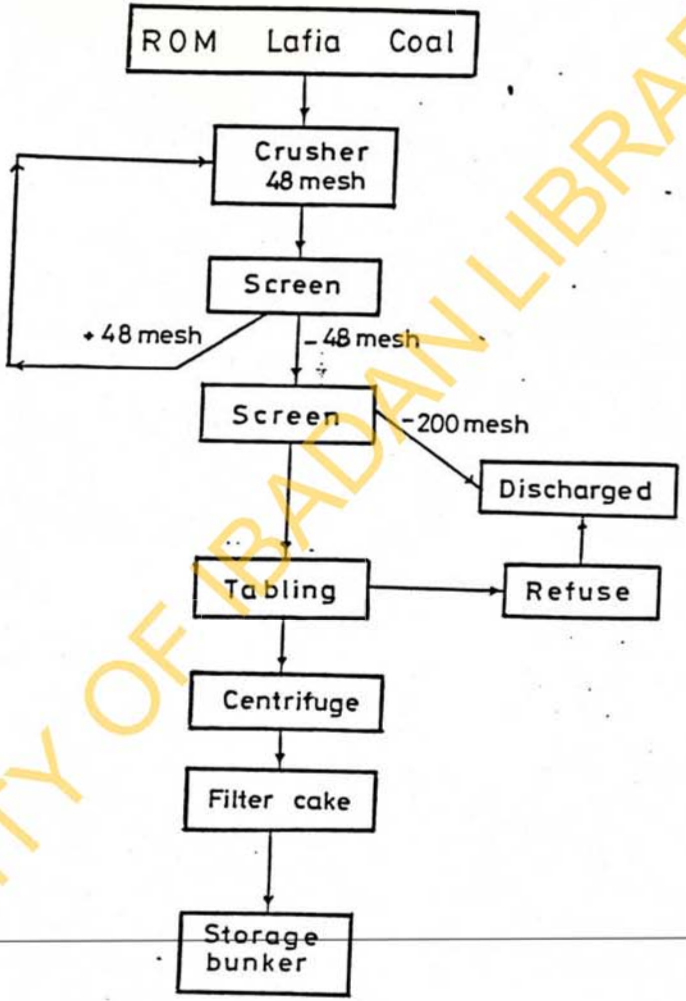

Fig. 35. Tentative flowsheet for once wet concentration tabling to desulphurize Lafia coal.

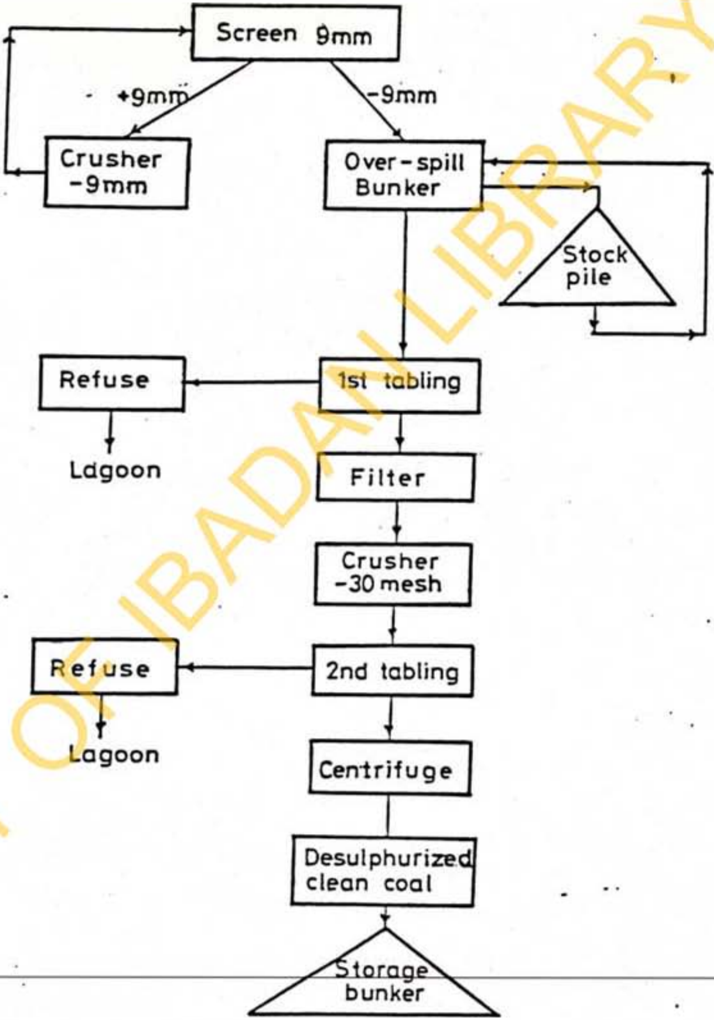

Fig.36: Tentative Flow Diagram for the Double wet concentration Tabling to Desulphurize Lafia coal.

available for use, sale or transportation.

For the once tabling process the only variation is that minus 200 mesh coal dust is discharged while the minus 48 plus 200 mesh coal fraction is tabled. The refuse material emanating from the table is itself discharged. The desulphurized clean coal concentrate is filtered using a centrifuge. The filter cake is stored in a storage bunker ready for subsequent use.

8.3.5 CONCLUSION

Finely ground Lafia coal responds positively to wet table concentration with desulphurization of about 65%, yield above 80% for 6 mm by 0 particle size or finer top size coal, The results of the tests carried out indicated that Lafia coal can be desulphurized using the technique of wet concentrating table.

Feed water and steady power supply are the prerequisites for successful wet table desulphurization operation. It is a cheap process to operate requiring little attention and low over head costs. It is a technique that should be confirmed at the pilot scale to optimise its operative parameters for good process economy.

CHAPTER NINE

APPLICATION OF HEAVY MEDIA SEPARATION TO THE DESULPHURIZATION OF NIGERIAN LAFIA COALS

9.1. Introduction

The Heavy Media Separation (HMS) method has been extensively reviewed in Chapter 2. It entails the physical separation of coal from the admixture of siliceous, pyritic and other types of gangue based on the difference in the specific gravity of the impurities and clean coal. It is a method commonly used for desulphurization of coarse coal particles. At the pilot or industrial scale of testing, the materials used to set and adjust the media density are fine sand and magnetite as informed from the literature review. However, at the laboratory scale the bath is too small to keep the magnetite and/or sand in constant suspense to give a steady media density. For this reason organic liquids like carbon tetrachloride, benzene, and bromoform were used to establish the desired media specific gravity for the studies undertaken.

9.2. INSTRUMENTATION AND REAGENTS

A deep iron mortar with its pestle was used in crushing the coal lumps to smaller sizes and fines. An iron mortar and pestle were used because they contain little silica which could interfere with the result of the ash and other inorganic mineral content of the coal apart from iron. The British standard of sieve was used to separate the different particle sizes of coal.

For the Heavy Media Separation tests, a mixture of carbon tetrachloride and benzene was used as the dense medium to set the density gradations as was done under coal washing in Chapter Five. Although carbon tetrachloride and benzene are toxic chemicals if inhaled profusely, enough care was exercised to avoid such dangerous level of inhalation. Open air space outside was used for the experiments in place of the laboratory room. In that way the natural air-currents diluted the toxic vapours of the media chemicals.

The determination of ash, moisture, volatile matter content, sulphur, reflectivity, maceral counts of liptinite, vitrinite, and fusinite as well as the crucible swelling and ultimate chemical analysis of coal as received and the treated coal were carried out according to the methods previously described in Chapter 2.

9.3. PROCEDURE FOR SIEVE ANALYSIS AND HEAVY MEDIA SEPARATION OF SEAMS 12 AND 13 SAMPLES

The coal sample weighing 1 kg was crushed until it was 100 percent below 3mm. The lot was then mixed thoroughly. The pulverized coal was then sieved into the classes minus 3 plus 2mm, minus 2 plus 1mm, minus 1 plus 0.5mm and the rest was treated as minus 0.5mm. The size-classified samples were then weighed to determine the percentage weight distribution and then analysed for ash and total sulphur content.

Each class was then introduced into a medium of specific gravity 1.55 comprising of a mixture of carbon tetrachloride and benzene. A portion sank and the other floated. The floated sample was scooped off, drained and allowed to dry in an oven at 30°C for 24 hours. The portion that sank was filtered, drained dry and transferred into an oven at 30°C also for 24 hours. The float and sink portions were separately prepared for laboratory analysis for ash and sulphur in order to monitor the distribution of these parameters in the float and sink fractions.

9.4. RESULTS AND DISCUSSIONS

The results of sieve analysis and heavy media separation (HMS) are given in Tables 68 and 69 for seam 12, and Tables 70 and 71 for seam 13. Each set of results represent the mean of three replicate determinations.

Seam 12

From the results the minus 2 plus 1 class had the highest percentage weight distribution of 32.79 percent followed by the minus 3 plus 2mm with a value of 24.95 percent distribution. The ash and sulphur distribution did not follow that pattern. The ash distribution decreased regularly with the coal particle size as the minus 3 plus 2mm class had the highest ash content of 21.34 percent and minus 0.5mm had the lowest of 19.46 percent. The sulphur distribution pattern showed the finest particle size of minus 0.5mm had the highest total sulphur value of 2.27 followed by minus 1 plus 0.5mm class with sulphur value of 2.09 percent.

For the floats of seam 12 at the specific gravity of 1.55g/cm the coal recovery was quantitatively satisfactory over all the sizes as it was in each case (Table 68) higher than 85 percent.

TABLE 68: RESULTS OF SIEVE ANALYSIS OF LAFIA SEAM 12 COAL

Size (mm)	% Wt. Distribution	% Ash	% S
-3 + 2	24.98	21.34	2.05
-2 + 1	32.79	20.94	2.01
-1 + 0.5	18.38	20.36	2.09
-0.5	23.83	19.46	2.27
Run-of-mine coal	100	32.70	2.84

TABLE 69: RESULTS OF HEAVY MEDIA (S.G. 1.55) SEPARATION OF LAFIA SEAM 12 COAL

Size (mm)	% Recovery (Concentrate)	Analyses				Percent desulphurization achieved
		Concentrate		Tailing		
		% Ash	% S	% Ash	% S	
-3 + 2	88.02	17.13	2.01	40.54	3.30	29.2
-2 + 1	87.48	18.62	2.01	40.93	3.26	29.2
-1 + 0.5	87.48	14.33	1.96	42.40	3.35	31.0
-0.5	85.26	16.02	1.85	41.55	4.02	34.9
Run-of-mine coal	untreated -	32.70	2.84	-	-	-

Qualitatively the float of each size fraction was unsatisfactory with respect to the ash (>10<20%) and sulphur (2%) contents as these were still higher than tolerable in metallurgical coal (ash 10%, sulphur 1.0%). There were some obvious separations and rejections of the unwanted ash and sulphur if one compares the higher values of these entities in the tailings than in the concentrates or their smaller values in the concentrates than in the original coal pieces. The desulphurization achieved varied within the range of 29.2 to 34.9%.

Seam 13

The percentage weight distribution of the classes have values that are close to one another. The minus 2 plus -1mm class was highest at 33.57 percent distribution by weight and the minus 1 plus 0.5mm the lowest at 18.78 percent (Table 70). The ash and sulphur distribution in the classes are in each case respectively above 23.96 and 2.52 percent which are high values for these entities.

For seam 13 coal concentrate (Table 71) the recovery of coal was, for each size class, above 65.79 percent.

TABLE 70:

RESULTS OF SIEVE ANALYSIS OF LAFIA SEAM 13 COAL

Size (mm)	% Wt. Distribution	% Ash	% S
-3 + 2	23.98	26.57	2.56
-2 + 1	33.57	27.05	2.65
-1 + 0.5	18.78	25.90	2.60
-0.5	23.65	23.96	2.52
Run-of-mine coal	100	33.50	3.02

TABLE 71:

RESULTS OF HEAVY MEDIA (S.G. 1.55) SEPARATION OF LAFIA SEAM 13 COAL

Size (mm)	% Recovery (Concentrate)	Analysis				Percent desulphurization achieved
		Concentrate		Tailings		
		% Ash	% S	% Ash	% S	
-3 + 2	65.79	18.41	2.56	44.61	3.98	15.2
-2 + 1	66.01	18.19	2.65	43.65	4.22	17.3
-1 + 0.5	67.65	17.29	2.60	45.54	4.19	13.9
-0.5	67.01	15.84	2.52	45.04	4.98	16.6
Run-of-mine coal	untreated	33.5	3.02	-	-	-

The ash and sulphur of the concentrate were also high with the minimum ash found in size class minus 0.5mm at a value of 15.84 percent as well as the minimum value of total sulphur of 2.52 percent. Desulphurization achieved ranged between 15.2% and 17.3% (Table 71). This is a pointer to the fact that heavy media separation may not produce substantial desulphurization of Lafia coal.

9.4.1. PROCEDURE FOR HEAVY MEDIA SEPARATION OF CORE SAMPLES OF SEAMS 5,7,11,12,13,14,18,19,20 AND COMPOSITE CORE SAMPLES OF SEAMS 12 AND 13

It was decided to compare the behaviour of coals of other seams with that of seam 12 towards heavy media separation washery. Nichol's hydrometer was used to follow specific gravity adjustment. 400cm^3 of carbon tetrachloride (S.G. 1.594), and 144.6cm^3 of benzene (S.G. = 0.95) were mixed to prepare the medium of specific gravity of 1.55. Coal samples ground to pass 16 mesh aperture were separated into the floats and sinks using the medium of 1.55 specific gravity. The floats were then analysed for moisture, ash, volatile matter and total sulphur contents.

For comparison proximate analysis was carried out on the raw coal. In addition to proximate analysis, ultimate analysis was done on the concentrate to determine the elements carbon, hydrogen, nitrogen, sulphur and oxygen content of the coal. The methods of analysis have been described in detail in Chapter 4. Petrographic maceral analysis was done on the concentrates. The degree of reflectivity of the coal was also determined. Caking characteristics using crucible swelling number was also determined. The results of these determinations are contained in Tables 72 and 73.

9.4.2. RESULTS AND DISCUSSIONS

Moisture content in Lafia coal is low (range 1 - 3, mean 2) and portrays the coal as mature. Both the raw coal and the media-concentrated coal were low in moisture (Tables 72 and 73). The ash of the raw coal was higher than 26.0 percent in all the composite samples tested.

Volatile matter of both the raw and the heavy media separated coals was between 26.4 and 30.6 which is regarded as medium volatile matter.

TABLE 72: RESULTS OF ANALYSIS OF LAFIA COAL CONCENTRATE FROM MANY SEAMS AT SPECIFIC GRAVITY 1.55

Se/No	Sa/No	Seam No	M	A	VM	Stot	P	R^o %	Lip	Vit	Semi-vit	Fusi-nite	CSN (mm)	Dilatation %	C	H	N	S	O	Hygroscopic moisture %
1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21
1	1494, 1495, 1500	12	0.8	10,7	-	5,14	-	1,09	-	93	4	8	-	149	-	-	-	-	-	1.10
2	1514		2.6	17.6	30.8	2.11	-	-	-	93	5	2	15	100	87,93	5,09	1.91	1.66	3.41	2.17
3	1519, 1524, 1526	12	1.2	19.9	29.1	0.69	0.004	1.14	-	93	6	1	18	88	86.97	6.78	1.86	0.86	4.53	1.42
4	1529		0.9	15.7	29.6	3.09	0.003	-	-	95	5	-	-	132	87.75	5.58	1.97	1.83	2.80	1.22
5	1531	13	1.1	15.1	29.7	2.31	0.006	-	-	92	6	2	-	138	87.62	5.23	2.17	1.70	3.53	1.54
6	1534-1535	12	1.8	19.5	28.4	1.32	0.004	1.09	-	93	4	3	17	80	87.88	5.40	1.88	1.51	3.33	1.63
7	1539	20	4.1	14.4	30.3	1.03	0.002	-	-	95	5	-	18	140	87.52	5.34	2.35	1.22	3.57	2.31
8	1544-1548	12	1.1	15.1	29.3	2.55	0.005	1.07	-	94	6	-	-	114	86.96	5.37	2.07	0.40	5.56	1.39
9	1556-1558	11	1.7	14.8	28.8	2.46	0.002	-	-	94	5	1	21	149	87.72	5.62	2.02	2.37	2.27	1.59
10	1560	7	1.3	16.2	28.7	3.21	0.005	-	-	92	6	2	-	106	86.48	5.22	1.58	1.63	5.09	1.41
11	1565	20	1.9	17.5	31.2	3.46	0.01	-	-	94	4	2	15	116	86.24	5.42	1.92	2.50	3.92	2.15
12	1570	13	2.7	15.1	29.4	2.99	0.003	-	-	95	5	-	20	142	88.57	5.85	1.96	1.81	2.31	2.04
13	1616-1617	20	2.8	14.2	30.6	1.79	0.002	-	-	93	7	-	20	110	88.74	5.64	1.93	1.46	2.23	2.16
14	1623-1625	13	2.1	17.3	29.2	1.18	0.001	-	-	94	4	2	21	120	88.28	5.50	1.61	1.35	3.26	1.64
15	1627-1639	12	1.2	14.9	29.8	3.07	-	1.07	-	96	4	-	-	152	87.09	5.16	2.12	1.79	3.92	1.15
16	1641	12	3.6	11.6	27.8	0.95	0.002	1.12	-	94	5	1	21	136	-	-	-	-	-	2.37
17	1653	14	2.0	14.7	26.7	2.63	0.002	-	-	91	9	-	13	59	88.93	5.32	1.64	1.75	2.36	1.99

TABLE 72:(contd.)

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21
18	1655	13	1.2	14.2	27.6	1.44	0.002	-	-	94	5	1	20	120	88.38	5.44	1.94	1.19	3.05	1.33
19	1657-1675	12	1.1	16.6	27.1	1.90	0.001	1.14	-	93	5	2	17-18	106	88.52	5.23	2.11	1.61	2.53	1.19
20	1664	12	0.9	16.4	27.1	..	0.002	1.17	-	92	7	1	20	107	87.42	5.32	2.09	1.99	3.18	1.07
21	1679	12	1.0	18.6	28.4	2.04	0.002	1.10	-	94	6	-	18	104	88.22	5.37	2.33	0.69	3.39	1,24
22	1678-1685	12	1.1	13.7	28.1	-	-	1.11	-	92	5	3	-	95	87.16	5.22	1.70	2.58	3.34	0.81
23	1691	5	0.6	9.0	27.7	3.05	0.004	-	-	96	4	-	-	141	87.72	5.38	1.99	1.46	3.45	1.14
24	1692	5	1.0	14.8	29.0	3.37	0.007	-	-	94	6	-	17	137	86.86	5.50	2.21	1.94	3.49	1.47
25	1693	5	0.7	11.0	28.2	2.80	0.004	-	-	95	5	-	-	144	87.88	5.17	1.86	1.98	3.16	1.19
26	1698	14	1.4	14.3	26.4	2.54	0.006	-	-	91	7	2	15	87	87.58	5.38	1.90	2.41	2.78	1.76
27	1699-1709	12	0.9	16.8	26.8	2.08	0.01	1.18	-	94	4	2	20	108	87.93	5.42	2.07	2.35	2.23	1.06
28	1716	18	2.2	13,5	28.8	2.39	0.002	-	-	94	6	-	22	140	87.85	5.42	2.13	1.23	3.37	1.81
29	1723-1730	12	1.1	17.8	28.8	2.45	-	1.09	-	93	4	3	21	119	88.14	5.70	1.46	1.42	3.28	1.10
30	1731	13	0.9	16.1	28.5	3.25	0.009	-	-	93	5	2	20	117	87.36	5.26	2.14	1.49	3.75	1.34
31	1732	13	0.8	15.8	27.9	3.14	0.002	-	-	92	7	1	-	116	85.96	5.48	1.95	3.32	3,79	1.40
32	1733	13	0.8	12.2	28.3	1.79	0.002	-	-	93	6	1	19	147	88.05	5.46	1.95	0.79	3.75	0.93
33	1746-1750	19	1.0	15.8	27.0	2.36	-	-	-	94	6	-	-	116	88.13	5.27	1.68	1.38	3.54	1.21
34	1752	18	1.7	15.4	26.6	2.15	0.002	-	-	94	4	2	-	100	88.45	5.35	2.10	1.64	2.46	1.49
35	1761	19	0.9	19.8	28.8	2.16	0.002	-	-	94	5	1	-	121	-	-	-	-	-	-
36	1767	14	1.2	14.8	26.7	3.77	0.004	-	-	94	4	2	-	57	88.47	5.24	2.10	1.65	2.54	1.68
37	1768-1777	12	0.9	16.8	26,2	2.14	-	1.11	-	90	5	5	17-19	113	88.71	5.58	1.58	1.40	2.73	1.03
38	1782	7	0.7	9.8	26.3	2.54	-	-	-	93	4	3	-	142	88.00	5.36	2.12	2.81	1.71	0.94
39	1789-1796	12	0.6	14.5	27.7	3.00	-	1.15	-	93	7	-	-	137	87.83	5.10	1.26	1.53	4.19	0.99
40	1800-1816	12	0.7	16.2	26.7	2.46	-	1.15	-	93	5	2	24	125	87.77	5.27	1.64	1.14	4.13	0.97
41	1820-1881	12	1.1	16.5	29.0	-	0.0001	1.07	-	95	4	1	20	107	86.84	5.32	2.16	2.91	2.77	1.02

LEGEND

M = moisture
A = ash
VM = volatile matter
Stot = total sulphur
P = Phosphorus

R^{o}% = Random vitrinite reflectance % in oil
Lip = Liptine t.C
Vit = Vitrinite
CSN Crucible swelling number

C = carbon
H = Hydrogen
N = Nitrogen
S = Sulphur
O = Oxygen

Table 73: Results of analysis of composite core samples of Lafia coal at specific gravity 1.5

S/N	Composite Sa/ No.	Bore-hole No.	Seam No.	Thickness (m)	Raw coal proximate analysis, % wt				Concentrate proximate analysis % wt				Plasticity		Petrographic maceral analysis				Ultimate analyses (elementary analyses), % wt daf					Hygrosc. moisture %
					M	A	VM	Stot	M	A	VM	Stot	CSN	%Dilatation	R°%	Vit %	Semi vit %	Fusinite	C	H	N	S	O	
1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25
1.	1336A	B-51	-	0.95	0.6	27.6	28.8	1.33	0.9	14.2	27.4	2.16	26	135	1.23	94	4	2						-
2.	1538A	B-56	12	0.55	0.8	39.9	30.0	3.66	1.8	19.5	28.4	1.32	17	80	1.09	93	4	3	87.88	5.40	1.88	1.51	3.33	1.63
3.	1639A	B-58	12	0.40	0.7	29.6	30.8	3.48	1.2	14.9	29.8	3.07		152	1.07	96	4	-	87.09	5.16	2.12	1.79	3.92	1.15
4.	1648A	B-59	12	0.80	0.5	37.4	32.0	2.51	3.6	11.6	27.3	0.95	21	136	1.12	94	5	1	-	-	-	-	-	2.37
5.	1675A	B.60	12	1.50	0.7	34.6	30.0	2.16	1.0	16.5	27.1	1.67	19	106	1.16	93	5	2	87.97	5.29	2.10	1.80	2.84	1.10
6.	1685A	B-61	12	0.60	0.7	38.5	31.3	2.74	1.10	13.7	28.1	-	-	95	1.11	92	5	3	87.16	5.22	1.70	2.58	3.34	0.81
7.	1707A	B-64	12	0.50	0.7	30.1	29.5	2.26	1.1	17.8	28.8	2.45	21	119	1.09	93	4	3	88.14	5.70	1.46	1.42	3.28	1.10
8.	1777A	B-67	12	0.80	1.0	44.3	29.4	2.52	0.9	16.3	26.2	2.14	18	113	1.11	90	5	5	88.71	5.58	1.58	1.40	2.73	1.03
9.	1816	B-66	12	1.40	0.7	29.1	28.0	2.73	0.7	16.2	26.7	2.46	24	125	1.15	93	5	2	87.77	5.27	1.64	1.14	4.13	0.97
10.	1835A	B-68	12	1.20	1.0	29.9	30.4	2.28	1.1	16.5	29.0	2.28	20	107	1.07	95	4	1	86.84	5.34	2.16	2.77	2.77	1.02

LEGEND

S/N = Serial Number
m = meter
M = moisture
A = ash
VM = volatile matter

S_{tot}^{t} = total sulphur
CSN = crucible swelling number
R°% = Random vitrinite reflectance % in oil
Vit = Vitrinite

C = Carbon
H = Hydrogen
N = Nitrogen
S = Sulphur
O = Oxygen

Sulphur rejection achieved by the heavy media separation was not significant at the particle size of 1mm. This state of affairs indicated that much finer grind of the coal was required for sulphur liberation and subsequent gangue rejection. At much finer grind, however, the heavy media separation method of desulphurization has limited success. The heavy media separation method of desulphurization which operates better with coarse coal becomes inefficient in the case of finely disseminated pyrite. Desulphurization levels achieved were 11.8, 22.7, 62.2., 63.9 percent in some of the experimental runs (Table 72). In some instances, there was no sulphur reduction and in some the ashy materials washed off were non-pyritic. In the latter type of situation the coal concentrate showed higher sulphur content than the raw coal. From the result of experiment No.1, Table 73 it is seen that raw coal contained 1.33 percent total sulphur while the concentrate contained 2.16 percent and thus indicating 62.4 percent worsened position of sulphur content. In experiment No.10 of results, Table 73 the sulphur contents of the raw coal and that of the heavy-media-separated coal were the same value of 2.28 percent which indicated there was no sulphur reduction in that borehole seam 12 composite core sample.

The results of heavy media separation tests put together point to the fact that Lafia coal could be best washed when fine-ground and only desulphurization methods that treat fine-ground coal would give good results in the attempt to desulphurize the coal.

Ash reduction achieved was in the range of 42.4 to 63.2. From the results, Table 73, experiment No.1 ash reduction was 48.6 percent; No.2 ash reduction was 51.1. percent. The resultant ash content in the coal concentrate in the experiments No.1 and No.2 were 14.2 and 19.5 percent which are values too high to be used for high grade pig iron production via the blast furnace process.

The elementary carbon content of Lafia coal concentrate was between 86.86 and 88.93 percent, as in Tables 72 and 73. Hydrogen content had a value lying between 5.0 and 5.6 percent. Elementary nitrogen value ranged between 1.46 and 2.33 percent. Elementary oxygen had the value that ranged between 2.36 and 5.09 percent. It can thus be seen from the ultimate (elementary) analysis that Lafia coal was high in carbon content, high in hydrogen content, high in nitrogen content, and also high in oxygen content.

Of these, it is only the high carbon content that is in advantage in metallurgical coal evaluation. The high values of other parameters diminish from the coke product quality of the coal.

The plasticity of the heavy media separated coal concentrate as measured by the crucible swelling number and the percentage dilatation was high which is a valuable character in coke manufacture.

Coal petrology gave a random reflectance of the vitrinite of Lafia coal to be between 1.07 and 1.23 which is indicative of high rank coal. The maceral components had values of vitrinites that range between 90 and 96 percent, semi-vitrinite of between 4 and 6, and fusinite of between 1 and 5. These maceral counts indicate fusible, plastic coal which will require the addition of coal that has strengthening, non-fusible fusinite (e.g. Enugu coal) for metallurgical coke manufacture.

9.5. CONCLUSION

From the results of investigations carried out using Heavy Media Separation (HMS), the conclusion can be drawn that Lafia coal requires to be ground fine before the finely-disseminated pyrite crystals, carbo-pyrite and other carbo-minerites could be effectively liberated. Using mixtures of benzene and carbon tetrachloride (Sp. Gr. 1.55) did not give encouraging results for desulphurization of Lafia coal irrespective of the particle size of the coal.

This is not surprising since this method is best suited for coarse coal. However, about 29% desulphurization of Lafia coal (Table 73) was achieved with coarse coal particle sizes, with most of the pyritic sulphur remaining in the washed coal. Invariably, finer coal particle sizes were used for investigation but without much improvement in coal desulphurization. Therefore, Heavy Media Separation would not be recommended for Lafia coal desulphurization.

CHAPTER TEN

CONCLUSION AND RECOMMENDATIONS

10.1. CONCLUSION

Coal occupies a strategic position in the world energy scene. With so much instability in the world petroleum market these days coal is the sure hope and bridge to the future energy supply up to the early years of the 2000s [139,140]. The advanced countries as well as the developing nations will all continue to depend on coal for power generation, metallurgical industry, industrial and commercial thermal energy, home-heating and other domestic energy requirements to augment woodfuel or to totally replace same in response to the campaign to counter desert encroachment.

Nigeria is in dire need of metallurgical coal for the success of the Ajaokuta Steel Company (ASCL). With transport, handling and stocking infractructures yet to be developed to cater for large scale (1 million metric tonne) annual supply, it has been estimated [32] that a metric tonne of imported coking coal might cost up to US150$ ex-Ajaokuta.

At today's exchange rate of the naira (US1.0$ = ₦4.29 as at 1st Nov., 1987) a metric tonne of metallurgical coal would then be costing ₦643.5, Six hundred and forty three naira, fifty kobo by the time it is off-loaded in Ajaokuta. Judging from such phenomenal cost of coal it was thought useful to undertake an applied research at the doctoral level to develop a technology for making Nigerian coal resource usable for the metallurgical industry.

Proximate and ultimate chemical analyses were carried out on Lafia coal as well as two other major coal deposits in Nigeria, that is, Enugu and Okaba coals so as to characterize them.

Out of the vast coal resources of the country, it is only Nigerian Lafia coal deposit which has shown agglomeration and cokeability. For reasons of such desirable physico-chemical properties and the dearth of coking coal in Nigeria, Lafia coal was chosen for this study.

The chemical analysis results gave the relative proportion of types of sulphur in Lafia coal as organic sulphur (0.79 - 1.00%), pyritic sulphur (1.01 - 4.04%), and sulphate sulphur (0.01 - 0.07%). Thus it became known that the major form of sulphur in Lafia coal is pyritic sulphur.

Total sulphur in Lafia coal ranged between 1.9 - 5.6%. High sulphur coal when carbonized will produce high sulphur coke. Pig iron produced using high sulphur coke will itself (i.e the iron) have sulphur partitioned into it. It has been known that high sulphur steel is brittle. So in effect it is necessary to desulphurize Nigerian Lafia coal to render it suitable for use as raw material for Ajaokuta Steel Complex.

The study of "Desulphurization of Nigerian Lafia coal for metallurgical coke preparation" in itself gives a direction to the methods of investigation. It is firstly meant to be an applied research investigation with the hope that a process for the appropriate industrial use will emanate from the study. Secondly, the physico-chemical nature of coal is such that chemical desulphurization being reducto-oxidative type of reaction would reduce from the coking propensity of coking coal. From the literature survey done chemical desulphurization has not resulted in producing coal for coke preparation.

The obvious choice of route for the desulphurization of Nigerian Lafia coal was thus physical methods which are known to produce clean coal suitable for preparing metallurgical coke.

Coal Petrology has proved useful as an analytical tool in monitoring the process and progress of coal desulphurization. The instrumentation and reagents required to put coal petrology into operation were simple and easy to maintain and operate. Principally they consisted of the white-light microscope and its accessories, the sample polishing machine and consumables like carborundum (SiC) and alumina or chromite polishing powders. With practical coal petrology it was possible to see the coal macerals as well as the inorganic mineral inclusions. As desulphurization progressed the diminishing particles and crystals of the impurities were followed visually. At the same time the resultant concentration of the coal macerals became apparent and evidently convincing.

Photomicrographs of Nigerian Lafia, Enugu and Okaba coals have been produced. All other studies of Nigerian coals carried out previously have not produced such clear photomicrographs. This is probably because such earlier studies carried out principally by foreigners have found Nigerian coals too strange for their understanding.

This research work has therefore sort of Nigerianised the study and understanding of local coals.

Other areas of applied coal petrology which is another whole study on its own and which is very worthwhile is to relate the quantitative maceral and carbo-minrite composition of Nigerian coals to the volatile matter and ash of these coals. It will also be of interest to metallurgical coke preparation to relate the abundance (or otherwise) of vitrinite, exinites and inertinite macerals of Nigerian coals to the strengths of coke produced from different coking blends.

The pyritic nature of the greater part of the high sulphur content of Nigerian Lafia coal has made xanthate pyrite collector useful in an effort to desulphurize Lafia coal. The desulphurization which was done in two stages involved a first stage normal flotation of coal as the froth float concentrate and the second stage of reverse flotation when coal was depressed and pyrite impurities were floated off. The underflow in the second flotation stage was the clean coal concentrate 2. Another variant of the investigation was two double normal flotation in which coal was froth-floated in both stages and shaly as well as pyritised gangue were the underflow.

Then, comparing the two variants it was observed that the total sulphur retained in the clean coal product of the two stage (Normal and Reverse) flotation ranged between 1.02 and 1.21 percent while that of the double normal flotation ranged between 0.94 and 1.70 percent. That is to say that the froth float concentrate from the two stage flotation, normal and reverse, was a lot cleaner than the concentrate from the double normal froth flotation. It was observed that once it was finely ground to about minus 60 mesh sieve size two stage flotation of Lafia coal produced a clean coal that would be suitable for metallurgical coke preparation, the ash and sulphur having been lowered by 73.3% and 60.9% respectively. The clean coal product yield of 48.4% was however, low.

The justification for proposing the double flotation venture for use of Lafia coal despite of its low percentage yield lies, however, in the fact that the dearth of coking coal in the world makes such coals too costly as an import item on an industrial scale. Although local production entails a sizeable capital investment it is still the cheaper option than importation of coking coal.

However, because of the geochemical characteristics of Nigerian Lafia coal as proved by the experimental investigations carried out in this study it requires to be desulphurized using physical methods to meet the qualities of a metallurgical coal. The desulphurization becomes feasible only after crushing and grinding the coal to fines of minus 60 mesh size. The three techniques of desulphurization of Lafia coal that have proved effective are 1) washability test, 2) wet concentrating table, 3) the two-stage froth flotation.

Since the coal seam structure (section 1.10) is variable washability test will always have to be done precedent to either of wet concentrating table or the two-stage froth flotation to establish desulphurization and ash removal chracteristics of separate seam gross samples and consignments. While the latter two methods are capable of desulphurizing the coal the first choice is the two-stage froth flotation because its clean coal concentrate is cleaner than the wet concentrating table clean coal product; its infrastructure is cheaper to establish, operate and maintain.

10.2. RECOMMENDATIONS

1. The washability test results of Nigerian Lafia coal gave an encyclopedic view of the behaviour of the coal towards improvement studies. The characteristic curve, the float cumulative curve, as well as the sink cumulative curve, the specific gravity curve and the ±0.1 specific gravity distribution curve, all came out clearly only after the coal had been crushed to minus 60 mesh sieve particle size. Any consideration of Lafia coal outside its fine state would not be very useful. In addition to giving an accurate picture of the coal when ground fine, coal for both carbonization and combustion normally require being ground into such fines as minus 60 mesh for best results. In fact there is presently the practice of coal powder injection (CPI) into the blast furnace for improved performance and greater economy of the process and this practice requires the coal to be pulverized. To dry the cleaned coal at this particle size offers no problem as the centrifuge and/or vacuum drying systems have been in use for such fine coal size without the risk of oxidation. The method of doing the washability which has been described in this study is highly recommended for use for this coal whenever its behaviour is to be examined.

Its reproducibility has been confirmed by the mean of nine separate determinations which agreed closely with the data and curves generated by a single run.

2. The wet concentrating table is also recommended for use to desulphurize Lafia coal judging from the results obtained from its use in this study. It gave clean coal with considerable ash and sulphur removal. It is a process that is not sophisticated to set up and to operate. coal improvement results are obvious before one's eyes ás the process goes on. the process flow-sheet developed in figures 30 and 31 apply to industrial scale of desulphurization beneficiation of Lafia coal. In figure 30 it is a once wet concentration tabling to desulphurize the coal. The run-of-mine coal is pulverized to minus 48 mesh and the product is tabled to produce clean coal which is centrifuged and the resultant cake transported on a converyor belt to a storage bunker. The second flow sheet (Figure 31) is first tabled at particles size minus 9mm. The product of first tabling is further crushed to minus 0.5mm which is in turn re-tabled to produce the final desulphurized clean coal which is either used immediately or stocked in the bunker. Refuse generated during the processes is sent to refuse lagoon which is kept moquito-free by regular sanitation-attention.

3. The unique two stage froth flotation method of desulphurization is also recommended for industrial plant use for desulphurizing Nigerian Lafia coal. from the results of this study either the double normal froth flotation or the two stage (normal/reverse) flotation produced good level of sulphur and ash reduction. Industrial size of flotation washery of coal fines is already in operation and so it will be no new machinery-design. The chemical reagents too have been in use from time on mineral sulphides. The application now will be on coal and not metalic sulphides.

The products of the three processes, namely, the washability test, the wet concentrating table and the two stage froth flotation will meet the requirement for metallurgical coke preparation. Since in coke preparation coals of several types and ranks are mixed, then, during the carbonization blends mixing desulphurized Nigerian Lafia coal is blended with those other complimentary coals that are low (about 0.5%) in sulphur, e.g. Enugu coal.

4. As pointed out earlier-on it is not known of a chemical desulphurization method that produces metallurgical coal. It is a challenge to coal chemists to come up with a method of desulphurizing coal chemically without destroying its cokeability.

5. Pilot plant and later the industrial scale tests of the findings of this study are recommended since the scale of operation described herein has been laboratory scale. It is desirable that there be a confirmation of these findings at the larger scale in pilot plants and in industries before the processes could be passed on to enterpreneurs.

REFERENCES

1. International Committee for Coal Petrology (ICCP), 1963, International handbook of coal petrology, 2nd edn., Paris, Centre National de l'a Recherche Scientifique.

2. Hendricks, T.A., 1945, The origin of coal, Chap.1 in Chemistry of coal utilization, H.H. Lowry, ed., vol. I, London, Chapman and Hall.

3. Van Krevelen, D.W., 1981, Coal, Coal Science and Technology 3, Typology - Chemistry-Physics-Constitution, Chap. 1, Amsterdam, Elsevier Scientific Publishing Company.

4. Gibson, J., 1979, Coal: an introduction to its formation and properties, Chap. 1, in Coal and Modern coal processing, an introduction, Pitt, G.J. and G.R. Millward, eds., London, Academic Press.

5. Deluc, J.A., 1979, Physique et Morales sur les montagnes et sur phistoire de la terre et de phomme, Lettres, 5, 213 - 225.

6. Rogers, H.D., 1843, Origin of coal, Repts, Am.Assoc. Geologists and naturalists, 71, 434 - 467

7. Hutton, J., 1795, Theory of the Earth with Proofs and illustrations, London, Codell and Davies, Vol.1, 565 - 586.

8. Potomie, H., 1910, die Entstchung der steinkohle und der Kaustobiolithe, Berlin, Gebruder Borntraeger, 5th edn.

9. Daubree, A., 1846, Origin of Fusain, Bull.Soc.geol. France, 3, 153 - 157.

10. Karsten, C.J.B., 1826, Mode of origin of Fusain (mineral charcoal), Arch. Bergbau Huttenw., 12, 3 - 244.

11. White, D., and R. Thiessen, 1913, Fusain in Coal, U.S. Bur. Mines, Bull. 38, 390.

12. Lessing, R., 1925, Nature and origin of Mineral Matter in Coal, J.Soc. Chem. Ind., 44, 277 - 283.

13. Given, P.H., 1960, The distribution of hydrogen in coals and its relation to coal structure. Fuel., A jnl. of fuel science, 39, 147.

14. Report of the United Nations Conference on new and renewable sources of energy, 1981, Nairobi, 10 to 21 August, 1981, A/CONF.100/11, United Nations, New York.

15. Elliot, C.R., 1982, Coal desulphurization prior to combustion, Park Ridge, Noyes data corporation.

16. Von Beroldingen, F., 1950, Beol achtungen, Zweifel, und Fragen die mineralogie uberhaupt, und insberondere ein naturliches mineral-system betreffend, Hannover, 1st edn., 17(8).

17. U.S. Bureau of Mines, 1969, International Coal Trade, Vol. 38(8), 153 - 70.

18. Deubrouck, A.W. and P.S. Jacobsen, 1974, coal cleaning - state of the art, Coal and Environmental Technical Conference and Equipment Exposition, Louisville, Ky, U.S.A.

19. Rose, H.J., 1945, Classification of coal, in Chemistry of coal utilization by H.H. Lowry, John Wiley and sons, Inc., New York, U.S.A., 1, 24 - 85.

20. Wilson, C.L., 1982, Coal - Bridge to the future, Report of the world coal study, Cambridge, Ballinger.

21. Wilson, C.L., 1982, Future Coal Prospect, Country and Regional Assessments, World Coal Study, Cambridge, Ballinger Publ. Co.

22. Hutton, C.A. and R.N. Gould, 1982, Cleaning up Coal, A study of coal cleaning and the use of cleaned coal, Allen, C.R. (Ed.), Cambridge, Ballinger Publishing Company.

23. Perch, M. and C.C. Russel, 1949, A study of coal classification and its application to the coking properties of coal, <u>Trans.AIME-SME</u>, 184, 205 - 212.

24. U.S. Bureau of Mines, 1968, Coal - bituminous, lignite and anthracite, 1968 yearbook preprints, 12 - 34.

25. Rees, P.B., 1975, Exploitation of Deposits: Exploration, The Mining Engineer, 134, 1 - 25.

26. Noyes, R., 1982, Coal Resources, characteristics and ownership in the U.S.A., Park Ridge, Noyes Data Corporation.

27. Mitchell, D.R. and H.B. Charmbury, 1963, Cleaning and Preparation in Lowry, H.H., (Ed.), Chemistry of coal utilization, supplementary volume, New York, Wiley and Sones, Inc., Ch.8, 320 - 321.

28. Kennedy, T.H. and A.W. Thornton, 1949, Sulphur in coke, Yearbook of Amercian Iron and Steel Institute, 222 - 42.

29. Swardt de, A.M.J., and P.O. Cassey, 1961, The Coal Resources of Nigeria, Geol.Surv. of Nigeria, Bull. No.28, Lagos, Govt. Printers.

30. Powell Duffryn Technical Services Ltd., 1961, on Nigeria: First Report to the Undersecretary of State for the Colonies, Colonial Office, Dover House, White Hall, S.W.1, on the Government Colliery, Enugu, The Characteristics of the Coal produced and the Investigations into the other coal and mineral resources, Colonial Reports, Miscellaneous No 33 of 1903 - 1904, Cd 2876 of 1904 - 5, Cd 4994 No 67 of 1505 - 6.

31. Final Draft Report, 1984, Industrial Scale Carbonization Tests in W.Germany, report submitted to the Federal Ministry of Mines, Power and Steel, Steel Development Department, Lagos, by Eisenbau Essen, GMBH.

32. United Nations Economic Commission for Africa, 1986, Development of Coal Resources in Nigeria, Based on advisory services to the Government of Nigeria, E/ECA/NRD/ERU/COAL/280/8/86.

33. British Carbonization Research Association (BCRA) Tests' Results - Private (classified) property of the Federal Ministry of Mines Power and Steel.

34. Chapman, W.R. and D.C.R. Jones, 1955, "The removal of sulphur from coal", Jnl. Inst. Fuel, 28, 102 - 108.

35. Bhatnagar, J.N., 1957, Extracts of organic sulphur in coal, Indian Pat. 54960.

36. Roy, M.M., 1956, Coal sulphur variety, Naturwissenschaften, 43, 21--497.

37. Kavcic, R., 1954, Organic Sulphur in coal, Bull.Sci. 2, 12 - 13, RPF Yougoslavie.

38. Potowski, J.J., and A.B. Harlampovich, 1936, Coal Organic Sulphur, Fuel 15, 229 - 32.

39. Lissner, A., and W. Gobel, 1952, Sulphur Types in Coal, Bergakademie, Freiberg. Forschungsh, A., Bergbau 14, 31 - 4.

40. Mott, R.E., 1936, The history of coke making and of the Coke Oven Managers' Association, (COMA), in the 1936 Yearbook of the Coke Oven Managers' Association, Mexborough, Yorkshire, C.O.M.A., Year Book Ltd.

41. Bashforsh, G.X., 1960, The Manufacture of iron and steel, Vol. I, Iron Production, 2nd Edn., London, Chapman and Hall, p.102.

42. Landry, B.A., 1949, Fundamentals of coal sampling, Bur.of Mines Bull. 454.

43. Bailey A.L. and B.A. Landry, 1949, Sampling of Coal for float-and-sink, Mining Engineering Jnl, 16, p.8.

44. Merle, J.J. and Mullins, R.A., 1950, Laboratory control in coal washing and drying plants, Trans.AIME-SME, 187, 609 - 615.

45. COL, G.D., 1938, An explation of washability sink data on coal, U.S. Bureau of Mines, I C 7045.

46. Keller, R.E. and W.W. Anderson, 1938, "Determining the Washability characteristics of a coal", Commercial Testing and Engineering Company, 228N Lassalle Street, Chicago, Illinois 60601.

47. Picard, J.L. and C.F.J. Rozenhart, 1938, Procedure for Flota-and-sink Analysis, Div. Report FMP 67/14-WRL, Dept. of Energy, Mines and Resources, Ottawa, Canada.

48. U.S. Bureau of mines, 1967, Methods of analysing and Testing Coal and Coke, Bull. 638.

49. Leonard, J.W. and D.R. Mitchell, 1968, Coal Preparation, New York, AIME, 3rd edn., 9 - 5.

50. Pitt, G.J. and G.R. Millward, 1981, Coal and Modern Coal Processing: an Introduction, London, Academic Press, 2nd edn.

51. Wheelock, T.D., 1977, Coal desulphuriztion, Chemical and Physical methods, American Chemical Society, Washington, D.C., U.S.A.

52. Leonard, J.W. and D.R. Mitchell, 1968, Coal Preparation New York, AIME, 3rd edn., 10 - 32.

53. Yancey, H.F., 1931, Determination of shapes of particles and their influence on treatment of coal on Tables, Trans. AIME, 94, 355 - 402.

54. Dell, R.C., 1978, Determination of froth-flotation-washability data, Institute of Fuel Journal, 30, 523 - 526.

55. Sun, S.C., 1954, Effects of oxidation of coals on their flotation properties, Trans.AIME-SME, 187, 30 - 39.

56. Sun, S.C., and R.E. Zimmerman, The Mechanism of Coarse coal and mineral froth flotation, Trans.AIME-SME, 187, 616 - 622.

57. Gandrud, B.W. and H.L. Riley, 1955, Increasing coal flotation-cell capacity, Trans.AIME-SME 1137 - 1142.

58. Baker, A.F., Miller, K.J., and A.W. Deurbrouck, 1973, Desulphurization of coal by froth flotation, Proceedings 6th International coal preparation congress, Paris, France, 27E, 13.

59. Gieseke, E.Q., 1962, Flocculations and filtration of coal flotation concentrate and tailings, Trans.AIME, 226, 352 - 358.

60. Plante, E.C. and K.L. Sutherland, 1949, Effects of Oxidation of Sulphide minerals on their flotation properties, Trans.AIME 183, p.160

61. Miller, K.J., 1975, Coal-pyrite flotation, Trans.AIME-SME, 258, 30 - 33.

62. Miller,K.J., 1974, Flotation of pyrite from coal, U.S. Patent No 3807557.

63. Miller, K.J. and A.F. Baker, 1972, Flotation of pyrite from coal, TPR, 51, U.S. Bureau of Mines

64. Eliot, R.C., 1978, Coal desulphurization prior to combustion, Park Ridge, Noyes Data Corporation, New Jersey, U.S.A.

65. Kester, W.M., 1965, Magnetic demineralization of pulverized coal, Mining Engineer, 17, 8 - 13.

66. Kester, W.M., Leonard, J.W., and E.B. Wilson, 1967, Reduction of Sulphur from steam coal by magnetic methods, Mining Congress Jnl., July, 1967, 71 - 75.

67. Yurovsky, A.Z. and I.D. Remesnikov, 1958, Thermomagnetic method of concentrating and desulphurizing coal, Koks i Khimiia, 1, 8 - 13.

68. Miller F.G., 1964, Reduction of sulphur in minus 28-mesh bituminous coal, Trans.AIME-SME, 229, 11.

69. Bension, W. and C.C. Burgess, 1966, Development of fine aperture sieve bends for slurry classification and application in conjunction with classifying cyclones to sulphur reduction in fine coal, Paper E2, Fifth International Coal Preparation Congress, Pittsburgh, p.11.

70. Schiffman, L.E., 1950, Kerosine flotation of bituminous coal fines, Trans.AIME-SME, 187, 1047-1056.

71. Schiffman, L.E. and E.D. White, 1951, Floating fines with Kerosine, Coal Age, 56, 78 - 82.

72. Wolk, R., Steward, N., and S. Alpert, 1977, Solvent refining for clean coal cobustion, <u>Engineering Index Annual</u>, 76, p.1015.

73. Masciantonio, P.X., 1965, The Effects of Molten Caustic on Pyritic Sulphur in Bituminous Coal, Fuel, <u>A Jnl. of the fuel Science</u>, 44, 269 - 275.

74. Lorenzi, L., Land, J.S. and R.A. Meyers, 1972, Conversion of organosulphur compounds to sulphones using $KmnO_4$, HNO_3, H_2O_2/HOAC, and hydroperoxides, <u>Coal Age</u>, 77, (11), <u>No</u> 3,909, 211.

75. Meyers, R.A., 1977, Coal Desulphurization, Marcel Dekker, Inc., New York, U.S.A.

76. Reed, E., 1968, Organic Chemistry of Bivalent Sulphur, Vol.II, p.64, New York, Chemical Publishing Company, Inc.

77. LaCount, R.B. and S. Friedman, 1977, Chemical desulphurization of coal using air at 100psi and temperature 150 - 180°C, <u>Jnl.Org.Chem.</u> 42, p. 2751.

78. Friedman, S., LaCount, R.B., and R.P. Warzinski, 1977, Use of air under pressure to desulphurize coal, Trans.AIME, <u>Jnl.Eng.Power</u>, 99A, p.361.

79. Worthy, W., 1975, Organic Sulphur Removal from coal using aqueous alkali at 300°C, <u>Chem.Eng.News</u> July 7, 1975, pp.24 - 25.

80. Agarwal, J.C. and S.S. Sareen, 1975, Chemical desulphurization of coal as a viable alternative to stack gas scrubbing, <u>Min. Congr.Jnl</u>, 61, (3), 40 - 43.

81. Sarren, S.S., Gilberti, R.A. and L.J. Petronic, 1977, Ammonia/Oxygen/Water system for pyrite removel from coals, AICLE Symp.Ser., 73 (165), 183 - 189.

82. Diaz, A.F. and E.D. Guth, 1975, Conversion of dibenzothiophenic sulphur to sulphone, U.S. Patent No 3,909, 211.

83. Smith, E.G., 1975, Organic Sulphur removal from coals by oxydesulphurization, Am.Chem.Soc.,Div. of Fuel Chemistry, 169th National Meeting, Philadelphia, 11 - 27.

84. Hutton, C.A. and R.N. Gould, 1982, Advanced Physical Coal Cleaning, in Cleaning up Coal, Allen,R.C. (Ed.), Ballinger Publishing Company, 115 - 119, Cambridge, Ma, U.S.A.

85. Nelson, H.W. Snow, R.D. and D.B. Keyes, 1933, Pyrite Sulphur removal by passing fine air/oxygen bubbles through coal/water suspension, Ind. and Eng. Chem. 25, (12), 1355 - 1371.

86. Agarwal, J.C., Giberti, R.A. and S.S. Sareen, 1975, Chemical Desulphurization of coal, Mining Congr.Jnl., 61, (3), 40 - 59.

87. Sareen, S.S., Giberti, R.A. and L.J. Petrovic, 1975, The Use of Oxygen/Water for Removal of Sulphur from Coals, 80th National Meeting of the Am.Inst. of Chem. Eng., Boston, Massachusetts, 13 - 39.

88. Argawal, J.C., Giberti, R.A. and L.J. Petrovic, 1976, Chemical means of coal desulphurization, U.S Patent No 3, 960, 513.

89. Majuma, H., and E. Peters, 1966. Acid leaching of pyrite at >100°C, Trans.Metall.Soc.AIME, 1409 - 1419.

90. Gaseous Fuels, Coal and Coke, ASTM Standards, 1976, Part 26, Methods D2015 - 66, D2492 - 68, D3174 - 73, D3177 - 75.

91. Stambaugh, E.P., Levy, A., and R.D. Giammar, 1976, Hydrothermal Coal Desulphurization with Combustion Results, Proceedings of the Fourth National Conference, 386 - 394.

92. Stambaugh, E.P. Levy, A. and K.C. Sekhar, 1976, Hydrothermal coal desulphurization with combustion results, Proceedings of the fourth National Conference, 386 - 394.

93. Sum, S., and M.Wu., 1974, Desulphurization by chlorination of coal in the gas phase at high temperature (400°C) and elevated pressure, AIME Annual Meeting, Preprint, Dallas, 28.

94. Imperial, G.R., and P.L. Walker, 1962, Low temperature chlorinolysis, Special Research Report SR-31, Pennsylvania State University, 1 - 18.

95. Zarubina, Z.M., Laylikova, N.N. and Y.I. Shmuk, 1959, Investigations of Microbiological Oxidation of Coal Pyrite, Izvestia Akad Nauk SSSR Otdel Tekh Nank, Metal i Topl, 1, 117 - 119

96. Silverman, M.P., Rogoff, M.H. and I. Wender, 1963, Removal of pyritic sulphur from coal by bacterial action, Fuel,A Jnl of fuel science, 42, 113 - 124.

97. British Standards. 1017, 1942, Methods for sampling of coal and coke, part 1; Sampling of Coal, London, British Standards Institution, 82.

98. British Standards Institution (BSI), Methods of Coal Analysis, B.S.1016, Part 4.

99. Ode, W.H., Coal Analysis and Mineral Matter, in Chemistry of coal utilization, suppl. volume, H.H. Lowry (ed.), London, John Wiley & Sons Inc., Ch.5., 202 - 224.

100. Brit. Standards Assoc., 1942, Brit. Standard 1016, British Standard Methods for The Analysis and Testing of coal and coke: Forms of Sulphur in coal.

101. Hillman, J., 1979, Coal Preparation Projects in the U.K., SME, 1602 - 1608.

102. Fiedner, A.C. and W.A. Selvig, 1951, Determination of forms of sulphur in coal, U.S. Bur. Mines Bull. 492, 16 - 17.

103. Shimp, N.F., J.K. Kuhn and R.J. Helfinstine, 1977, Review of the methods for determining sulphur and forms of sulphur in coal, in Energy Sources, 3, 93 - 109.

104. Powell, A.R. and S.W. Parr, 1919, Methods for the determination of sulphate and pyritic sulphur in coal, Bulletin Americal Institute of Mining Engineers, 2041 - 2049.

105. Teichmann, R.F.J., 1945, Quick determination of sulphate sulphur in coal using strong acid digestion, S.African Mining Eng. Jnl., 56, Part. 2, 99 - 103.

106. Motts, R.A., 1950, Digestions at boiling temperatures for the quick determinations of sulphate and pyritic forms of sulphur in coals, Fuel, 29, 53 - 61.

107. Edwards, A.H., Daybell, G.N., and W.J.S. Pringle, 1958, Comprehensive investigation of methods for determining forms of sulphur in coal, Fuel, 37, 47 - 61.

108. Radmacher, W., and P. Mohrhauer, 1953, Reduction of pyrite by nascent Hydrogen and estimation of hydrogen sulphide for the determination of pyritic sulphur in coal, Gluckauf, 89, 503 - 11.

109. Eschka, A., 1874, Determination of total sulphur in coal, Ost. Z. Berg. U. Huttenw., 22, 11 - 13.

110. Battye, T., Johnson, R., and H.C. Wilkinson,1954, Eschka method of determining total sulphur in coals, Jnl. Appl. Chem., 4, 119 - 23.

111. Marrison, F.C., and R.A. Mott, 1956, Total sulphur determination in coal based on Eschka method, Chem. & Ind. (London), 1956, 715 - 16.

112. Amercian Society for Testing Materials, D271-58, Assoc. Francaise de Nomalisation, M03006. 1963, Bomb - calorimeter method of determination of total sulphur in coal.

113. Skau, E.L. and I.L. Newell, 1933, A rapid volumetric method for determination of sulphur in coal and coke, Analyst, 5, (3), 180 - 2

114. Strambi, E., 1943, Determination of total sulphur in coal, Carole, 16, 61 - 5.

115. Seuthe, A., 1939, High-temperature combustion method Gluckauf, 75, 409 - 11.

116. Beet, A.E., and R., Belcher, 1940, High Temperature Combustion determination of coal sulphur, Fuel, 19, 42 - 5.

117. Deutscher Normenauschuss: DIN51724 1955; British Standards Inst., British Standards 1016, Part 6, 1958.

118. Mott, R.A. and W.C., Wilkinson, 1956, High Temperature Combustion determination of coal sulphur Fuel, 35, 6 - 18.

119. Radmacher, W. and P., Mohrhauer, 1954, Use of lithium metal to estimate coal sulphur, Z. Anl. Chem., 141, 419 - 29.

120. Edgecombe, L.J., 1956, Ion-Exchange Separation and titrimetric determinations of sulphur and chlorine in coal, Fuel, 35, 38 - 48.

121. British Coke Research Association, 1972, Rapid determination of sulphur in coal by x-ray fluorescence spectrometry, Reprint No 71, 1-7.

122. Chakrabarti, J.N., 1978, Analytical Procedures for sulphur in coal desulphurization products, in Analytical Methods for coal and coal products by C. Karr, Jr., Academic Press, New York, U.S.A., 306 - 309.

123. Brown, G. and R. Kanaris-Sotiriou, 1969, The determination of sulphur in soils by x-ray fluorescence Analysis, Analyst 94, 782 - 6.

124. Karr, C., Jr., 1978, Analyst.Methods for Coal and coal products, Vol.I, London Academic Press.

125. Kreider, R.E. and J.G. Foulds, 1954, Bomb Volumetric method for sulphur in refined petroleum products, Anal.Chem. 26, 1983 - 5.

126. Radmacher, W., and P., Mohrhauer, 1952, Rapid determination of sulphur in coal, Brennstoff-Chem., 33, 129 - 35.

127. Elphick, J.O., and T., Gerson, 1952, Estimation of sulphur in coal by conductometric titration, fuel, 31, 438 - 44.

128. Wilkinson, H.C., 1956, The use of 4-amino-4-chlorodiphenyl hydrochloride for the determination of sulphur in coal, Analyst, 81, 9 - 11.

129. Majumdar, S.K., and N.G. Banerjee, 1957, Use of versenate to estimate sulphur in coal, Jnl. Proc. Inst. Chemists (India), 29, 213 - 15.

130. Gordiyevskii, A.V., and E.E. Radnayeva, 1958, Amperometric determination of sulphur in coal, Koks i khim, 7, 14 - 15.

131. Hicks, D., and G. Nagelschmidt, 1943, Medical Research Council (British), special Report series No 244, 153 - 86.

132. McNair, H.M. and E.J. Bonelli, 1965, Basic Gas Chromatography, consolidated printers, Berkeley, California, U.S.A., 1 - 306.

133. Thompson, B.H. and J.E. Garside, 1953, Coal gasification, Gas Engrs., Reprint No 437, 1 - 32.

134. Nelkon, M., and P. Parker, 1982, Advanced Level Physics, Part 3, ch. 20, 5th edn., London, Heinemann Educational Books, 311 - 404.

135. British Standards 1453 Type A8 on Steel Filler rod, 1978, London, British Standard Institution, 18.

136. Bernes, B., 1968, A new method for the decomposition and comprehensive analysis of silicates by atomic absorption spectrometry, Anal.Chem., 40, 1682 - 86.

137. Smith, R.D., Campbell, J.A., and W.D. Felix, 1977, Trace element concentrations in some coal samples and possible emissions from coal combustion in Sweden, Swedish Water and Air Pollution Research Institute, B 358, Gothenbury, Lovblad.

138. Smith, R.D., Campbell, J.A. and W.D. Felix, 1980, Atmospheric Trace Element Pollutants from Coal Combustion, SME-Preprint 79 - 265, AIME Meeting, New Orleans.

139. Onwumechili, C.a., 1973, Proceedings of the second symposium on Science and Technology in National Development, Lagos, 5 - 32.

140. Tooley, P., 1971, Fuels, Explosives, and Dyestuffs, London, John Murray.

APPENDIX

Plates showing different aspects of
Lafia Coal Desulphurization study.

Plate 1
Mannual Coal Sampling
(NMDC, Jos, Nigeria).

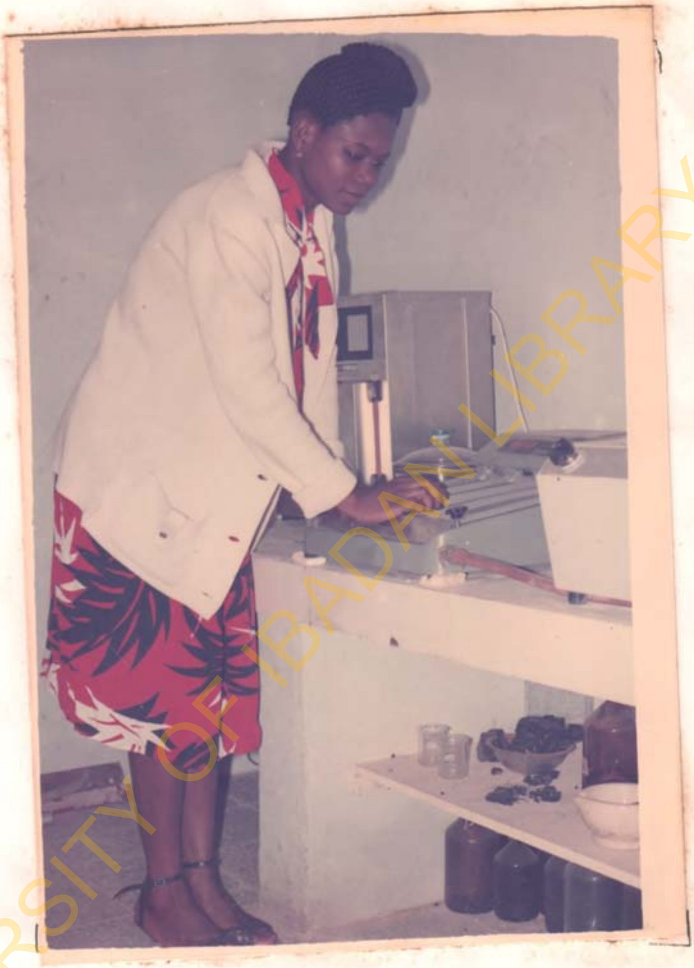

Plate 2
Mannual Coal Polishing
(in NMDC, Jos, Nigeria).

Plate 3

Polished Lafia Coal Piece Showing in situ massive, framboids, patches and crystals of pyrite deposition on cell-wall tissue (high vol.A. Bit, VM 33%) reflected light, oil immersion, x 320.

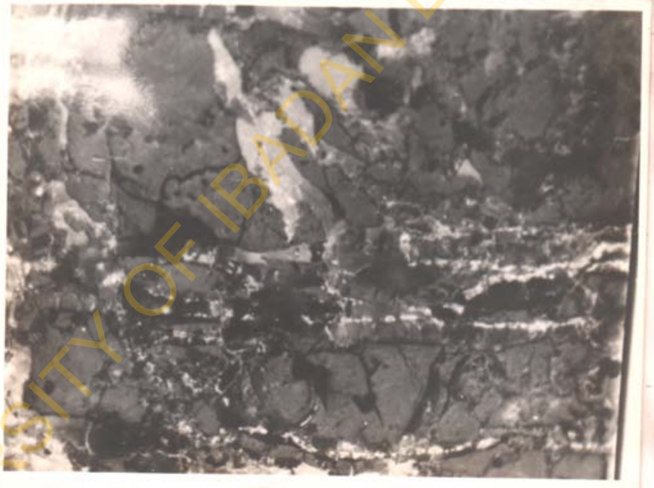

Plate 4
Polished Lafia Coal Piece
showing siderite in cleats
and fissures (reflected
light, oil immersion, x500).

Plate 5
Polished Lafia Coal particulate block showing pyrite crystals, patches, massive and framboids before physical desulphurization (reflected light, oil immersion, x500).

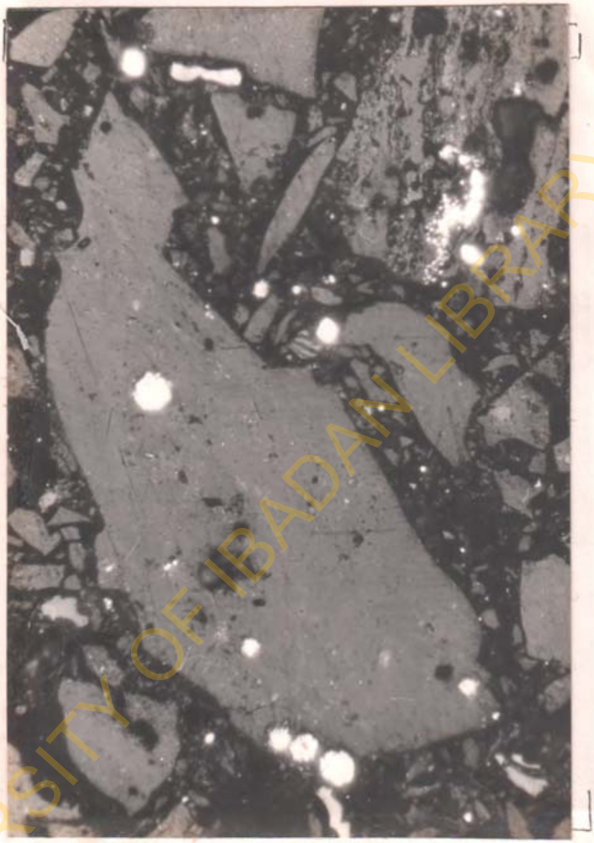

Plate 6
Polished Lafia Coal particulate block of first stage froth flotation desulphurization, (reflected light, oil immersion, x 500), showing some framboids and crystals aggregates of carbo-pyrite entrapped within macerals.

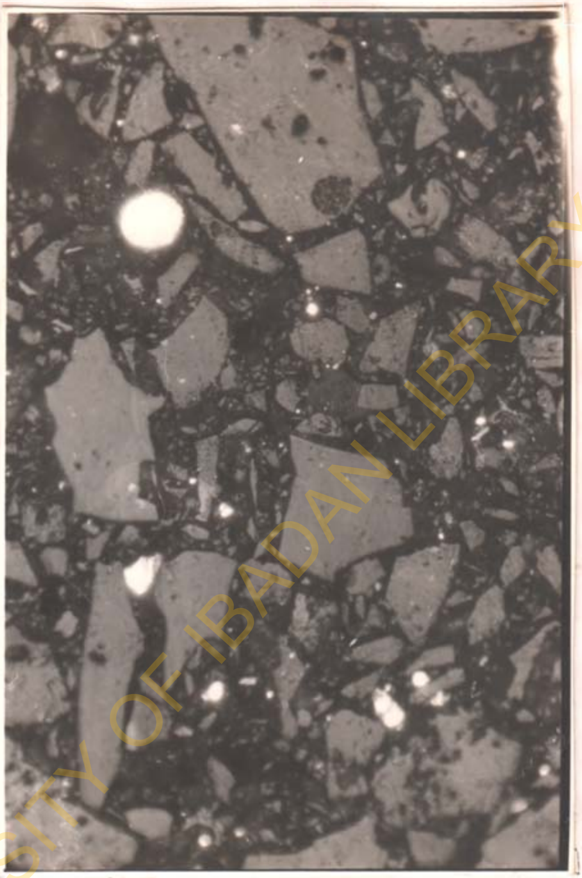

Plate 7
Polished Lafia Coal particulate block of second stage froth flotation desulphurization (reflected light, oil immersion, x500) showing only few entrapped framboid and single crystals of carbo-pyrite.

Plate 8
In general typifying vitrinite with woody structure (tellinite) after van krevelen, D.W., [3] in Coal Science and Technology 3.

Plate 9

In general typifying structureless vitrinite (collinite) after van krevelen, D.W., [3] in Coal Science and Technology, 3.

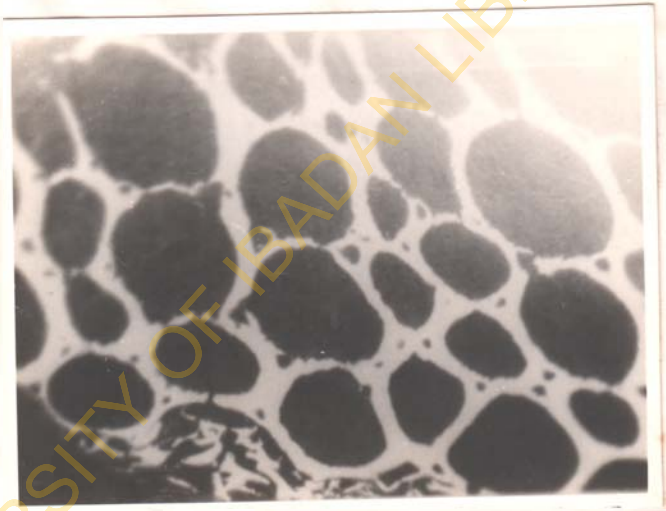

Plate 10
In general showing typical fusinite, after van krevelen, D.W., [3] in Coal Science and Technology, 3.

Plate 11
To show crushed fusinite cell-walls (Bogenstruktur.), after van krevelen, D.W., [3], in Coal Science and Technology.

Plate 12
General structure of semi-fusinite after van krevelen, D.W., [3] in Coal Science and Technology, 3.

Plate 13(a)
General structure of megaspore, after van krevelen, D.W., [3] in Coal Science and Technology, 3.

Plate 13(b)
General structure of microspores, after
van krevelen, D.W., [3] in Coal Science
and Technology, 3.

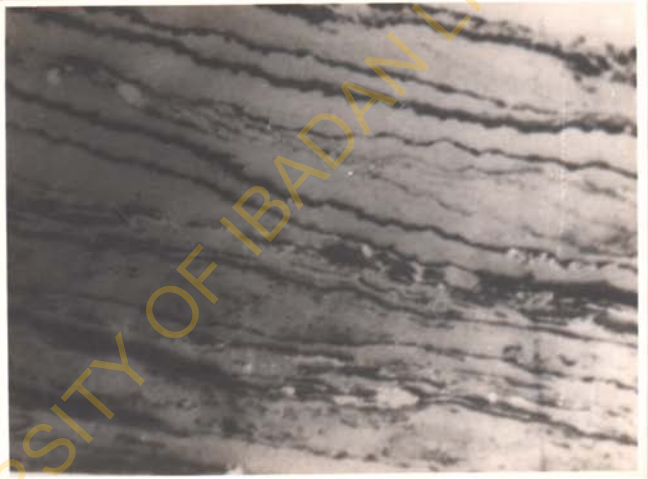

Plate 14
General structure of cuticle after van krevelen, D.W., [3], in Coal Science and Technology, 3.

Plate 15
In general typifying Resinite in coal, after van krevelen, D.W., [3], in Coal Science and Technology, 3.

Plate 16
General structure of alginite, after van krevelen, D.W., [3], in Coal Science and Technology, 3.

Plate 17
General structure of sclerotinite in coal, after van krevelen, D.W., [3], in Coal Science and Technology, 3.

Plate 18
Typifying granular micrinite in coal, after van krevelen, D.W., [3], in Coal Science and Technology, 3.

Plate 19
Laboratory determination of
crucible swelling number of
Nigerian Lafia Coal.

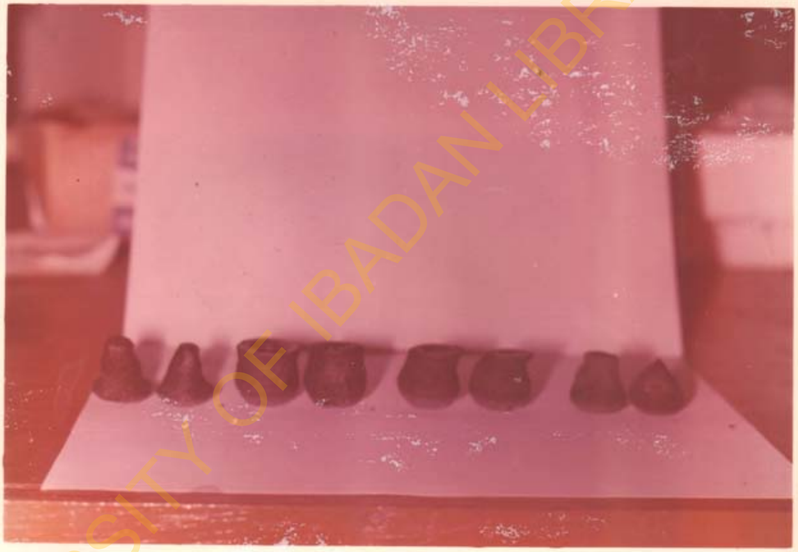

Plate 20
Coke buttons of Nigerian Lafia Coal for crucible swelling number determination.

Plate 21
Flotation cell used in coal desulphurization,
(N.M.D.C., Jos, Nigeria).

Plate 22
Photomicrographs of Okaba Coal, Nigeria, (reflected light, oil immersion, x500) showing megaspore, microspores, fusinites resinites, vitrinite, and mineral inclusions.

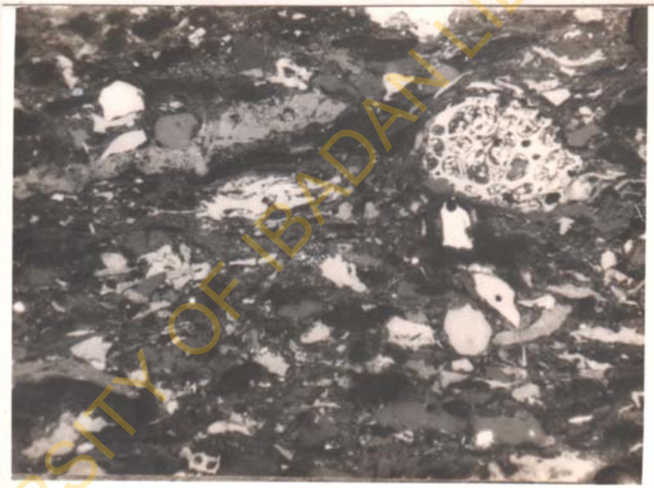

Plate 23
Photomicrograph of Enugu Coal, Nigeria, (reflected light, oil immersion, x500) showing Bogenstruktur, fusinite, microspores, alginite, semifusinite, vitrinite, and mineral inclusions.