

**Petrographic and Geochemical Characterization of
Part of A Permian Coal Deposit of the South-
Eastern Kalahari sub-basin, Moiyabana area,
eastern Botswana**

**By
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TECHNOLOGY (BIUST)**

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August 2018

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CERTIFICATION

The undersigned certifies that he/she has read and hereby recommends for acceptance by the College of Science a dissertation titled: **Petrographic and Geochemical Characterization of Part of A Permian Coal Deposit of the South-Eastern Kalahari sub-basin, Moiyabana area, eastern Botswana** in fulfilment of the requirements for the degree of Master of Science of the BIUST.



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Abstract

The Moiyabana Coalfield, located ~80 km west of Mahalapye village in the South Eastern Kalahari sub-basin, is one of the potential areas for coal mining in Botswana. This project discusses the petrographic and geochemical results of a detailed investigation carried out on a borehole core of the Moiyabana area, eastern Botswana. Geologically, the Moiyabana coal seams occur in sedimentary rocks of Permian Age and are hosted in the Karoo Supergroup sequence. All coal seams are found within the Eccra Group, which comprises of fluvio-deltaic mudstones and siltstones. A total of 8 coal samples from one borehole core were collected from the western part of Moiyabana Coalfield. The maceral analysis reveals a predominance of inertinite group macerals, with minor vitrinite and liptinite group macerals, suggesting possible oxidizing conditions for a prolonged period of time when accumulated vegetal matter was exposed to the surface during peatification. Mineralogical analysis shows that these coals are characterized by high mineral matter dominated by silicate minerals. In terms of coal rank, using vitrinite reflectance in the range of 0.84-1.04 (mean RoVmr% values), placing the Moiyabana coal samples in the medium rank C bituminous coal, borderline B, using ECE-UN rank classification. Proximate analysis data indicate low contents of moisture and volatile matter, moderate fixed carbon and high ash content. The low to medium sulphur content suggests a possible fresh water environment after the deposition of precursor peat. Major oxides of Si, Ti and Al positively correlate with ash content, indicating inorganic affinity, while other oxides have negative correlation with ash content revealing organic modes of occurrence. Elements As, Al, B, Ba Cd, Cu, Cr, F, Ge, Hg, Li, Mo, Ni, Pb, Sb, Si, Ti, U and V exhibit positive correlations with ash yield, suggesting inorganic affinity, whereas elements Ca, Cl, K, Mg, Mn, Na, P, S, Se, Sr, Th and Zn negatively correlate with ash yield, signifying a potential organic mode of occurrence. When compared to the concentration ranges (Coal Clarkes) in coals worldwide, the generally low concentration of trace elements in the Moiyabana coals, particularly those considered to be hazardous air pollutants, gives an advantage for this coal should it be used for power generation.

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Table of Contents

1	INTRODUCTION	1
1.1	Background to the study	1
1.2	Geographical Overview	6
1.3	General Geology.....	7
1.4	Previous Work	12
1.5	Aim and Objectives.....	13
1.6	Organisation	14
2	LITERATURE REVIEW.....	15
2.1	Coal Sampling	15
2.2	Sample preparation	16
2.3	Coal Petrography.....	16
2.3.1	Macro-petrography	17
2.3.1.1	Humic coal	17
2.3.1.2	Sapropelic coal	18
2.3.2	Micro-petrography	18
2.3.2.1	Coal preparation for micro-petrography	18
2.3.2.2	Analytical Techniques in micro-petrography	19
2.3.2.2.1	Maceral analysis.....	19
2.3.2.2.2	Vitrinite reflectance measurements	20
2.3.2.3	Microscopic description of coal	21
2.3.2.4	Mineral content of coals	22
2.3.2.5	Depositional environment models.....	23
2.3.2.5.1	Petrographic Facies.....	23
2.3.2.5.2	Mineral matter proportions.....	23
2.4	Coal Quality	24
2.4.1	Chemical properties of coal.....	24

2.4.1.1	Proximate analysis.....	24
2.4.1.1.1	Proximate analysis test methods	25
2.4.1.1.2	Proximate analysis as coal quality component	25
2.4.1.2	Ultimate analysis.....	25
2.4.1.2.1	Ultimate analysis test methods	26
2.4.1.2.2	Ultimate analysis as coal quality component	28
2.4.1.3	Calorific value analysis	29
2.4.1.3.1	Calorific value analysis test methods.....	29
2.4.1.3.2	Calorific value analysis as coal quality component.....	30
2.5	Coal Rank.....	30
2.5.1	Biochemical phase	31
2.5.2	Geochemical phase.....	31
2.6	Classification of coal	33
2.6.1	ASTM Classification	33
2.6.2	South African Classification.....	34
2.6.2.1	Rank	34
2.6.2.2	Petrographic composition	34
2.6.2.3	Ash yield	35
2.6.3	Russian Classification	35
2.7	Elements in Coal.....	36
2.7.1	Techniques in geochemical analysis of elements.....	37
2.7.1.1	Inductively coupled plasma –atomic emission spectrometry (ICP-AES)	37
2.7.1.2	Inductively coupled-plasma mass spectrometry (ICP-MS).....	37
2.7.1.3	Graphite furnace atomic absorption spectrometry (GFAA)	38
2.7.2	Mode of occurrence of elements	38
2.7.2.1	Major elements	38

2.7.2.2	Trace elements	39
2.7.3	Trace elements emission during coal combustion.....	40
2.7.4	Other analysis	41
2.7.4.1	Chlorine	41
2.7.4.2	Phosphorus.....	41
2.8	Summary	41
3	METHODOLOGY	43
3.1	Sampling.....	43
3.2	Sample Preparation	44
3.3	Coal Petrography.....	45
3.4	Proximate Analysis	46
3.5	Total Sulphur Analysis	46
3.6	Ultimate Analysis (C, H, O and N analysis)	47
3.7	Calorific Value Analysis	47
3.8	Geochemical Analysis	47
3.8.1	Major elements analysis.....	47
3.8.2	Trace elements.....	48
4	RESULTS.....	49
4.1	Coal Petrography.....	49
4.1.1	Lithotype Results.....	49
4.1.2	Macerals analysis.....	53
4.1.2.1	Vitrinite Maceral Group	55
4.1.2.2	Inertinite Maceral Group	57
4.1.2.3	Liptinite Maceral Group.....	58
4.1.3	Vitrinite Reflectance Measurements.....	60
4.1.4	Mineral Matter	63
4.2	Proximate Analysis	67

4.3	Ultimate and Sulphur Analysis.....	69
4.4	Calorific Value (CV)	72
4.5	Geochemistry	74
4.5.1	Major elements.....	74
4.5.1.1	Comparison of vertical variation of major elements and ash content in borehole JVP20	75
4.5.1.1.1	SiO ₂ , Al ₂ O ₃ , TiO ₂	75
4.5.1.1.2	Mg, Mn, Fe, Ca and S oxides	76
4.5.1.1.3	Na, K and P oxides.....	78
4.5.1.2	Correlations with ash contents and inter-element correlations...	79
4.5.1.2.1	Si, Ti and Al oxides.....	80
4.5.1.2.2	Mg, Fe, Mn, Ca, Na, K, S and P oxides.....	80
4.5.2	Trace elements.....	83
4.5.2.1	Trace element concentrations and vertical variation.....	83
4.5.2.1.1	Trace Elements of Major Concern.....	85
4.5.2.1.2	Trace Elements of Moderate Concern.....	86
4.5.2.1.3	Trace Elements of Minor Concern	87
4.5.2.2	Mode of occurrence of trace elements.....	88
4.6	Summary	91
5	DISCUSSION.....	92
5.1	Coal Petrography.....	92
5.1.1	Macroscopic Character	92
5.1.2	Microscopic Character.....	93
5.1.2.1	Maceral Analysis.....	93
5.1.2.2	Vitrinite Reflectance Measurements (Coal rank).....	94
5.1.2.3	Mineral matter	95
5.1.3	Peat-Forming Condition	97

5.1.3.1	Petrographic facies	97
5.1.3.2	Mineral matter concentration	99
5.2	Coal quality	100
5.2.1	Proximate analysis	100
5.2.2	Ultimate analysis	101
5.2.3	Calorific Value (CV).....	102
5.3	Geochemistry	103
5.3.1	Major elements.....	103
5.3.2	Trace elements.....	106
5.3.2.1	Trace elements concentration.....	106
5.3.2.2	Trace Elements Association with Mineral Matter in Coal	110
5.3.2.3	Trace elements as indicators of peat-forming facies.....	112
5.4	Summary	114
6	CONCLUSION	116
7	Recommendations	119
8	REFERENCES.....	120

List of Figures

Figure 1.1: Map showing the various coalfields of Botswana (Modified after Chatupa, 1991).	2
Figure 1.2: Sub-Basin of the Botswana Kalahari Karoo Basin (after Smith, 1984). 3	
Figure 1.3: Central Botswana Kalahari Karoo Basin and its sub-divisions (Belts) (redrawn after Segwabe, 2008).....	5
Figure 1.4: Location of PL93/2007 in Moiyabana Coalfield, eastern Botswana (Modified after Stephenson and McLean, 1999).	6
Figure 1.5: Stratigraphic scheme with depositional environment for Karoo Supergroup in Botswana (Smith, 1984).	8
Figure 1.6: Geology of Moiyabana area, eastern Botswana (Modified after Ermanovics, 1978).	12
Figure 2.1: Summary of major analytical techniques used in coal petrography (taken from Falcon and Snyman, 1986).	20
Figure 3.1: The JVP20 borehole core – coal horizon only.	44
Figure 4.1: Megascopic representation of JVP20 correlated with Smith (1984) formation and sample locations in the Morupule Formation at Moiyabana Coalfield, Botswana.	52
Figure 4.2: Variation of macerals and mineral matter within coal seams intersected at borehole JVP20, Moiyabana Coalfield.....	53
Figure 4.3: Photomicrographs of vitrinite group maceral from Moiyabana coal samples, under reflected polarised light in transparent oil immersion examination. a) Collotelinite with disseminated pyrite (white spots) and clay (grey/brown). b) Homogeneous band of collotelinite. c) Collotelinite showing banding as differentiated by colour and reflectance. d) Collodetrinite (Cd) forming groundmass in which sporinite and cutinite (liptinite) and inertodetrinite (inertinite) are embedded. e) Collotelinite (Ct) with clay (Cy). f) Corpogelinite (Cg) occurring in association with inertodetrinite (In) and secretinite (Sc).	56
Figure 4.4: Photomicrographs of inertinite macerals from Moiyabana coal samples, under reflected polarised light in transparent oil immersion examination. a) Semi-fusinite. b) Fusinite. c) Secretinite(Sc) associated with semifusinite(SF). d) Fusinite. e) Semifusinite. f) Inertodetrinite.....	58

Figure 4.5: Photomicrographs of liptinite macerals from the Moiyabana coal samples, under reflected polarised light in transparent oil immersion examination. a) Resinite in vitrinite maceral. b) Sporinite (Sp) and cutinite (Cu) macerals types in colloditrinite (Cd) groundmass. c) and d) Liptodetrinite (Ld).....	59
Figure 4.6: Histograms showing distribution of random vitrinite reflectance (Rom %) measured in half V-steps using values from Table 4.3.	62
Figure 4.7: Petrographically observable mineral matter concentrations showing vertical variation.	63
Figure 4.8: Photomicrographs of clay occurrences in the Moiyabana coal samples; under reflected polarised light in transparent oil immersion examination. a) Clay groundmass with stringers of collotelinite. b) Thin bands of clay in collodetrinite. c) Syngenetic clay- disseminated. d) Clay as cell-filling in fusinite.....	64
Figure 4.9: Photomicrographs of quartz (Qz) in association with clay (Cy) from sample G; under reflected polarised light in transparent oil immersion examination.....	65
Figure 4.10: Photomicrographs of calcite occurrences in Moiyabana coal samples, under reflected polarised light in transparent oil immersion examination. a) Detrital calcite-restricted to occur in the organic matter. b) Fracture filling – occurring in veins and/or cleats. c) Epigenetic calcite vein cutting across collodetrinite.....	66
Figure 4.11: Photomicrographs of pyrite occurrences in Moiyabana coal samples, under reflected polarised light in transparent oil immersion examination. a) Epigenetic pyrite in cleats and veins. b) Syngenetic pyrite clustered together in vitrinite. c) Massive pyrite nodule as cell filling cavities. d) Syngenetic pyrite disseminated in vitrinite.....	66
Figure 4.12: Proximate analysis graphs depicting weight (%) loss as temperature (°C) increases.	68
Figure 4.13: Plots of proximate component's variations with coal samples of Moiyabana Coalfield.	69
Figure 4.14: Plots of ultimate constituents content vertical variation with coal samples of Moiyabana Coalfield.	70
Figure 4.15: Fixed carbon compared to carbon on a vertical scale for Moiyabana coal samples.....	71

Figure 4.16: X-Y scattergrams proximate analysis components vs. carbon of Moiyabana coal samples, eastern Botswana.	72
Figure 4.17: Variation relationship between calorific value and proximate parameters for coal samples of Moiyabana Coalfield.	73
Figure 4.18: Variation relationship between calorific value and ultimate parameters in coal samples of Moiyabana Coalfield.	73
Figure 4.19: Plots of calorific value vs. carbon (top) and ash (bottom).	74
Figure 4.20: Plots of major elements content showing positive vertical correlation with ash.	76
Figure 4.21: Plots of major oxides content compared to ash content in vertical scale showing negative correlation with ash yield.	77
Figure 4.22: Plots of total S, SO ₃ and sulphide values in vertical scale.	77
Figure 4.23: Plots of major oxides content showing vertical variation with ash of Moiyabana coal samples.	78
Figure 4.24: Major oxides showing relationship with ash content.	82
Figure 4.25: Plots of mineral matter concentrations and ash yield showing vertical variation in Moiyabana coal samples.	83
Figure 4.26: Plots of trace elements of major concern concentrations showing vertical variation in Moiyabana borehole core, JVP20.	86
Figure 4.27: Plots of trace elements of moderate concern concentrations showing vertical variation in Moiyabana coal samples.	87
Figure 4.28: Plots of trace elements of minor concern concentrations showing vertical variation in Moiyabana coal samples.	88
Figure 5.1: Plot showing the coalification path for Moiyabana coal samples (Bustin et al. (1983) based on van Krevelen, 1961). Values from Table 4.6.	94
Figure 5.2: Coal facies diagram of TPI versus GI showing the peat-forming conditions of the Moiyabana coal samples.	98
Figure 5.3: Ternary diagram illustrating depositional conditions based on the macerals and mineral matter content (after Singh and Singh, 1996).	100
Figure 5.4: Vertical relationship between trace elements.	109
Figure 5.5: Vertical relationship between elements.	110

List of Tables

Table 1.1: Correlation of formations in the Karoo Supergroup of Botswana (from Smith, 1984).....	10
Table 2.1: Macroscopic description of coals in sections and boreholes (reproduced from Thomas, 2013; after Ward, 1984).....	18
Table 2.2: Stopes-Heerlen classification of macerals groups, macerals and sub-macerals of hard coal (reproduced from Thomas, 2013; after Ward, 1984).....	22
Table 2.3: Major stages of the development from peat to meta-anthracite (reproduced from Taylor et al., 1998; after Levine, 1993).	32
Table 2.4: ASTM classification of coal by rank (reproduced from Malvadkar et al., 2004).....	33
Table 2.5: South African Classification using vitrinite reflectance (SANS 11760-2007).....	34
Table 2.6: South African Classification using vitrinite content (SANS 11760-2007).	35
Table 2.7: South African Classification using ash yield (SANS 11760-2007).....	35
Table 2.8: Russian classification of coal by mark (reproduced from Thomas, 2013).....	36
Table 4.1: Lithological succession encountered in borehole JVP20, Moralan area, Moiyabana Coalfield.	51
Table 4.2: Maceral compositions (in vol%) of JVP20 coal samples, Moiyabana Coalfield, eastern Botswana.	54
Table 4.3: Random vitrinite reflectance (RoV) measurements with mean and standard deviation values from Moiyabana Coalfield, Botswana.	61
Table 4.4: Summarised maceral groups and mineral matter data of the Moiyabana coal samples, eastern Botswana.	63
Table 4.5: Results of the proximate analysis and Calorific Value (CV) of Air Dried Coal Seams.	67
Table 4.6: Elemental composition (a.d.b) of Moiyabana coals, eastern Botswana.	70
Table 4.7: Major elements analytical data (weighted %) of Moiyabana coal samples, eastern Botswana.	75

Table 4.8: Example of how to calculate values of Pearson's correlation coefficient for oxides in coal using SiO ₂ and TiO ₂ oxides (after Shao et al., 2004).	79
Table 4.9: Values of Pearson's correlation coefficient for major elements in coal.	80
Table 4.10: Trace elements data (ppm) of Moiyabana borehole core, JVP20 compared to other coals.	84
Table 4.11: Values of Pearson's correlation coefficient for trace elements in Moiyabana coal samples.....	90
Table 5.1: Values of GI and TPI ratios of Moiyabana coal samples.....	97

1 INTRODUCTION

1.1 Background to the study

Coal is a heterogeneous material containing a mixture of organic and inorganic components in varying proportions (Liu et al., 2005; Yossifova et al., 2011). The inorganic sediments comprising of sand and clay in the early stages of coal formation, infiltrate into the vegetal matter and are embedded in the vegetative structure during the process of peatification and coalification. Although the inorganic fraction typically constitutes only a small portion of the coal composition, most of the problems associated with coal are related to this portion (Ward, 2002). By definition, a coal must have less than 50 wt% of mineral matter; greater than 50% the rock is classified as carbonaceous shale. The inorganic matter in coal is a result of several separate processes associated with peat accumulation (Dai et al., 2012; Ren et al., 2006). These inorganic constituents in coal provide not only geological information about depositional conditions and coal-bearing sequence formation (Ren, 1996), but also useful environmental information relevant to pollution control of major, minor, and trace elements during coal mining, cleaning, combustion and utilization (Dai et al., 2008b; Sia and Abdullah, 2011).

Coals in Africa, occurring in Morocco in the north to South Africa in the south, were deposited during varying ages ranging from Carboniferous to the Miocene and all are hosted in sedimentary successions (Hancox, 2016). In southern Africa, the Karoo Supergroup (KSG) hosts all known coal deposits, ranging in age from about 300 to 150 million years before present (Cairncross, 2001). Coal plays a small role in the North African countries due to the wide diversity in the energy mix. Conversely, in the southern African countries, coal is the primary source of energy. It is common knowledge that most countries in southern Africa, including Botswana, South Africa, Zambia and Zimbabwe, depend mainly on coal for their electrical generation. Hancox (2016) in his latest publication (The Coalfields of South-Central Africa: A current perspective) states that South Africa and Botswana, specifically are highly dependent on coal for electricity generation accounting for about 90% in total.

In Botswana, the KSG succession is represented by the Kalahari Karoo Basin (KKB) and the Tuli Basin (Smith, 1984), both estimated to host about 212 billion tonnes of coal resources based on private companies and geological survey reports on coal resource estimations for coalfields in Botswana (Fig. 1.1) (Chatupa, 1991). Meyer (2015) challenged this figure on the basis of improper methods of quantification of these coal reserves. Hancox (2016) acknowledges the fact that it is difficult to properly quantify Botswana's coal resource base, but affirms that it is probably in the order of tens of billions of tonnes. Although private companies were involved in extensive coal exploration, Botswana Geological Survey (currently Botswana Geoscience Institute) has not compiled actual resources figures based on those exploration activities. Some open file information (published) generated by private companies (Golding, 2015) indicates compliant resources in the order of 60 Gt Gross Tonnes *in situ*.

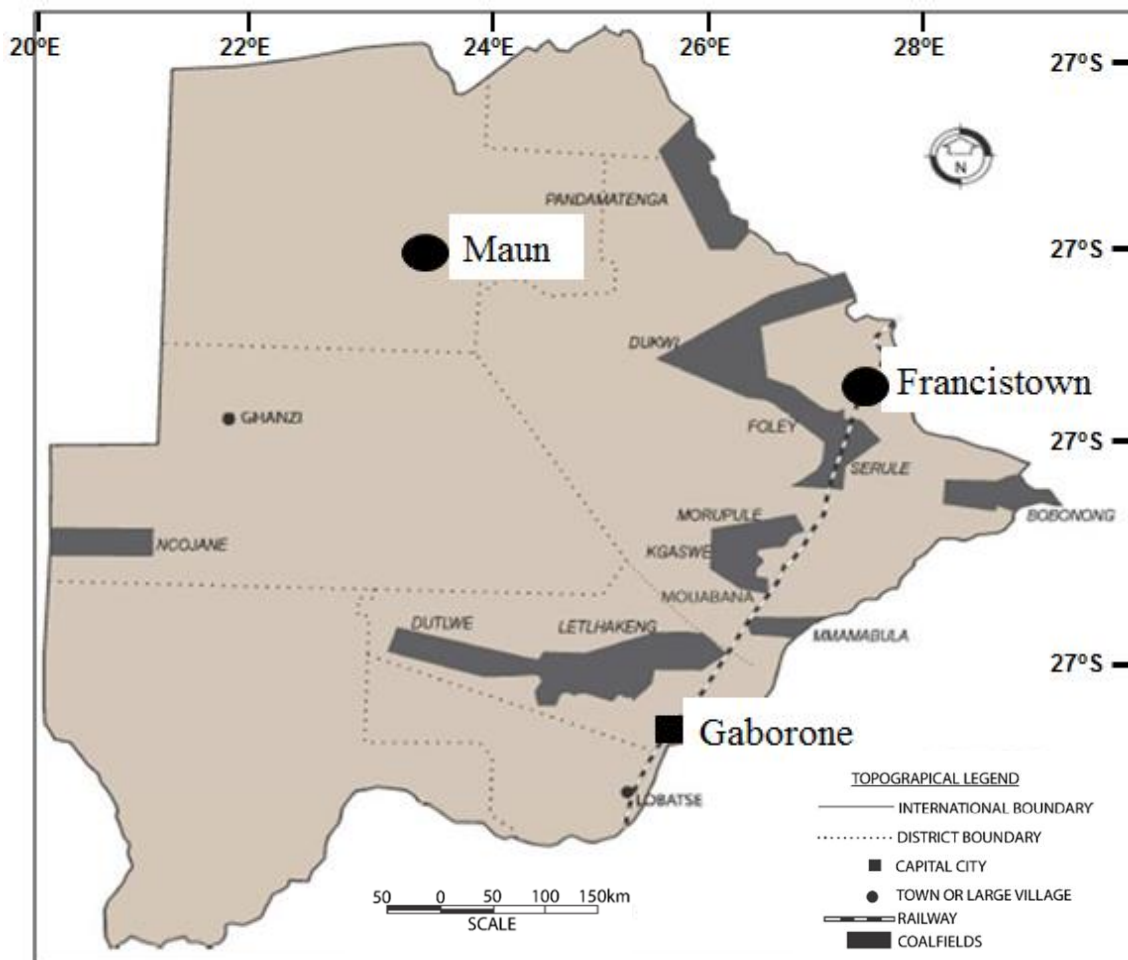


Figure 1.1: Map showing the various coalfields of Botswana (Modified after Chatupa, 1991).

Smith (1984) divided the KKB into four prominent areas of Southwest Botswana, Northeast Botswana, Northwest Botswana and Central Kalahari sub-basin (Fig. 1.2). Because of its huge size, the Central Kalahari sub-basin was further subdivided into four belts mainly for descriptive purposes (Smith, 1984). They include the Western, South-eastern, Southern and Northern Belts (Fig. 1.3). This subdivision is based on lithostratigraphy, involving the correlation between local successions with respect to geological setting and facies changes.

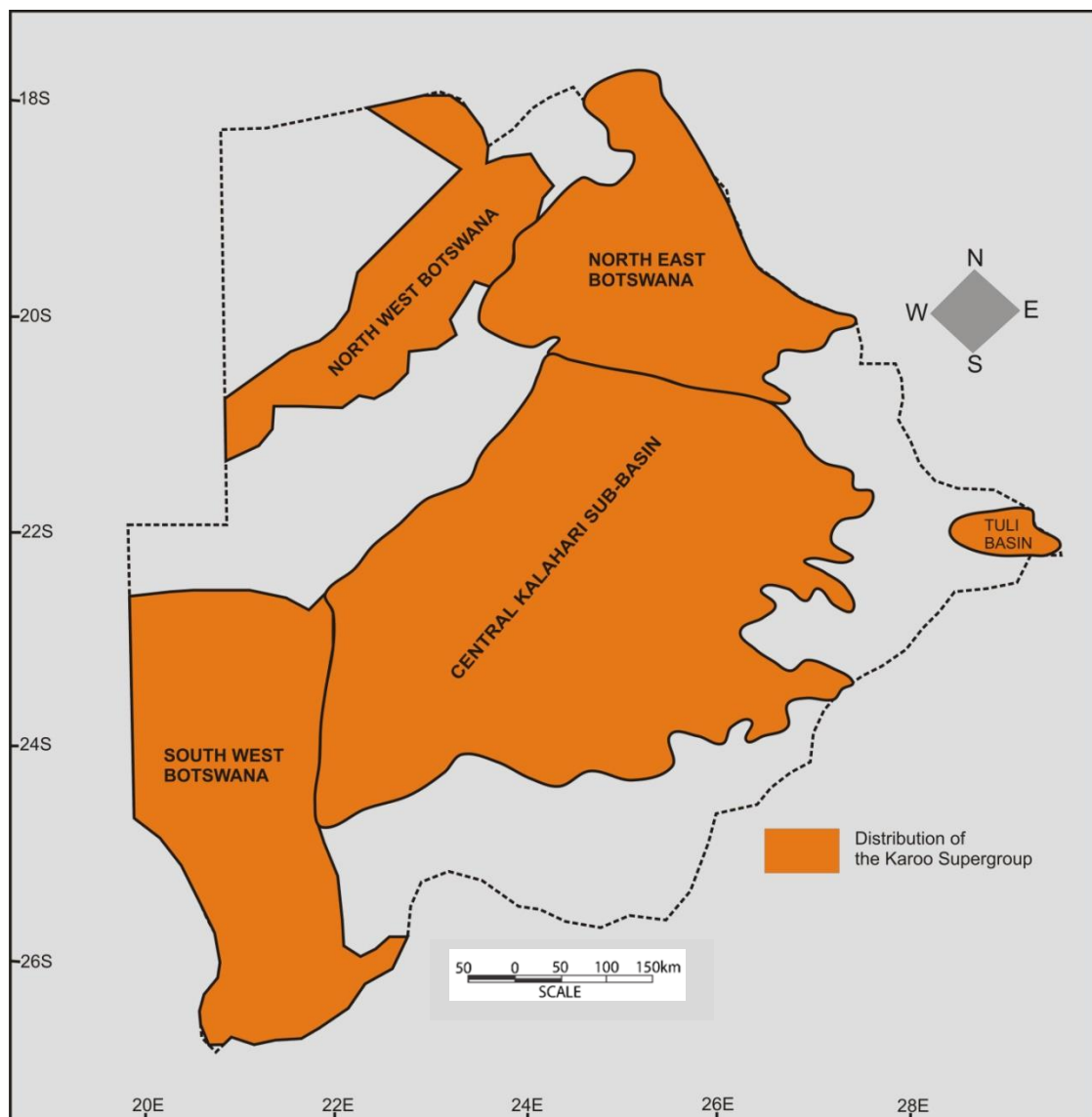


Figure 1.2: Sub-Basin of the Botswana Kalahari Karoo Basin (after Smith, 1984).

This study will focus on Moiyabana Coalfield, which is one of the coalfields located in the South-east belt (Fig. 1.1 and Fig. 1.4). Many of these coalfields, such as Morupule (Kgwaswe), Lechana and Mmamabula Coalfields, have been targeted for coal exploration since the 1970's due to the quality and close to surface coal deposits. Botswana's only operational coal mine is in the Morupule Coalfield. Currently, available published papers on petrography and geochemistry of the various coal seams in the KKB are scarce to absent.

This project adds knowledge to the limited public domain information on coal of Botswana, focusing on the Moiyabana Coalfield. Detailed petrography of organic (type), inorganic (grade), and rank (maturity) of coal was undertaken. Furthermore, geochemistry of major and trace elements in terms of distribution, concentration and mode of occurrence of these elements was also investigated.

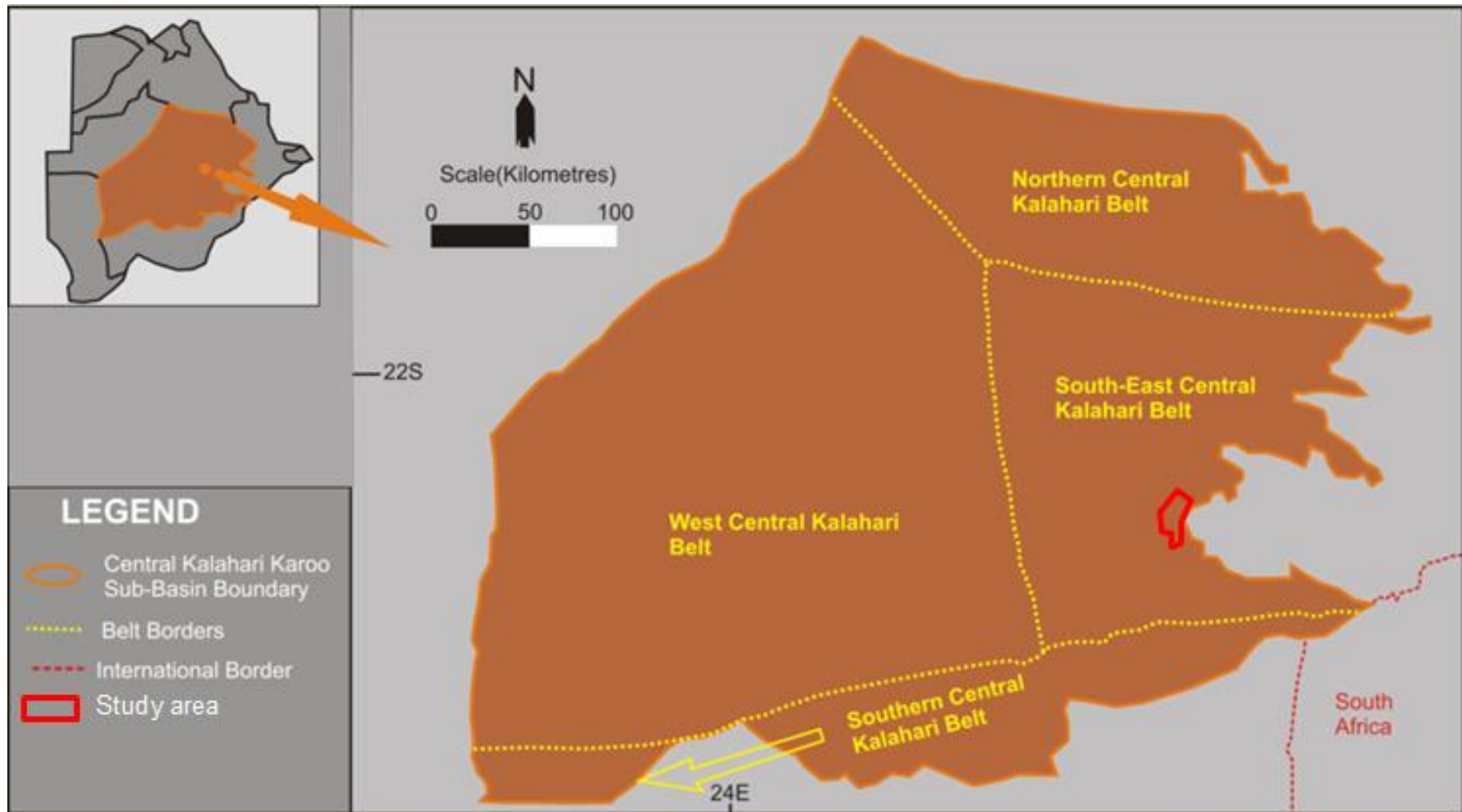


Figure 1.3: Central Botswana Kalahari Karoo Basin and its sub-divisions (Belts) (redrawn after Segwabe, 2008).

1.2 Geographical Overview

The current study focuses on the western part of the Moiyabana Coalfield where Jaquar Venture (Pty) Ltd was allocated prospecting licence PL93/2008 by the Government of Botswana. This area is located in the central district, approximately 10 km to the north of Kodibeleng village and about 70 km northwest of Mahalapye village, situated between longitude 26°00' and 26°25'E and latitude 22°00'S and 23°25'S and covers an area of about 410 km² (Fig. 1.4). Palapye village is located about 100 km northeast of the licence area. Topography of this area is mostly flat land, with only small tributary rivers running from north to south. The area is sandy and only accessible through a 4x4 vehicle during the dry and rainy period of the year. There are no coal seams exposed on the surface, and most of the area is under cover of recent Kalahari Beds, averaging a thickness of about 25 m.

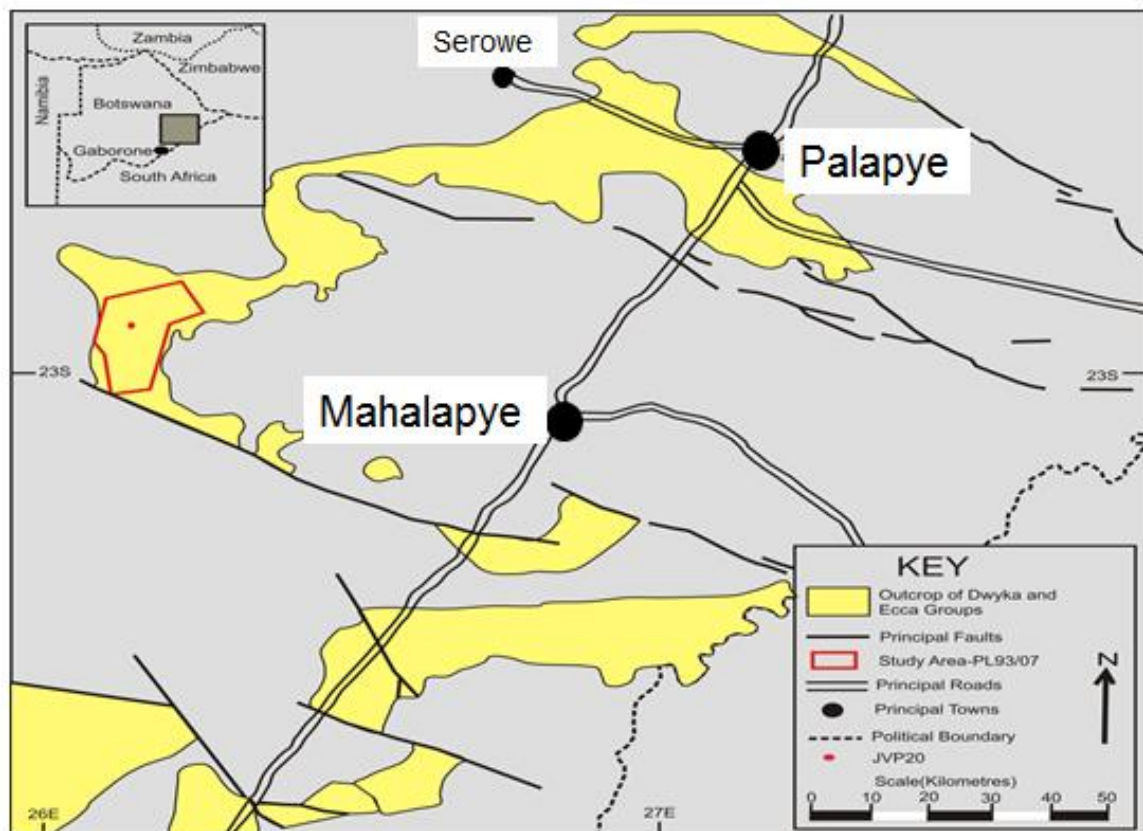


Figure 1.4: Location of PL93/2007 in Moiyabana Coalfield, eastern Botswana (Modified after Stephenson and McLean, 1999).

1.3 General Geology

Coal in southern Africa, including Botswana, occurs in sedimentary rocks of Permian age. These palynologically derived ages generally range between Artinskian to Kungurian and Ufimian to Kazanian for some of the slightly younger coals (Cairncross, 2001) and they are assigned to the KSG (Johnson et al., 1996; SACS, 1980). One of the outstanding geological feature of Botswana is the centrally-located sedimentary basin infilled by Karoo sediments, elongated in NE-SW direction and covers approximately 70% of Botswana's total surface area (Carney et al., 1994). Potentially exploitable areas for coal in Botswana are centred on the eastern and south-western flanks of the KKB (Hancox, 2016), attributed to coals in this region being close to the surface.

Karoo rock structures and stratigraphy in Botswana are derived mainly from coal and diamond exploration work, and geophysical surveys undertaken by both Government of Botswana (through the Department of Geological Survey) and private companies. Earlier work on the KSG of Botswana was pioneered by Smith (1984), Green et al. (1980), Carney et al. (1994) and more recently by Modie (2007) and Segwabe (2008). Smith (1984) established a lithostratigraphic scheme of nomenclature for the KSG in Botswana. He divided the KSG into five groups based on the correlation with Main Karoo Basin (MKB) in South Africa, which include: Dwyka, Eccca, Beaufort, Lebung and Stormberg Lava Groups (Fig. 1.5). Correlations of different formations across the different sub-basins are mainly based on this lithostratigraphic scheme (Table 1.1).

The Dwyka Group forms the basal lithological unit comprising of rocks of glacial origin constituting of varvites, mudstones, siltstones, sandstones and tillites (Carney et al., 1994). This Group is overlain by the Eccca Group which comprises of marginal marine to fluvio-deltaic mudstones and siltstones and coals from swampy environments (Carney et al., 1994; Smith, 1984). All the known coal reserves in Botswana are hosted in this Group. In most of the well-studied sub-basin, the Eccca Group is subdivided into lower, middle and upper Eccca.

Age (Ma)	STRATIGRAPHY	LITHOLOGY	DEPOSITIONAL ENVIRONMENT
185	SLG	Basalt	Continental flood basalts: extensional tectonics related to on-set of Gondwana break-up
225	LEBUNG GROUP	Red beds Sandstone Silt-mudstone Sandstone, rare conglomerates	Arid continental palaeo-climate: Aeolian Fluvial Lacustrine
265	BG	Silt-mud limestone	Transitional (Lacustrine)
278	ECCA GROUP	Mudstone	De-glaciation and amelioration of the climate: Fluvio-deltaic Swamps Lacustrine/marginal marine
290	DWYKA G	Mudstone varvites, siltstone, sandstone, tillites	Glacial palaeo-climate Sub-glacial Glacio-fluvial Glacio-lacustrine

Figure 1.5: Stratigraphic scheme with depositional environment for Karoo Supergroup in Botswana (Smith, 1984).

The Lower Eccca (represented by Makoro Formation in the SE/Morupule column in Table 1.1) commences with argillites and carbonaceous argillites containing rare extra clasts possibly released from ice rafts (Smith, 1984), to poorly sorted fluvio-

deltaic sandstones occurring in upward fining cycles (Green et al., 1980). The Middle Eccca (represented by Kamotaka and Morupule Formations in the SE/Morupule column in Table 1.1) is typical of the delta top and fluvially-dominated delta plain, with the coal progenitors seen as peat swamps that subsequently were buried following channel changes and basin subsidence (Modie, 2007). The Upper Eccca subgroup (represented by Serowe Formation in the SE/Morupule column in Table 1.1) is believed to have accumulated at a time when many of the basin's topographic irregularities had been infilled and when rates of subsidence were more uniform over a greater area (Carney et al., 1994).

Overlaying the Eccca Group is the Beaufort Group (BG) (Fig. 1.5) which is composed of lacustrine mudstones, silty mudstones and/or limestone (Segwabe, 2008). The Lebung Group, above the Beaufort Group, comprises lacustrine, fluvial and aeolian to arid continental palaeo-climate siltstone and sandstone with rare mudstone (Smith, 1984). The Lebung Group is overlain by the Stormberg Lava Group (SLG) (Fig. 1.5) comprising of lavas from the continental flood basalts resulting from volcanism related to extensional tectonics during the breakup of Gondwana (Carney et al., 1994).

The general geology of the Moiyabana area is shown in Figure 1.6 and the lithostratigraphy of the study area, described by Smith (1984), is illustrated in Table 1.1 under SE Kalahari/Morupule belt column. At the bottom of the stratigraphy is the Dukwi Formation (the basal unit), which consists of tillites, diamictites, conglomerates and laminated siltstones and mudstones. The Dukwi Formation belongs to the Dwyka Group, which is overlain by the Eccca Group comprising of Makoro, Kamotaka, Morupule and Serowe Formations (Table 1.1). The Makoro Formation comprises of interbedded silty mudstones and siltstones with rare sandstone. The Kamotaka Formation overlaying Makoro comprises of coarse grained to pebbly sandstone that can have a maximum thickness of 40 m. Very thin coaly units of about 10 cm to 1 m are usually present in this formation. The Morupule Formation overlies the Kamotaka Formation and has thicknesses to a maximum of 70 m, and consists of carbonaceous mudstone, shale, coal and rare medium- to coarse-grained sandstone. The Morupule Main seam (currently mined at Morupule Mine), upper Morupule seams and Taukome bright seam are

Table 1.1: Correlation of formations in the Karoo Supergroup of Botswana (from Smith, 1984).

GROUPS	SOUTH WEST BOTSWANA	KWENENG AND WESTERN CENTRAL KALAHARI	MMAMBULA	MORUPULE AND SE KALAHARI	NORTH EAST BOTSWANA AND NORTHERN BELT	NORTH WEST BOTSWANA	TULI BASIN
STORMBERG LAVA	STORMBERG LAVA GROUP (UNDIVIDED)						BOBONONG LAVA FORMATION
LEBUNG	NAKALATLOU SANDSTONE	NTANE SANDSTONE FORMATION				BODIBENG SST FM	TSHEUNG SST FM
						SAVUTI FM	THUNE FM
	DONDONG FM	MOSOLOTSANE FORMATION			(North east only) NGWASHA FM		KOREBO FORMATION
					PANDAMATENGA FM		
BEAUFORT	KULE FM	KWETLA FM	TLHABALA FORMATION			? MARAKWENA FM TALE FM ?	SESWE FORMATION
ECCA	OTSHE FM	BORITSE FM	BOROTLO FM	SEROWE FM	TLAPANA FM	?	MOFOIAHOGOLO FORMATION
			MMAMABULA FM	MORUPULE FM			
		KWENENG FM	MOSOMANE FM	KAMOTAKA FM	MEA ARKOSE FM		
	KOBE FM	BORI FM	BORI FM	MAKORO FM	TSWANE FM		
DWYKA	MIDDLEPITS FM KHUIS FM MALOGONG FM	DUKWI FORMATION				?	

contained within the Morupule Formation. The Tacoma bright seam forms the upper contact with the Serowe Formation, which consists mainly of siltstones and coals. There is a well-defined and consistent “siltstone marker” above the Taukome bright seam. The Serowe bright seam forms the top boundary of Serowe Formation.

Belonging to the Beaufort Group, the Thabala Formation overlies the Serowe Formation. This comprises almost entirely of non-carbonaceous brown to grey mudstones. The Mosolotsane Formation overlies the Thabala Formation, and constitutes the lower part of the Lebung Group, which consists of coarse to medium grained sandstone and various sequences of red-brown mudstone, siltstone and fine sandstone. The upper part of the Lebung Group, Ntane Sandstone Formation, overlying the Mosolotsane Formation, consists of a uniform sequence of fine to medium red-brown sandstones of aeolian origin. Although the Stormberg lavas are rarely intersected, they occur as dark purple grey amygdaloidal basalt. At the top of these units are Kalahari beds, which usually comprise of sand, clay soils, calcareous conglomerates, calcretes and silcretes of up to 20 m thickness.

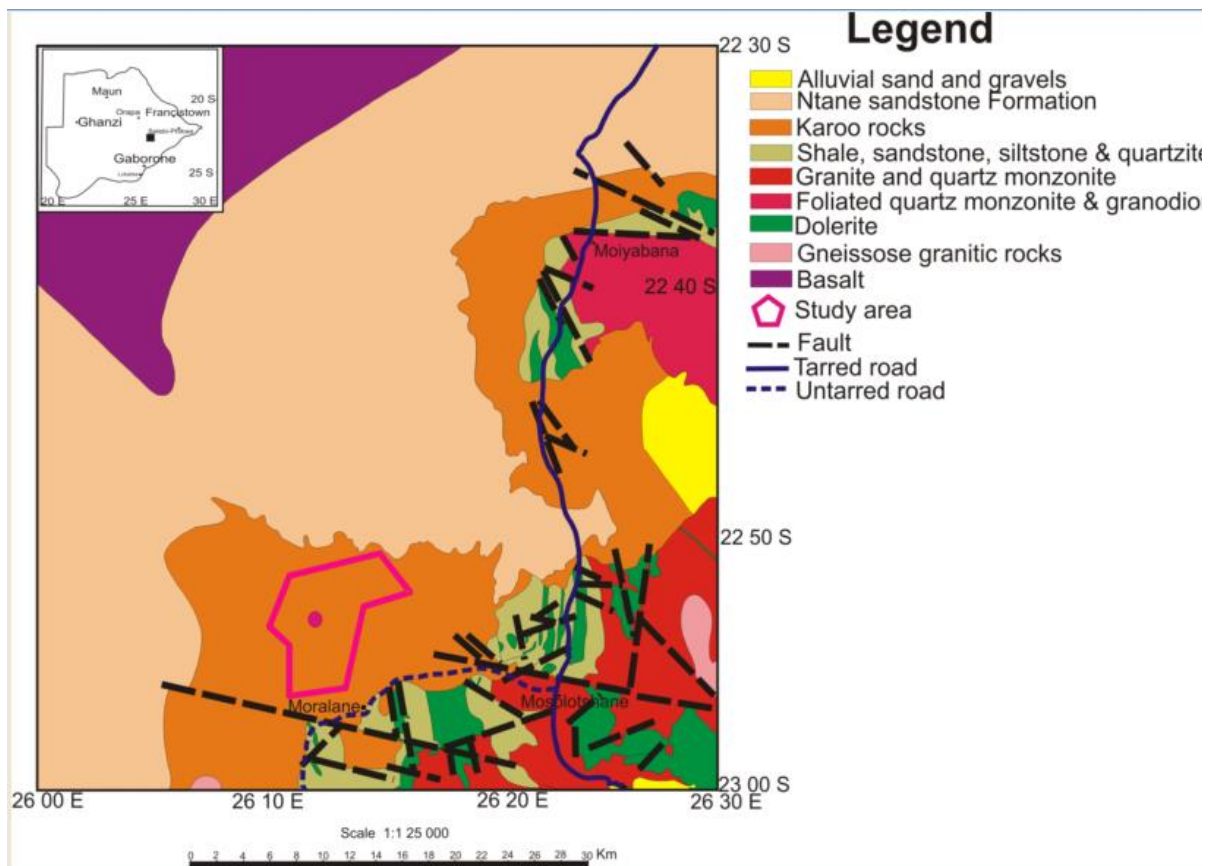


Figure 1.6: Geology of Moiyabana area, eastern Botswana (Modified after Ermanovics, 1978).

1.4 Previous Work

Although the occurrence of coal in Botswana has been known since early 1930's (Hancox, 2016), it was only around mid-1970's to late 1980's that Shell Coal Botswana (Pty) Ltd carried out extensive exploration in the Moiyabana Coalfield and other areas which includes Foley, Morupule, Mmamabula, Lechana, Dukwe and Pandamatenga over an initial area of 3 550 km² (Shell Coal, 1977). After the completion of the aforementioned drilling campaign by Shell, it was established that there is very little outcrop of the Karoo sequence in the Moiyabana Coalfield. Therefore, the geology of this area is determined from the drilling.

Further knowledge on the Moiyabana West Coalfield was obtained by Ermanovics (1978), who worked on the geology of the Moiyabana area (Fig. 1.6). It was estimated that the thickness of the Karoo sequence in the west side of Moiyabana Coalfield was at least 365 m thick. Smith (1984) continued the work in this area by

correlating coal seams from this area with others occurring in KKB and relating them to the MKB in South Africa.

Current licence holder (Jaquar Ventures Pty Ltd) and Hodges Resources Ltd jointly explored the area from 2008 to 2012. The objective of the drilling campaign by the joint venture in this area was to confirm the resources previously reported by Shell in 1977. In 2012, Hodges Resources withdrew its involvement in the exploration activities at Moiyabana Coalfield, leaving Jaquar Ventures to retain 100% of prospecting licence PL93/2008. To date, Jaquar Ventures Pty Ltd is the last company to carry out the latest drilling campaign in September 2014. Borehole JVP20 is part of the last drilled cored boreholes from PL93/2008 as part of the shallow coal of Moiyabana West Coalfield.

1.5 Aim and Objectives

The aim of this study was to undertake a detailed investigation on the Morupule Formation coal seams deemed profitable at Moiyabana Coalfield from a single borehole core to obtain an understanding of coal petrography and geochemistry. It is very important to clearly state that by the use of only one borehole core samples in this study means that the results are not intended to represent the coalfield, rather to indicate coal quality associated with that core only, but could be used to infer coal quality of the area.

The petrographic component of coal is important in that it ascertains coal quality, composition, and coal rank (Taylor et al., 1998) of a given coal sample. This study will utilise maceral constituents to aid in the reconstruction of the palaeo-environmental conditions of deposition and to assess the coal quality of the coal samples. Vitrinite reflectance will be used in quantifying rank of the Western Moiyabana coal samples. The geochemical analysis results obtained in this study will be used to understand the vertical distribution of ash in relation to major and trace elements occurrence in the coal samples of Moiyabana Coalfield. The mode of occurrence of trace elements in particular, is vital as some trace

elements, if they are not determined, may have detrimental effects during coal utilisation.

Therefore, this dissertation has the following objectives:

- (1) To characterize the constitution, rank and economic potential of individual Morupule Formation coal seams at Moiyabana Coalfield from a single borehole core.
- (2) To delineate the vertical distribution of ash content in relation to organic matter and geochemical components.
- (3) To establish and delineate the modes of occurrence of the trace elements in the coal, and to quantify the percentages of these occurrences for each specific trace elements therein.
- (4) Lastly, reconstruction of the paleo-environmental conditions of deposition which prevailed during formation of the Morupule Formation coal seams.

1.6 Organisation

This dissertation is arranged into six individual chapters with each focusing on a specific area of this research project. Chapter 1 contains background information, geographical overview, general geology, previous work and closing with aims and objectives. Chapter 2 details literature review regarding all the relevant components which are important for the overall achievement of the project. Chapter 3 deals with all the employed methods during the sampling phase and the analytical procedures used in this project. Chapter 4 encompasses all the data collected during the analyses carried out in this project, which are interpreted and discussed in Chapter 5. Chapter 5 is the discussion section of this study. Finally, Chapter 6 provides a summary of the important conclusions derived from this study.

2 LITERATURE REVIEW

In this chapter the literature review discusses the characteristics of coal for technological utilisation purposes based on the petrographic, chemical and geochemical components. Although the literature covers a wide variety of parameters with regard to petrography, chemical analysis and geochemistry of coal, this review will focus on five major themes which emerge repeatedly throughout the past work on this topic. These themes include: coal petrography, coal quality, coal rank, coal classification and elements occurrence in coal. The methods, procedures and techniques employed during analysis on the above topics will be reviewed. Lastly, a review of some environmental hazardous trace elements during coal combustion will be covered.

2.1 Coal Sampling

In collection of geological sample, which is represented by a body of material removed by approved methods, should be protected against any form of contamination, and subsequent testing should primarily involve determination of the nature, composition, and percentage of specified constituents in a given sample. If not carried out properly, sampling could result in wrong analytical data. With regard to coal analysis, different sampling protocols are used for different types of coals based on the ultimate usability of coal (Thomas, 2013). For producing reliable data, caution should be taken to avoid contamination during coal sampling (Speight, 2005). As coals are often heterogeneous in nature (Speight, 1994), Gould and Visman (1981) strongly emphasise on the necessity to obtain a representative sample for analysis based on the coal composition variability. However, sampling protocols for coal utilisation are outlined in available coal standards (SABS / ISO / ASTM) for overall reliable data.

The storage of coal prior to analysis is very important as any chemical/physical alterations due to weathering of coal after completion of sampling procedure might lead to degradation of coal quality. Speight (2005) emphasises that the reliability of the storage procedure is the degree to which the coal sample remains

unchanged. This is achieved by storing coal samples in closed plastic bags to avoid moisture loss and rainwater infiltrating the collected samples.

2.2 Sample preparation

Most of the countries and companies use different sample preparation based on the type of coal and where it is found. The differences in coal types lead to different international and national coal standards (Thomas, 2013). Although different coal types are involved, the usability of that coal determines the sampling preparation to be used (e.g. thermal, metallurgical). Generally, sampling preparation encompass drying, crushing, and thoroughly mixing of crushed sample particles to achieve unbiased sample. Speight (1994) cautiously warns not to compromise standards, but advocate for rapid coal preparation steps and in as few steps as possible, to minimize both water loss and oxidation of the coal.

The nature of the test methods to be applied during sample preparation depends mainly on the particle size distribution. A wide distribution of various particle size is understood to cause segregation (Speight, 2005). Based on the above contention by Speight (2005), Thomas (2013) concluded that crushing and/or grinding coal from a large particle to a very small particle should involve a reasonable number of steps that are based on the starting particle size and nature of the coal.

2.3 Coal Petrography

Although very old, petrographically the Stopes–Heerlen system classifies coals (humic and sapropelic) macroscopically into lithotypes based on visual observation of features greater than 3 mm (Malvadkar et al., 2004). Microscopically, the use of microscopy to study both organic and inorganic constituents in coal and the degree of metamorphism to which they have been subjected subsequent to their time of burial is accepted (Falcon and Snyman, 1986). This technique utilises coal properties which can be directly correlated to the petrographic constituents in coal, making it possible to predict the technological behaviour of the studied coal sample.

2.3.1 Macro-petrography

Lithotype refers to different macroscopically identifiable layers in coal seams (Thomas, 2013). They are used as the principal basis for coal resource and reserve determination, but it is not applicable in predicting specific coal properties such as ash content and coal rank (Malvadkar et al., 2004).

2.3.1.1 Humic coal

Humic coals are mainly heterogeneous and composed of a diversified mixture of macroscopic plant debris (Thomas, 2013). The Stopes–Heerlen system proposed four lithological types for describing humic coals (Table 2.1). Lithotypically, the system describes them as follows: 1) vitrain as black, glassy, vitreous material with a bright lustre, occurring as thin bands and usually brittle. It is a feature characterising most humic coals; 2) clarain as bright with a silky lustre between vitrain and durain, and occurs in fine laminations; 3) durain as grey to black with dull lustre and fractures into rough surface fragments. It is imperative to note that only lenses thicker than 3-10 mm are referred to as durain and; 4) fusain as black, soft, and friable and easily disintegrates into a black fibrous powder. In coals, fusain occurs as lenses concentrating in the discrete layers, which are usually several millimetres thick.

In the commercial usage of coal, very thin coal layers/lenses in borehole cores and exposures present difficulties for the strict use of the terms, as mentioned above, to describe coals (Thomas, 2013). In the practical use of coal, only a limited number of lithologically distinct units are required. As coal is used mainly for commercial purposes, various sources (AS 2418-1995; McCabe, 1984; Schopf, 1960), have proposed an alternative terminology that, although essentially retaining the basic classification, has a more descriptive lithological bias as shown in Table 2.1.

In the USA, Schopf (1960) introduced the term attrital coal whereupon he subdivided vitrain and fusain into five levels of lustre ranging from bright to dull (Table 2.1). The Australian system (AS 2418-1995) is broadly similar in approach,

but more descriptive in terminology as it defines vitrain and fusain as bright and dull coal respectively (Table 2.1).

Table 2.1: Macroscopic description of coals in sections and boreholes (reproduced from Thomas, 2013; after Ward, 1984).

Coal Type	Stopes (1919)	Schopf (1960) ASTM Standard	Australian Standard
Banded (Humic) coal	Vitrain	Vitrain	Coal, bright
			Coal, bright, dull bands
	Clarain	Bright	Coal, dull and bright
		Moderately bright	Coal, mainly dull with numerous bright bands
	Durain	Attrital mid-lustre coal	Coal, dull minor bright bands
		Moderately dull	
		Dull	
	Fusain	Fusain	Coal, dull
Non-banded (Sapropelic) coals	Cannel coal	Cannel coal	Coal, dull conchoidal (cannaloid)
	Boghead coal	Boghead coal	
	Impure coal	Bone coal	Coal, stoney or shaley
		Mineralised coal	Coal, heat altered
			Coal, weathered

2.3.1.2 Sapropelic coal

Defined as homogenous in nature, sapropelic coals are composed of a restricted variety of microscopic plant debris (Thomas, 2013), which are believed to have formed from the biological and physical degradation products of coal peat-forming environments. Sapropelic coals are classified into two main types, cannel and boghead coal. Based on Thomas (2013), cannel coal is described as black and dull, homogenous and breaks with a conchoidal fracture, whereas boghead coal is described as algal. Macroscopically, all sapropelic coals essentially look similar and can only be readily distinguished microscopically (Malvadkar et al., 2004).

2.3.2 Micro-petrography

2.3.2.1 Coal preparation for micro-petrography

The use of microscopy, carried-out by the use of reflected white light, on a polished surface of high quality under oil immersion summarises coal micro-petrography analysis. The crushed material (coal particles) is used to prepare

polished epoxy mounted samples, which are then polished to obtain a polished surface ready for petrographic analysis (Falcon and Snyman, 1986). It should be emphasised that polished surface of the coal pellets must be of high quality and mechanisms in place to procedurally check the surfaces of the blocks for quality purposes. The procedures of sample preparation for this type of analysis are standardized in SABS ISO 7404-2 and ASTM D2797-2007.

2.3.2.2 Analytical Techniques in micro-petrography

The analytical techniques usually carried-out involve the use of conventional microscopically investigations in the reflected light at a total magnification of between 250 and 500 (Falcon and Snyman, 1986). The equipment mostly deployed in petrographic analysis is an ore microscope, with photomultiplier, digital read-out and where possible, a computer attachment for the processing of data (Malvadkar et al., 2004). The type (organic component) of coal is determined petrographically by group macerals analysis following the standards test method SABS ISO 7404-3 and ASTM D2799-2009. The rank (coal maturity) is measured by means of vitrinite reflectance following standard SABS ISO 7404-5.

2.3.2.2.1 Maceral analysis

The maceral analysis in coals based on Falcon and Snyman (1986) descriptions, is performed using optical microscopy with a point-counter coupled to the microscope stage. Based on the recorded number of total points, the microscope stage is moved in a series of fixed intervals across the whole sample (Fig. 2.1). Suárez-Ruiz (2012) acknowledges the importance of white light in maceral analysis, and recommends supplementary observations in fluorescence mode. Maceral analyses are carried out under oil immersion using various oil-immersion objectives. A minimum of 500 points are counted in maceral analysis. Although it depends on the purpose of the analysis, visible minerals may be ignored, counted collectively or individually (Falcon and Snyman, 1986). Thomas (2013) observes that although the automated methods using computerized image analysis were developed in the last decades for maceral analysis, the manual point-counting method is extensively used.

2.3.2.2.2 Vitrinite reflectance measurements

In reviewing literature regarding vitrinite reflectance measurement, it stands out from many publications that incident white light microscopy is used to measure the optical parameter that serve to describe the degree of coalification reached by a coal following the standard test methods SABS ISO 7404-3, and ASTM D2798-2009. By measuring the homogenous bands of collotelinite, reflectance measurements are taken on the surface of vitrinite particles and carried out in monochromatic green light by means of a photomultiplier (Falcon and Snyman, 1986). As a precaution, half an hour is enough for all electrical components to be switched on before instrument calibration procedure is performed.

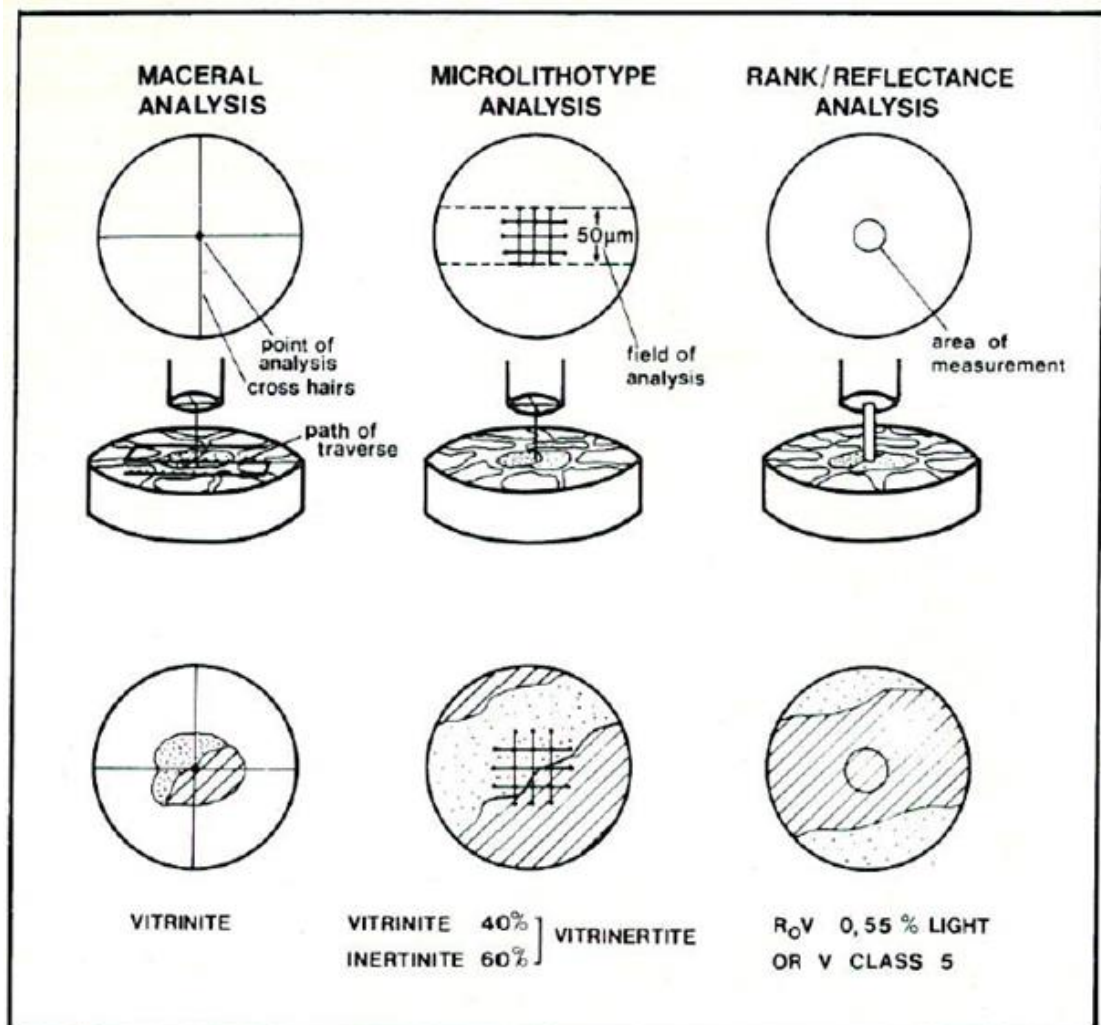


Figure 2.1: Summary of major analytical techniques used in coal petrography (taken from Falcon and Snyman, 1986).

As outlined by Suárez-Ruiz (2012), two distinct reflectance measurements include: 1) maximum reflectance – typically used in metallurgical assessments, performed with the polarizer in the 45 degrees position with the microscope stage rotated through 360 degrees and the maximum reading recorded; and 2) random reflectance – mainly for combustion assessments, performed with no rotation of the microscope stage and the polarizer removed from the optical path of the microscope (Falcon and Snyman, 1986). A range between 50 to 100 readings is taken on vitrinite bands which fall within the graticule area of measurement. Such areas of measurement are encountered along traverses 0.5 mm apart and with steps of 0.5 mm along the horizontal cross lines (Fig. 2.1). As a rule, reflectance of any vitrinite grain should not be measured more than once. Calibration of the instrument should take place before and after the measurements of every sample.

2.3.2.3 Microscopic description of coal

The original classification of maceral groups was first described by the Stopes-Heerlen System (Thomas, 2013) as depicted in Table 2.2. The three main maceral groups recognised by this system are vitrinite, liptinite, and inertinite. In this classification, the macerals groups are described as follows: vitrinite as derived from coalfield plant tissues and gels; liptinite as formed from plants protective coatings, reproductive organs, and resinous cell fillings; inertinite produced from oxidation of vitrinite and liptinite precursors (Malvadkar et al., 2004). This system further divided groups into macerals and subsequently to submacerals (Table 2.2).

Table 2.2: Stopes-Heerlen classification of macerals groups, macerals and sub-macerals of hard coal (reproduced from Thomas, 2013; after Ward, 1984).

Maceral Group	Maceral	Sub-maceral
Vitrinite	Telinite	Telocollinite
		Gelocollinite
	Collinite	Desmocollinite
		Corpocollinite
Exinite (Liptinite)	Sporinite	
	Cutinite	
	Suberinite	
	Resinite	
	Alginite	
	Liptodetrinite	
	Fluorinite	
	Bituminite	
	Exudatinite	
Inertinite	Fusinite	
	Semi-fusinite	
	Macrinite	
	Micrinite	
	Sclerotinite	
	Inertodetrinite	

2.3.2.4 Mineral content of coals

In defining mineral matter in coal, Speight (2005) describes it as those inorganic constituents in coal including all the elements that are not part of the organic coal substance. On the other hand, Thomas (2013) defines mineral matter as the non-combustible inorganic fraction in coal, which is made up of minerals that are either detrital or authigenic in origin, and which are introduced in coal in first or second phases of coalification. Thomas (2013) further suggests that the minerals transported and deposited in the peat swamp by wind and water be called detrital. The remaining minerals, all of which formed in place (authigenic), are divided into those that formed contemporaneously with coal formation (syngenetic) and those whose formation followed the initial stages of coalification (epigenetic).

The mineral matter in coal can be classified broadly as inherent, included and adventitious (Taylor et al., 1998). Based on Taylor et al. (1998) descriptions; inherent mineral matter as the inorganic constituents closely associated with the organic matter in coal, making it very difficult to separate it by methods available at present; included mineral matter as that which forms an inorganic entity within pulverized coal particles; and adventitious mineral matter as the inorganic material that is less associated with the organic matter in coal, in that making it easy to be

separated from coal. Taylor et al. (1998) state that the occurrence of mineral matter in coal seams takes different forms. They outline the common forms as layers, nodules, fissure fillings, rock fragments, and small particles finely distributed throughout coal seams.

2.3.2.5 Depositional environment models

2.3.2.5.1 Petrographic Facies

The petrographic facies with the use of maceral proportions in coal provides a wealth of information with regard to organic source materials relating to the accumulation of peat and conditions that prevailed during coal deposition (Kalkreuth et al., 1991). In the determination of the depositional environment through the analysis of macerals, facies-critical macerals are used as indicators for the palaeo-environment during the early stage of coal formation (Kalkreuth and Leckie, 1989). In an analysis of Australian Permian coals, Diesel (1986) used the Gelification Index (GI) and Tissue Preservation Index (TPI) ratios to determine the depositional environment. Many workers (Casareo et al., 1996; Kalkreuth et al., 1991; Marchioni et al., 1994) following Diesel (1986) fundamental principle, used this method in other geological areas. The GI and TPI are calculated from the formulae given below:

$$GI = \frac{\text{Vitrinite} + \text{Macrinite}}{\text{Semifusinite} + \text{Fusinite} + \text{Inertodetrinite}}$$

$$TPI = \frac{\text{Telinite} + \text{Collotelinite} + \text{Semifusinite} + \text{Fusinite}}{\text{Collodetrinite} + \text{Macrinite} + \text{Inertodetrinite}}$$

2.3.2.5.2 Mineral matter proportions

In his study of wet and dry conditions in association with the rate of subsidence, Goodarzi (1985) comprehensively investigated the importance of mineral matter in coal with regard to the environmental reconstruction. Using the basic principle followed by Goodarzi (1985), Singh and Singh (1996) determined the depositional environment by combining the proportions of both macerals and mineral matter. In

his model, factors that directly affect the amount of mineral matter in coal such as the influx of surface water in the swamp, aeolian sediment blown into the swamp and to a lesser extent magmatic input through faults (dolerite dykes) as a source of minerals into the swamp (Smith, 1984) are taken into consideration.

2.4 Coal Quality

By reading some publications, a simple but precise definition of coal quality could be understood as those chemical and physical properties of coal that influences its potential use. Chemical analyses (proximate, ultimate and calorific value) are the fundamental components for coal quality characterisation as far as the coal utilisation in the technological industry is concerned. The techniques, methods and procedures employed to analyse for coal quality based on the mentioned components will be described in this section.

2.4.1 Chemical properties of coal

The four components which are determined in proximate analysis include; moisture, volatile matter, fixed carbon and ash contents. In the ultimate analysis, carbon, hydrogen, oxygen (usually estimated by difference), nitrogen and sulphur elemental composition is determined (Speight, 2005). Calorific value determines the heating value of coal.

2.4.1.1 Proximate analysis

Speight (2005) defines the four proximate parameters as follows: 1) total moisture as the loss in weight under rigidly controlled conditions (i.e. temperature, time, and airflow); 2) volatile matter as all expelled substances due to thermal decomposition of the coal in the standard crucible under the controlled conditions; 3) fixed carbon is calculated from the addition of the other three parameters then subtracted from 100; and 4) ash as that inorganic residue that remains after combustion.

2.4.1.1.1 Proximate analysis test methods

With regard to test methods based on literature review, ASTM D3172-2007 standard test method repeatedly shows predominance in terms of proximate analysis. In this method, all the four components of proximate analysis are determined at the same. The actual analysis involves subjecting the coal sample to prescribed temperature levels under prescribed time intervals for determination of the proximate parameters (Speight, 2005). This method determines moisture content at low temperature, volatile matter at increased temperature than moisture, ash at high temperature than volatile matter and fixed carbon as the difference of these three values summed and subtracted from 100.

2.4.1.1.2 Proximate analysis as coal quality component

Proximate analysis parameters are very important in the overall usability of coal. Taylor et al. (1998) outline the individual importance of these parameters as follows: 1) moisture content - plays a very important role in buying/selling of coal based on the moisture holding capacity of coal. Very high moisture holding capacity in coal is not desired in the commercial use of coal (e.g. during coal combustion); 2) volatile matter content - in choosing the best match between a specific type of coal burning equipment and the coal to use with the equipment; 3) fixed carbon content - in determining the efficiency of coal-burning equipment; and 4) the ash data -allow engineers to predict the frequency and quantity of ash disposal.

2.4.1.2 Ultimate analysis

The determination of carbon in ultimate analysis represents all carbon present as organic carbon occurring in the coal substance (Speight, 2005). The carbon bonded in carbonates is also incorporated during analysis. Speight (2005) describes the hydrogen determination in coal to involve all hydrogen present in the organic materials and all hydrogen associated with water. Nitrogen is determined from the organic matrix of coal. Sulphur determination involves all forms of sulphur occurring in coal, and oxygen is determined from all inorganic

and organic types present in coal (Speight, 2005). If the value of oxygen is to be calculated from other elements in ultimate analysis, it is of great importance to add other elements that have direct bearing to the usability of coal, such as phosphorus, chlorine, forms of sulphur and selected trace elements (Taylor et al., 1998).

2.4.1.2.1 Ultimate analysis test methods

A common standard test method for determining collectively all elements in the ultimate analysis is called ASTM D3176-2002. Other test methods exist for analysing elements in the ultimate analysis individually.

Carbon and Hydrogen: Regarding carbon and hydrogen, the generally accepted methods are ASTM D3178-1997; ISO 609-1996; and ISO 625-2003. These methods involve determination of carbon and hydrogen contents through combustion in a closed system. The resulting carbon dioxide and water are collected in an absorption train.

Nitrogen: Different methods of nitrogen determination exist in various laboratories across the world and they include; Kjeldahl, Kjeldahl–Gunning and Dumas methods. According to Speight (2005), these methods can be summarised as follows;

The Kjeldahl method: involves nitrogen determination where pulverized coal is boiled with concentrated sulphuric acid containing potassium sulphate and a suitable catalyst to reduce the time for digestion. This common method follows standard test methods ASTM D3179-2002 and ISO 333-1997.

The Kjeldahl–Gunning method: nitrogen present in the coal sample is converted into ammonium salts by the destructive digestion of the sample by a hot mixture of concentrated sulphuric acid and potassium sulphate. Speight (2005) warns that proper precaution should be taken in carrying out this procedure, especially the digesting and distillation steps. This method follows the standard test methods ASTM D3179-2002; and ISO 333-1997.

The Dumas method: nitrogen determination consists of an oxidation method in which a mixture of coal and copper oxide is heated in an inert atmosphere to produce carbon dioxide, water, and nitrogen. Although the Dumas method has been employed for many years, various modifications have been made to increase accuracy and precision.

Sulphur: Determination of sulphur is an important aspect in coal utilization, hence, there are considerable amounts of published works relating to the development of methods to improve the efficiency of the techniques as well as improve the accuracy and precision of the sulphur determination (Gorbaty et al., 1992). Total sulphur analysis follows standard test methods ASTM D3177-2002 and ASTM D4239-2002.

The three most widely used test methods for sulphur determination based on Speight (2005) work are summarised as follows: (1) the Eschka method – involves converting all types of sulphur in the coal (e.g., pyrite, sulphates, sulphides, organically bound sulphur) to sodium sulphate which in the process is converted to water-insoluble barium sulphate. It is then filtered, dried, and weighed; (2) the bomb washing method – the determined sulphur is precipitated as barium sulphate from the oxygen bomb calorimeter washings following test methods ASTM D2015-2007 and ASTM D3286-2002; and (3) the high-temperature combustion method - the sample is burned in a tube furnace and sulphur oxides are collected and quantified by titration using standard test method ASTM D4239-2002.

Problems encountered during the sulphur analyses in coal include non-uniform distribution of pyrite particles, failure to convert all the sulphur to sulphate, and loss of sulphur as sulphur dioxide (Speight, 2005). The best mitigation for the non-uniform distribution of pyrite is provided by Speight (2005), who advocate for many sample increments to be collected to ensure that the gross sample is representative of the coal in question. As for the other two problems, no clear mitigation strategies are in place this far in the existing literature.

Oxygen: The occurrence of oxygen in both organic (i.e. hydroxyl, carboxyl, methoxyl and carbonyl groups) and inorganic (i.e. forms of moisture, silicates, carbonates, oxides, and sulphates) fractions makes it difficult for oxygen to be determined during ultimate analysis (Taylor et al., 1998).

A widely used standard test method (i.e. ISO 1994) for direct determination of oxygen involves pyrolyzing coal in the presence of nitrogen, and then followed by passing the volatile products over a hot platinized carbon at a temperature of about 1100°C. On passing through the volatile products, oxygen is converted to carbon monoxide (Gluskoter et al., 1981). In order to calculate the oxygen in the original sample, the iodine and carbon dioxide released during oxidization of the carbon monoxide can both be determined titrimetrically and gravimetrically respectively (Speight, 2005). Due to problems caused by oxygen various sources, the original procedure has been modified to reduce the contribution made by differences in oxygen sources.

Because most laboratories cannot afford the above mentioned method, they resort to manual calculation of oxygen, which involves subtracting from 100% other ultimate analysis derived elements (carbon, hydrogen, nitrogen, and sulphur), ash and moisture content as described in ASTM D3176-2002 standard test method. It must be noted that the resulting value for oxygen is therefore burdened with all the experimental errors during the determination of the other constituents (Thomas, 2013). For this reason, the oxygen content is still of low order of accuracy. Speight (2005) concluded that the oxygen determination by difference should not be designated percent oxygen but percent oxygen by difference.

2.4.1.2.2 Ultimate analysis as coal quality component

The ultimate analysis is very important in determining of the coal quality component. A combination of results from elements such as carbon, hydrogen and oxygen can be used in ratio form for classification of organic matter in coal seams. One example includes atomic ratios of H/C and O/C (van Krevelen, 1993)

for distinguishing between two major coal types (i.e. humic and sapropelic) based on the coalification path taken by coal samples under investigation.

The determination of elements such as sulphur and nitrogen by the ultimate analysis is also very crucial. Higher sulphur content (e.g. >2%) in the coals have detrimental effects during coal utilisation. During the burning of coal in power stations, sulphur dioxide is produced which causes atmospheric pollution (Thomas, 2013). The other problems caused by sulphur in coal include corrosion and fouling in power stations. Speight (2005) notes sulphur values ranging from 0.8 to 1.0% for steam coals used for electrical generation, <2% for coals used in the production of cement and a maximum of 0.8% for coking coals used on the manufacturing of steel. As for nitrogen in terms of the coal quality component, coals with low nitrogen are preferred in the industry. Thomas (2013) suggests a nitrogen value not higher than 2% because of NO_x emissions.

2.4.1.3 Calorific value analysis

The calorific value analysis is, undoubtedly, important for coal quality determination as it gives the overall specific heat energy in a given coal sample.

2.4.1.3.1 Calorific value analysis test methods

The isothermal calorimeter system using the ASTM D3286-2002 method and the adiabatic calorimeter system following test method ASTM D2015-2007 and ISO 1928-1976 are the two commonly used systems for calorific value determination. Both systems similarly burn a coal sample in an oxygen bomb covered in water surrounded by a jacket (Speight, 2005). The difference occurs in the temperature of the jacket. In the isothermal system, temperature of the jacket is kept constant, whereas for adiabatic system temperature of the jacket is adjusted to match that of the water temperature. In both systems, the total amount of heat produced is measured through temperature rise times the energy equivalent of the calorimeter. The isoperibol system (ASTM D5865-2004) is a newly introduced calorific value analysis method with added advantage over the other two systems in that the calorimeter has a jacket of uniform and constant temperature throughout the analysis (Speight, 2005).

A major concern regarding calorific value determination emanates from error created during temperature measurement. As a mitigation factor, most calorimeters are equipped with a digital thermometer with thermistor probes and microprocessors to control automatically the firing and record temperature at prescribed intervals, in that minimising most human errors occurring during the recording of temperature changes. The equipment maintenance and cleanness at all times is vital in the overall energy equivalent changes of the calorimeter (Speight, 2005). Any malfunction of the stirrers, electrical system or any damage to the oxygen bomb/calorimeter bucket introduces extra heat, which as a result lead to errors in the calorific value measurement (Thomas, 2013).

2.4.1.3.2 Calorific value analysis as coal quality component

By virtue of the fact that the calorific value equates to heating value, it is usually specified as the most important component to be determined on coal to be used for steam plants. It represents the combustible fraction of a coal sample (Speight, 2005). The gross calorific value (GCV) and the net calorific value (NCV) are terms used to differentiate between calorific value types. The difference in the two calorific values is in the latent heat of condensation of the water vapour in that the GCV assumes that the entire vapour produced during the combustion process is fully condensed, whereas the NCV assumes that the water is removed with the combustion products without being fully condensed (Thomas, 2013). To equalize all effects, the calorific value of coal should be compared based on the NCV basis (Speight, 2005). The GCV is mainly used to evaluate the effectiveness of beneficiation processes.

2.5 Coal Rank

Thomas (2013) describes rank as the alteration of vegetation to form peat, succeeded by the transformation of peat through lignite, subbituminous, bituminous, semi-anthracite, anthracite and meta-anthracite. Malvadkar et al. (2004) recognise two phases (i.e. biochemical and geochemical).

2.5.1 Biochemical phase

Malvadkar et al. (2004) describe the biochemical phase to basically include those processes that occur in the peat swamp after deposition and burial. This phase occurs in shallow depth where bacterial activities are common. These bacteria degrade the peat and may be assisted by the rate of burial, pH and level of ground water in the swamp (Thomas, 2013). If the water in the peat contains enough sulphur to support bacterial activities, the resulting process is sulphate ion changed to hydrogen sulphide or elemental sulphur. Instead of escaping the formative peat, Thomas (2013) explains that these two by-products are incorporated into the pre-existing pyrites or into the organic structures in peat.

An increase in density resulting from water squeezed out of peat is attributed to compaction of the previously buried peat receiving more plant materials (Malvadkar et al., 2004). The late stage of peatification involves breaking down of large pieces of plant tissues into smaller pieces due to bacterial attack (Taylor et al., 1998), in the process liberate carbon leading to significant increase in carbon content at this stage. The acceptable last stage of biochemical phase is when the hard brown coal is reached.

2.5.2 Geochemical phase

The geochemical phase involves more sediments piled on top of peat in the process resulting in an increase in both weight and compression in the peat (Malvadkar et al., 2004) which in turn buries the peat deeper. The primary factors that determine the geochemical phase progresses are temperature, pressure, and time (Taylor et al., 1998). A detailed account of coalification and its physical and chemical process is given by Taylor et al. (1998), who describe the major stages of coalification from peat to meta-anthracite. A brief summary of Taylor et al. (1998) description is given in Table 2.3 which outlines not only the rank of coal, but also the dominant processes and physico-chemical changes undergone in each stage in order to produce an increase in rank.

Table 2.3: Major stages of the development from peat to meta-anthracite (reproduced from Taylor et al., 1998; after Levine, 1993).

Coalification Stages	Approximate ASTM Rank Ranges	Predominant Processes	Predominant Physio-Chemical Changes
1. Peatification	Peat	Maceration, humification, gelification, fermentation concentration of resistant substances	Formation of humic substances, increase in aromaticity
2. Dehydration	Lignite to subbituminous	Dehydration, compaction, loss of O-bearing groups, expulsion of -COOH	Decreased moisture content and O/C, CO ₂ and H ₂ O ratio, increased heating value, cleat growth
3. Bituminisation	Upper subbituminous A to high volatile A bituminous	Generation and entrapment of hydrocarbons, depolymerisation of matrix, increased hydrogen bonding	Increased vitrinite Ro, increased fluorescence, increased extract yields, decrease in density and sorbate accessibility, increased strength
4. Debituminisation	Upper high volatile A to low volatile bituminous	Cracking, expulsion of low molecular weight hydrocarbon, especially methane	Decreased fluorescence, decreased molecular weight of extract, decreased H/C ratio, decreased strength, cleat growth
5. Graphitisation	Semi-anthracite to anthracite to meta-anthracite	Coalescence and ordering of pre-graphite aromatic lamellae, loss of hydrogen, loss of nitrogen	Decrease in H/C ratio, stronger XRD peaks, increased sorbate accessibility, anisotropy, strength ring condensation and cleat healing

2.6 Classification of coal

The difference in coal types and properties attributed to variation in age, origin, composition and rank necessitated classification standards which categorise various coals of different regions and setup based on their industrial usage. Falcon and Snyman (1986) established that coal sample taken in different parts of the world, but perceived to be of the same age behave differently due to differences in depositional setup and environmental conditions in different parts of the world. The main focus of classification standard relates to combustion assessment of coal based on the performance and economic value of coals in particular applications.

2.6.1 ASTM Classification

The American Society for Testing and Materials (ASTM) classification (ASTM D388-1999) is usually applicable to many coals across the world (Table 2.4). In this classification standard, two coal properties (i.e. the fixed carbon and calorific values on dry mineral matter free basis) are utilised. The higher rank coals are classified depending on the gross calorific value (on the moist basis) (Thomas, 2013).

Table 2.4: ASTM classification of coal by rank (reproduced from Malvadkar et al., 2004).

		Fixed carbon (%)		Volatile matter (%)		Gross calorific value(MJ/kg)	
		≥	<	≥	<	≥	<
Coal rank							
Anthracite	Meta-anthracite						
	Anthracite	92	98	2	8		
	Semi-anthracite	86	2	8	14		
Bituminous	Low volatile	78	86	14	22		
	Medium volatile	69	78	22	31		
	High volatile A		6	3		32.0	
	High volatile B					30.2	32.0
	High volatile C					26.7	30.2
Sub-bituminous	A					24.4	26.7
	B					22.1	24.4
	C					19.3	22.1
Lignite	A					14.6	19.3
	B						14.6

Although used as supplementary information, the standard test method ASTM D2798-2009 use ranks property and volatile matter correlated with mean maximum reflectance group of macerals. This classification is suitable for coals composed mainly of vitrinite.

2.6.2 South African Classification

In South Africa, the coals are classified using SANS 11760-2007. This classification standard utilises geological maturity (rank), petrographic composition (macerals) and the amount (as well as the nature and mode of association) of the mineral matter present.

2.6.2.1 Rank

By using the rank parameter, SANS 11760-2007 classifies coal into three broad categories; low, medium and high rank (Table 2.5). These primary rank categories are further divided into subcategories, shown in Table 2.5.

Table 2.5: South African Classification using vitrinite reflectance (SANS 11760-2007).

Coal rank		R_r (%)		Description
		$\geq$	$<$	
Broad	Sub-category			
High	A	4.0	6.0	
	B	3.0	4.0	
	C	2.0	3.0	
Medium	A	1.4	2.0	
	B	1.0	1.4	
	C	0.6	1.0	
	D	0.5	0.6	
Low	A	0.4	0.5	
	B		0.4	bed moisture <35 %, ash-free basis
	C		0.4	bed moisture > 35 % and < 75 %, ash-free basis

2.6.2.2 Petrographic composition

The petrographic (or maceral group) composition classifies coals based on the vitrinite content, categorises coals into four classes as shown in Table 2.6.

Table 2.6: South African Classification using vitrinite content (SANS 11760-2007).

Vitrinite content % by volume, mineral-free	Vitrinite class category
< 40	Low vitrinite
>40 and < 60	Medium vitrinite
> 60 and < 80	Moderately high vitrinite
> 80	High vitrinite

2.6.2.3 Ash yield

The inorganic content of coal, as represented by the ash yield, is used to classify coals into five categories as specified in Table 2.7.

Table 2.7: South African Classification using ash yield (SANS 11760-2007).

Ash yield % by mass (dry basis)	Ash class category
< 5	Very low ash
>5 and < 10	Low ash
>10 and < 20	Medium ash
>20 and < 30	Moderately high ash
>30 and < 50	High ash

2.6.3 Russian Classification

GOSSTANDART (GOST) 25,543-88 denotes the Russian coals classification system. This classification standard utilises mainly volatile matter (% d.a.f) and vitrinite reflectance index (Ro %) properties (Table 2.8), to measure the degree of metamorphism in coals. The term mark is used in the Russian system as opposed to rank, although these are not considered entirely synonymous. Moreover, calorific value is used for the classification of coals by types (brown coal, hard coal and anthracite).

Table 2.8: Russian classification of coal by mark (reproduced from Thomas, 2013).

			Volatile matter (%)	Vitrinite reflectance (Ro %)
Coal Type	Letter code	Name of mark		
Brown coal	B	Brown coal	10->48	<0.6
Hard Coals	D	Long flame coal	(+)30	0.40-0.79
	DG	Long flame gas coal	(+)30	0.50-0.79
	G	Gas coal	(+)30	0.50-0.99
	Gzho	Gas fat, semi-lean coal	<38	<0.99
	GZh	Gas fat coal	(+)38	0.50-0.99
	Zh	Fat coal	28-36	0.80-1.19
	KZh	Coke fat coal	24-30	0.90-1.29
	K	Coke coal	24-28	1.00-1.29
				1.30-1.69
	KO	Coke semi-lean coal	24-28	0.80-1.39
	KSN	Coke, weakly caking low metamorphic coal	<30	0.80-1.09
	KS	Coke, weak caking coal	30	1.10-1.69
	OS	Semi-lean caking coal	20	1.30-1.79
	TS	Lean caking coal	<20	1.4-1.99
	SS	Weakly caking coal	>20	0.7-1.79
	T	Lean coal	8-18	1.30-2.59
Anthracite	A	Anthracite	<8	>2.2

2.7 Elements in Coal

The elements in coal are analysed using different techniques based on the elements under investigation. The commonly used techniques for the determination of elements in coal include inductively coupled plasma-atomic emission spectrometry (ICP-AES), inductively coupled-plasma mass spectrometry (ICP-MS) and graphite furnace atomic absorption spectrometry (GFAA). Different methods are in place to determine the mode of occurrence of elements in coal analysis. The elements concentration in coal is very crucial in that, technological

utilisation of coal can be known prior to mining, and also aids in what precaution needs to be in place to guard against the pollution of the environment caused by elements liberated during combustion of coal.

2.7.1 Techniques in geochemical analysis of elements

2.7.1.1 Inductively coupled plasma –atomic emission spectrometry (ICP-AES)

Major and minor elements in coal can be determined by ICP–AES following the standard test method ASTM D6349-2007 in coal ash. This technique is covered comprehensively in Speight (2005) handbook of coal analysis. Based on Speight (2005), this technique can be summarised as involving analysing an ashed coal sample under standard conditions and ignited to constant weight. The ash is fused with a fluxing agent followed by dissolution of the melt in dilute acid solution. This method utilises the measurement of atomic emissions to analyse the elements.

2.7.1.2 Inductively coupled-plasma mass spectrometry (ICP-MS)

The determination of most trace elements concentrations in coal samples are usually carried out by the use of the ICP-MS technique. This technique is described comprehensively in many national and international standard test methods. Dai et al., (2011a) and Li et al., (2014) also provide excellent procedures to follow when using the ICP-MS technique. Prior to analysis of coal samples using ICP-MS technique, most standard test methods advocate for digestion procedures to be carried out to accomplish total decomposition of samples. This is achieved by ashing the coal sample and then subjecting it to microwave dissolution in a mixed acid or alternatively fusion with a mixture of lithium tetraborate and lithium metaborate flux without an ashing process (Lei et al., 2015).

2.7.1.3 Graphite furnace atomic absorption spectrometry (GFAA)

The GFAA technique is used to quantitatively analyse selected elements occurring in concentration below detection limit of ICP-AES and ICP-MS. This technique follows the standard test method (ASTM D6357-2000) for trace elements analysis. This test method determines elements which include; antimony, arsenic, beryllium, cadmium, chromium, cobalt, copper, lead, manganese, molybdenum, nickel, vanadium, and zinc in coal ash (Speight, 2005). In the test method, the coal to be analysed is ashed under controlled conditions, digested by a mixture of aqua regia and hydrofluoric acid, and finally dissolved in 1% nitric acid.

2.7.2 Mode of occurrence of elements

There are two commonly used methods (i.e. indirect and direct) for determining the mode of occurrence of elements in coal (Finkelman, 1994, 1995). Nicholls (1968) was the first researcher to use the indirect method. He used the statistical approach when conducting this method. Generally, elements in coal occur associated either with inorganic constituents (minerals) or with organic constituents (Zhang et al., 2002). According to Nicholls (1968), the concentrations of organic bounded elements in coal decrease or remain almost constant with increasing ash content in coal whereas the concentration of inorganic bounded elements in coal increase with increasing ash content in coal.

2.7.2.1 Major elements

In coal analysis, ash represents the residue of the combusted mineral matter, and it can be broken down and expressed as the series of major oxides (SiO_2 , Al_2O_3 , TiO_2 , CaO , MgO , K_2O , Na_2O , P_2O_5 , Fe_2O_3 and SO_3) (Zhang et al., 2002). In determination of oxides, the presence of large amounts of some oxides such as iron, calcium and sodium and/or potassium in steam coals boilers, which causes slagging and fouling resulting in ashes with low as fusion temperatures (Moosavirad et al., 2012) will be assessed prior to coal unitisation.

2.7.2.2 Trace elements

Prior to utilisation of coal, it is through understanding of the modes of occurrence of trace elements that we can predict trace element behaviour in beneficiation, combustion and environmental impacts associated with a particular trace element. It is through chemical characteristics and rank of coal that individual trace elements associate with the various constituents of the coal (Raask, 1985).

Coal experts, Raask (1985), Swaine (1990), and Clarke and Sloss (1992) have done excellent work as far as trace elements in coal are concerned. The enrichment of trace elements, especially potentially toxic trace elements, are influenced by highly distinctive geochemical environments during stages of coal formation (Moosavirad et al., 2012). Swaine (1990) argues that organic matter and diagenetic minerals can act as enrichment traps for these trace elements. In agreement are other workers (Dai et al., 2005; Finkelman, 1983; Spears et al., 1999) that the relationships between trace element concentration and ash yield can be used as an indicator of organic and inorganic affinity of trace elements in coal. Moosavirad et al. (2012) conclude that the ash content of coal and its geochemical character depends on the environment of deposition and subsequent geological history. Therefore, it is generally considered that most trace elements in coal are associated with the mineral matter (Gentzis and Goodarzi, 1997).

The association of trace elements with either organic or inorganic fractions is very important in the overall technological usage of coal (Moosavirad et al., 2012). Organically bound trace elements are very difficult to separate from the coal matrix (Thomas, 2013), whereas trace elements associated with the inorganic fraction are usually removed with the minerals in which they occur. Elements generally associated with the organic fraction are boron, beryllium and germanium, whereas those associated with the inorganic fraction are arsenic, cadmium, mercury, manganese, molybdenum, lead, zinc, and zirconium (Thomas, 2013). Raask (1985) notes that some trace elements have varying associations as evident by trace elements of gallium, phosphorus, antimony, titanium and vanadium in the organic fraction, whereas cobalt, chromium, nickel, and selenium are in the inorganic fraction.

2.7.3 Trace elements emission during coal combustion

In the modern world with environmental issues escalating every day, it is crucial to avoid at all cost, any increase in pollution to the environment through the emission of hazardous elements from coal combustion. It is through knowledge of the abundance of trace elements that a predictive tool can be used to meet the different requirements of coal users.

Early studies on trace elements representative concentration in coal by Swaine (1990), Finkelman (1993) and Spears and Zheng (1999) on the coal of the world, USA and UK respectively, were essential in estimating the environmental and health effects associated with toxic elements emissions during coal usage. For geochemical comparison, Ketris and Yudovich (2009) provide an excellent estimation of the world averages for trace elements content in coals, specifically detailing average content in hard coals, brown coals and all coals of the world including their ashes. Based on the coal averages from the above findings, many countries aligned their country emission limits to those average values. Although many trace elements concentration researches focused mainly on pollution related issues, the boron concentration in coal provide useful information regarding palaeo-salinity of coal during deposition (Swaine, 1990).

The concentrations of trace elements in coals which are deemed hazardous to the environment are regulated by world organisation authorities. As the leading country on issues of environmental pollutions, United States of America introduced the 1990 US Clean Air Act Amendments (CAAA), which was enacted to guard against elements which were in excess concentration in coal (US Environmental Protection Agency (US EPA), 1995). By the year 1992, the US EPA established the Centre for Air Toxic Metals (CATM) at the University of North Dakota Energy & Environmental Research Centre (EERC) to focus national research efforts on trace element emissions. Following the inception of research centres for toxic elements, a report was released (US EPA, 1998) on hazardous air pollutant emissions from electric steam generating units. The US National Research Council (NRC) classified trace elements of concern in coals based on environmental quality and health (Coal Trading Blog (CTB), 2011). Recently, the

US EPA published a final regulation on emission standards for hazardous air pollutants (HAPs) from coal-fired power stations which were adopted by other countries around the world with minor amendments based on every country's emission limit.

2.7.4 Other analysis

2.7.4.1 Chlorine

Chlorine occurs in coal (Chakrabarti, 1982) and is believed to be a factor not only in fouling problems but also in corrosion. It occurs predominantly as sodium, potassium, and calcium chlorides, but may also combine with organic matter in coal (Speight, 2005). The recommended chlorine levels in steam coals should be in the range of 0.2 -0.3% (air-dried).

2.7.4.2 Phosphorus

Apatite is a carrier of phosphorus in coal, thus a high concentration of apatite is undesirable in coal. Large amounts of phosphorus in coking coals used in the metallurgical industry, has been found to contribute immensely to producing brittle steel (Thomas, 2013). It is also undesirable in stoker firing coal as it causes fouling in the boiler. Coking coals should have a maximum phosphorus content of 0.1% (air-dried).

2.8 Summary

In review of literature regarding sampling procedure in coal analysis, it is fundamentally true that different sampling protocols are used for different types of coals. The sampling preparation employed by different companies and countries differs based on the type of coal and where it is found. Most coal sample preparation standards seem to fundamentally include drying (in air), crushing, and mixing a gross sample to obtain a sample which is ready to be analysed. In coal petrography, the petrographic constituents are directly correlated to important properties in coal to make it possible to predict the technological behaviour of a

coal from its petrographic composition. Macroscopic analysis involves descriptions of coal based on visual appearance. It is established through some literatures that microscopic analyses of coal carried out by use of reflected white light microscopy on the polished surfaces of high quality, under oil immersion determine dually the macerals and vitrinite reflectance in coal sample. The mineral matter in coal, forming the non-combustible inorganic fraction which is introduced in coal in the first or second phases of coalification is the main cause of many problems during coal combustion. By reading through several papers, a definition of coal quality has been understood as those chemical and physical properties of a coal that influences its potential use. Proximate, ultimate and calorific value analyses together quantify coal quality. Coals perceived to be of the same age, taken from different regions of the world have been noticed to behave differently, hence different classification standards are in place across the world. The geochemical analysis of both major and trace elements in coal is crucial in knowing the technological utilisation of coal prior to mining, and also aid in what precaution needs to be in place to guard against the pollution of the environment caused by elements liberated during combustion of coal. Different techniques are in place to determine the trace elements mode of occurrence based on the mineral matter association in coal.

3 METHODOLOGY

This chapter outlines all the sampling methods employed during the collection of data. Section 3.1 mainly gives the background to the sampling rationale which was used in this project. In Section 3.2, all the sample preparation procedures used in this study are described. Section 3.3 specifically deals with standard procedures used in coal petrography analyses. Section 3.4 outlines procedures and techniques followed when analysing proximate parameters. Sections 3.5 and 3.6 details background to the procedure followed in the analyses of ultimate and calorific value analysis respectively. Section 3.7 concludes this chapter by focusing on the geochemical equipment used to analyse both major and trace elements in coal.

3.1 Sampling

A total of 8 coal samples were collected from a drill core (JVP20) at Western Moiyabana Coalfield during the drilling campaign carried out by Jaquar Ventures (Pty) Ltd in October, 2014 (Fig. 3.1). A TNW core size (60.2 mm diameter) was used for this study. Sampling intervals were guided by lithological and geophysical logging interpretation. Only coal seams from the Morupule Formation were sampled due to their known current economic exploitation at Morupule Coalfield. Other seams found in this Coalfield are either too thin or have partings qualifying them to be of no economic importance. The collected coal samples were numbered and placed in plastic storage bags to prevent contamination and to minimize oxidation. All the 8 coal samples were subjected to petrographic (macerals constituents including mineral matter and vitrinite reflectance measurement), chemical (proximate, ultimate and calorific value) and geochemical analysis (ICP-AES and ICP-MS).

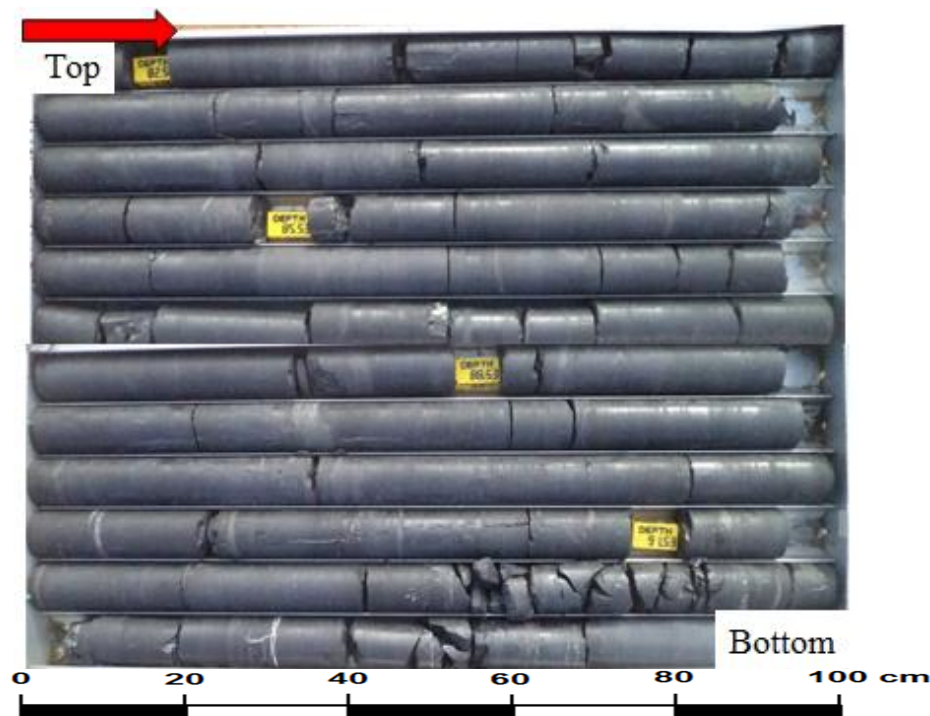


Figure 3.1: The JVP20 borehole core – coal horizon only.

3.2 Sample Preparation

Initial crushing of the core was carried out at the University of Johannesburg (UJ), Doornfontein Campus (DFC). A jaw crusher was used to reduce core particles to 25 mm, followed by roll crusher which reduced the core particles to 12 mm and lastly a cone crusher was used to further crush coal particles to 5 mm. Prior to further crushing of coal particles to -2 mm for petrographic analysis and -0.250 mm for proximate, calorific value (CV) and sulphur/carbon analysis, a cone and quartering method was used to split the samples until a representative mass of coal particles (about 200 g) of the sample was achieved. After the splitting of samples, further crushing of remaining coal particles was carried out using an Ultra Centrifugal Mill (Retsch ZM200) at the University of Witwatersrand in the School of Chemical Sciences and Metallurgical Engineering.

For petrography, further preparations were performed to make the coal blocks/pellets. The -2 mm split was used for coal petrography. A mixture of epoxy resin and hardener in the ratio of 7:1 was used for embedding the coal samples for pellet/block preparation. The mixture was left for 24hrs before polishing. One sample mount was made from each sample thus totalling to 8 coal blocks/pellets

for this project. After removing the blocks/pellets from cups and samples number engraved on the bottom and side, grinding and polishing commenced. The grinding stage uses 350, 500 and 1000 SIC paper, using a Struers Tegraforce polisher. Further three polishing stages include usage of Allegro Largo, Diapro DAL and OP-S suspensions to achieve a final polished surface. OP-S is a 0.05 micron suspension. All samples were cleaned and dried before analysis. SABS ISO 7404-2 was followed in preparation, carried out at UJ.

3.3 Coal Petrography

After carrying out the above preparation procedure, the pellets/blocks were analysed using a reflected polarised light microscope (Zeiss Axio Imager M2M) fitted with two digital cameras mounted at the top of the microscope. Prior to reflectance analysis, the system was calibrated using a Yttrium Aluminium Garnet (YAG) with reflectance of 0.9 under immersion oil. Immersion oil is used in coal microscopy to increase the resolution of the image produced. This is achieved by immersing both the objective lens and the specimen in a transparent oil of high refractive index, thereby increasing the numerical aperture of the objective lens. Typical oils used have an index of refraction around 1.515. A total magnification of x500 was used.

Fossil Software (Diskus) was used together with the microscope to analyse samples for both vitrinite reflectance (mainly on collotelinite) and maceral analysis. This software also allows the user to take photos in different places on the sample surface. Reflectance measurements were determined in the random mode (Rom) and the values reported were arithmetic means of at least 100 measurements per sample. The standard requires a minimum of 100 points, but in some instances the samples being assessed had very low vitrinite contents, making it impossible to analyse 100 different points. Vitrinite reflectance analysis was carried out to determine coal rank using standard test method SABS ISO 7404-5.

Macerals analyses were conducted to determine the macerals following standard test method SABS ISO 7404-3 for determination of macerals in coal. At least 500

points on each sample was counted and 27 categories were recorded; 4 mineral groups and 23 macerals. The results were expressed on a mineral included and excluded basis. Photomicrographs at a total magnification of x500 were taken during the analysis of both maceral and vitrinite reflectance analysis. These analyses were conducted by Prof. Wagner at Department of Geology, UJ.

3.4 Proximate Analysis

Proximate analysis was carried out using a Simultaneous Thermal Analyser-PerkinElmer STA 6000 with PYRIS SOFTWARE to process data during analysis, housed at the University of Witwatersrand. A mass of between 11.00 to 11.19 mg was used for proximate analysis. The sample was weighed in the crucible after zeroing the weight pan in the Pyris program. The sample mass was locked to stabilise the weight after which the analysis was started. Parameters which were measured include: moisture, volatile matter, fixed carbon and ash. On completion of analysis, calculation of moisture, volatile matter and fixed carbon contents are done on the graphs based on the following temperatures:

1. 0°C to 110°C - moisture content (MC) (%),
2. 110°C to 700°C - volatile matter (VM) (%) and
3. 700°C to 900°C - fixed carbon (FC) (%).

Value of ash was calculated by adding up (MC+VM+FC) and subtracting them from 100 i.e. ash content = $100 - (MC + VM + FC)$.

3.5 Total Sulphur Analysis

A Leco SC 632 instrument was used to analyse total sulphur at the University of Witwatersrand. After switching on the instrument, it was left to warm up to 1350°C, stepping up 300°C at a time. Analyse standard test (503-388-5.4%S) was used to check instrument precision and calibration status. A mass of 0.250 g per sample was required for this analysis. After every third sample, the standard test sample was analysed again to check the reliability of data. Two portions were analysed from each sample batch to check the repeatability of data analysis and subsequently the average value of the results used as the final value for each sample.

3.6 Ultimate Analysis (C, H, O and N analysis)

The elemental analysis of carbon, oxygen, hydrogen and nitrogen in the Moiyabana coal samples was determined following the procedure outlined in the ASTM D5373-2002 standard test method. The analysis was carried out at Australian Laboratory Services (ALS) (Pty) Ltd, Witbank, South Africa.

3.7 Calorific Value Analysis

A Bomb Calorimeter housed at the University of Witwatersrand was used in this research for calorific value (CV) analysis. A Benzoic Acid pill of 0.504 g was used to calibrate the machine prior to sample analysis and the machine set to 2500 kPa. The equipment was allowed to stabilise to the jacket temperature of approximately 36.3°C. A mass of 0.40 g was required for every sample analysis, which was weighed on the precision measuring balance (Mettler Toledo-Classic MS) using a crucible. Rubber on the cap of the bomb was insulated with grease to stop gas leakage. Cotton was placed on the firing wire and lowered to the sample in the crucible and then the cap lowered into the bomb. The bomb was filled with oxygen and lowered gently into place; the analysis will start automatically.

3.8 Geochemical Analysis

3.8.1 Major elements analysis

The major elements analysis was outsourced to ALS laboratory, South Africa. For the determination of major elements (ash analysis), the standard test method (ASTM D6349-2007) was employed with the use of ICP–AES technique.

The ICP-AES technique based on description by Speight (2005) involves ashing of samples under standard conditions and ignites to constant weight. The ash is then allowed to fuse with a fluxing agent upon which is then followed by dissolution of the melt in dilute acid solution. The solution was then analysed by ICP–AES for the major elements oxides (SiO_2 , CaO , MgO , Fe_3O_2 , Al_2O_3 , Na_2O ,

K₂O, TiO₂, SO₃, P₂O₅ and MnO). Speight (2005) warns that this technique is solely based on the measurement of atomic emissions.

3.8.2 Trace elements

The trace elements determination in coal was carried out employing the ICP-MS technique. This analysis was outsourced to UIS Analytical Services, Centurion, South Africa and the trace elements determined were As, B, Ba, Bi, Cd, Co, Cr, Ge, Hg, Li, Mn, Mo, Ni, Pb, Sb, Se, Sr, Th, U, V, Zn, Cl and F.

4 RESULTS

This chapter presents all the data generated during the experimental procedures carried out throughout the entire project, which are then discussed in Chapter 5. The results obtained macroscopically (lithotypes) aid in the field-based assessment of the coal samples. The results of the microscopic study enable a better understanding of the organic and mineral constituents in coal. Proximate analysis results are very important in buying/selling, choosing the best equipment for a specific coal type, determining the efficiency of coal burning equipment and predicting the quality of ash disposal. Ultimate analysis forms part of another very important component of coal results. It determines the main chemical elements in coal, which have direct bearing on the usability of the coal. Calorific value analyses forms part of the chemical component in coal and its determination gives an overview of the heating value of coal. Geochemical analysis considers the minerals, major elements and trace elements in coal. The determination of minerals and trace elements in coal can provide useful information relating to the environment for possible contamination of toxic trace elements in air, soil and groundwater during coal combustion and utilisation.

4.1 Coal Petrography

4.1.1 Lithotype Results

The results of the lithotype assessment of Moiyabana coals following the Australian standard of coal logging is shown in Table 4.1 and macroscopically represented in Figure 4.1. The Moiyabana coal samples are banded in nature and durain is observed to dominate the lithotypes imparting a dull appearance to these coals. Observed is also vitrain bands up to one centimetre thick, partly developed clarain bands and thin fusain lenticels. The recognised lithotypes thus include (Table 4.1):

- a) Dull coal: This is the dominant megascopic band type in this borehole of the Moiyabana coalfield. They are dull in appearance, hard and compact in

nature, and are known to have a higher specific gravity than other lithotypes.

- b) Banded dull coal: This megascopic band type occurs sporadically across the coal seams.
- c) Banded bright coal: In general, banded bright coal is uncommon in the Moiyabana coal samples except in some parts, where it occurs as stringers.

Table 4.1: Lithological succession encountered in borehole JVP20, Moralané area, Moiyabana Coalfield.

Sr. No	Depth(m)		Thickness (m)	Lithology	Sample ID	Seam Name (Smith, 1984)
	From	To				
1	0.00	12.18	12.18	Calcrete		
2	12.18	13.81	1.63	Conglomerate		
3	13.81	22.91	9.10	Sandstone		
4	22.91	23.22	0.31	Mudstone		
5	23.22	23.54	0.32	Weathered Coal		
6	23.54	23.92	0.38	Mudstone		
7	23.92	28.16	4.24	Sandstone		
8	28.16	35.26	7.10	Siltstone		
9	35.26	36.65	1.39	Sandstone		
10	36.65	42.18	5.53	Siltstone		
11	42.18	44.31	2.13	Coal/Carb mudstone		
12	44.31	44.66	0.35	Siltstone		
13	44.66	45.60	0.94	Mudstone		
14	45.60	45.84	0.24	Coal mixed		
15	45.84	48.10	2.26	Mudstone		
16	48.10	49.06	0.96	Coal/Carb Mudstone		
17	49.06	52.84	3.78	Mudstone		
18	52.84	54.12	1.28	Siltstone		
19	54.12	62.46	8.34	Mudstone		
20	62.46	72.16	9.70	Coal/Carb Mudstone		
21	72.16	73.39	1.23	Coal Dull		
22	73.39	74.28	0.89	Coal/Carb Mudstone		
23	74.28	75.29	1.01	Coal Dull Heavy		
24	75.29	75.59	0.30	Carb Mudstone		
25	75.59	77.32	1.73	Coal Dull		
26	77.32	77.63	0.31	Mudstone		
27	77.63	78.82	1.19	Coal Dull		
28	78.82	80.42	1.60	Siltstone		
29	80.42	82.18	1.76	Coal/ mudstone		
30	82.18	84.04	1.86	Mudstone		
31	84.04	86.52	2.48	Coal/ Minor Carb mudstone	H	F2b
32	86.52	87.54	1.02	Coal/ Minor Mudstone	G	F2b
33	87.54	87.68	0.14	Coal/Carb. Mudstone		F2a
34	87.68	93.00	5.32	Mudstone		
35	93.00	94.68	1.68	Siltstone		
36	94.68	96.03	1.35	Coal/Minor Mudstone	F	F1d
37	96.03	97.70	1.67	Coal	E	F1d
38	97.70	100.13	2.43	Coal/ Minor Carb Mudstone	D	F1c
39	100.13	101.57	1.44	Coal/Carb Mudstone	C	F1b
40	101.57	103.78	2.21	Coal Dull	B	F1a
41	103.78	104.66	0.88	Coal/Dull lustrous	A	F1a
42	104.66	105.18	0.52	Coal/Carb Mudstone		
43	105.18	106.35	1.17	Mudstone/Siltstone		
44	106.35	108.43	2.08	Coal/Carb mudstone/ Mudstone/ sandstone		
45	108.43	109.68	1.25	Siltstone		
46	109.68	112.69	3.01	Conglomerate		
47	112.69	115.62	2.93	Sandstone		

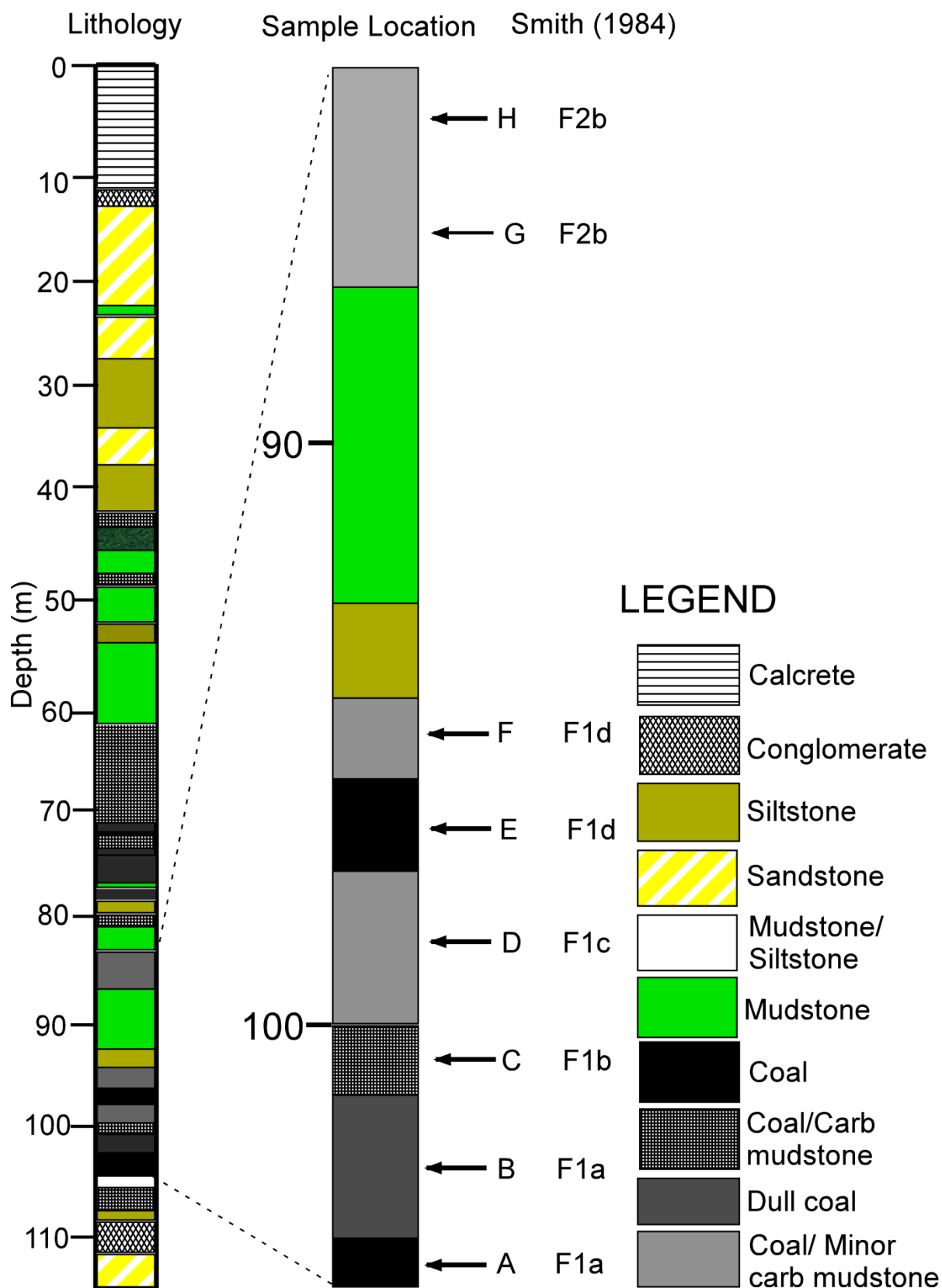


Figure 4.1: Megascopic representation of JVP20 correlated with Smith (1984) formation and sample locations in the Morupule Formation at Moiyabana Coalfield, Botswana.

4.1.2 Macerals analysis

The maceral constituent's proportions for coal samples from borehole JVP20 are characterised by the high quantity of inertinite and low proportions of vitrinite and liptinite macerals (Fig. 4.2). High mineral matter content constitutes another characteristic of Moiyabana coal samples. Table 4.2 shows the results of maceral compositions divided into group and type with the inclusion of observable mineral matter.

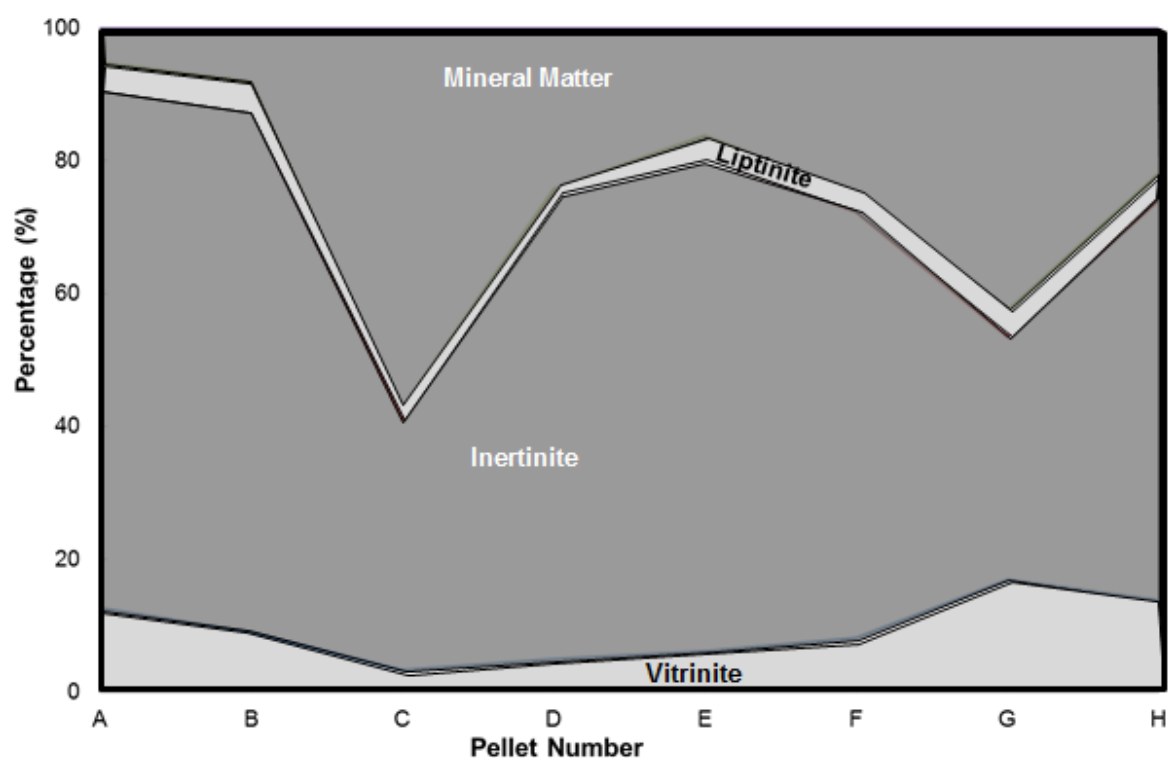


Figure 4.2: Variation of macerals and mineral matter within coal seams intersected at borehole JVP20, Moiyabana Coalfield.

Table 4.2: Maceral compositions (in vol%) of JVP20 coal samples, Moiyabana Coalfield, eastern Botswana.

	Sample	A		B		C		D		E		F		G		H	
GROUP	MACERAL (vol%)	Inn. mm	mmf	Inn. mm	mmf	Inn. mm	mmf	Inn. mm	mmf	Inn. mm	mmf	Inn. mm	mmf	Inn. mm	mmf	Inn. mm	mmf
Vitrinite	telinite	0.2	0.2	0.0	0.0	0.2	0.5	0.0	0.0	0.4	0.5	0.0	0.0	0.0	0.0	0.0	0.0
	collotelinite	9.4	9.9	6.6	7.2	2.4	5.7	2.8	3.7	1.8	2.1	4.8	6.4	14.0	24.2	10.0	12.9
	vitrodetrinite	0.2	0.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	collodetrinite	2.6	2.8	2.4	2.6	0.4	1.0	1.6	2.1	4.0	4.8	2.2	2.9	3.0	5.2	3.4	4.4
	corpogelinite	0.2	0.2	0.0	0.0	0.0	0.0	0.6	0.8	0.0	0.0	0.2	0.3	0.0	0.0	0.4	0.5
	gelinite	0.0	0.0	0.0	0.0	0.2	0.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	pseudovitrinite	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.0	1.3	0.0	0.0	0.0	0.0
Inertinite	fusinite	2.4	2.5	6.2	6.8	0.7	1.6	1.8	2.4	0.6	0.7	0.4	0.5	1.7	2.8	2.2	2.8
	semifusinite	24.4	25.8	23.4	25.5	10.0	23.3	15.5	20.4	19.0	22.6	15.9	21.2	15.6	27.2	16.2	20.8
	micrinite	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	macrinite	0.2	0.2	0.6	0.7	0.0	0.0	0.0	0.0	0.6	0.7	0.0	0.0	0.0	0.0	0.0	0.0
	secretinite	1.0	1.1	0.0	0.0	0.2	0.5	1.0	1.3	0.2	0.2	0.0	0.0	0.0	0.0	0.2	0.3
	funginite	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	inertodetrinite	50.3	53.1	48.2	52.5	26.8	62.7	52.2	68.9	54.1	64.5	47.9	63.9	19.3	33.6	41.5	53.2
Liptinite	sporinite	3.0	3.1	4.0	4.4	1.1	2.6	0.2	0.3	2.8	3.3	1.2	2.1	3.3	5.8	3.2	4.1
	cutinite	0.8	0.8	0.4	0.4	0.0	0.0	0.2	0.3	0.4	0.5	0.4	0.5	0.0	0.0	0.4	0.5
	resinite	0.0	0.0	0.0	0.0	0.4	1.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	alginite	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	liptodetrinite	0.0	0.0	0.0	0.0	0.2	0.5	0.0	0.0	0.0	0.0	0.6	0.8	0.7	1.2	0.4	0.5
	suberinite	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	exsudatinite	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Mineral matter	silicate (clay/quartz)	4.0		6.6		54.6		18.3		5.2		15.7		37.2		14.4	
	sulphide	0.4		0.2		1.8		2.8		4.4		5.0		0.0		0.8	
	carbonate	0.8		1.4		0.9		3.2		6.6		4.4		5.3		6.8	

(Inn.mm- including mineral matter; mmf-mineral matter free)

4.1.2.1 Vitrinite Maceral Group

Figure 4.3 present photomicrographs of vitrinite group macerals observed during petrographic analyses of the Moiyabana coal samples. The observed macerals of this group are collotelinite, collodetrinite, telinite, vitrodetrinite, corpogelinite, gelinite and pseudovitrinite. Collotelinite is the most abundant maceral of the vitrinite group (1.80 to 10.02 vol%), with an average value of 6.47 vol%. The structureless and homogeneous mass in thin and thick bands of collotelinite in most samples is evident (Fig. 4.3b). The isolated irregular fragments of different dimensions are features characterising collotelinite of the Moiyabana coal samples (Fig. 4.3c). Secondary infilling of mineral matter also occur in cracks and cavities of collotelinite (Fig. 4.3a and e). Furthermore, collotelinite shows banding in some places, differentiated by colour (light grey to medium grey) and reflectance (Fig. 4.3c). This banding is reported by Falcon and Snyman (1986) to qualitatively represent a difference in hydrogen content, the darker band more impregnated with hydrogen or resinous compounds than the lighter.

Collodetrinite content varies from 0.44 to 3.99 vol% with an average value of 2.45 Vol%. The collodetrinite in the study area coal samples is present as mottled vitrinitic groundmass binding other coal components (Fig. 4.3d) and often show lower reflectance than associated collotelinite, a feature which have been reported by Solaymani and Taghipour (2012). The dominant macerals embedded in the groundmass of collodetrinite maceral are sporinite and cutinite (Fig. 4.3d), a characteristic observed across all the Moiyabana samples. In some samples, a faint banding or stratification is evident, as seen in Figure 4.3d.

Occurring in very small amount, corpogelinite is observed in four coal samples. It ranges from 0.20 to 0.60 vol% with an average value of 0.30 vol%. This maceral occurs mostly in association with inertodetrinite maceral (Fig. 4.3f). The other macerals (telinite, vitrodetrinite, gelinite and pseudovitrinite) recorded low values with average values less than 0.20 vol% (Table 4.2).

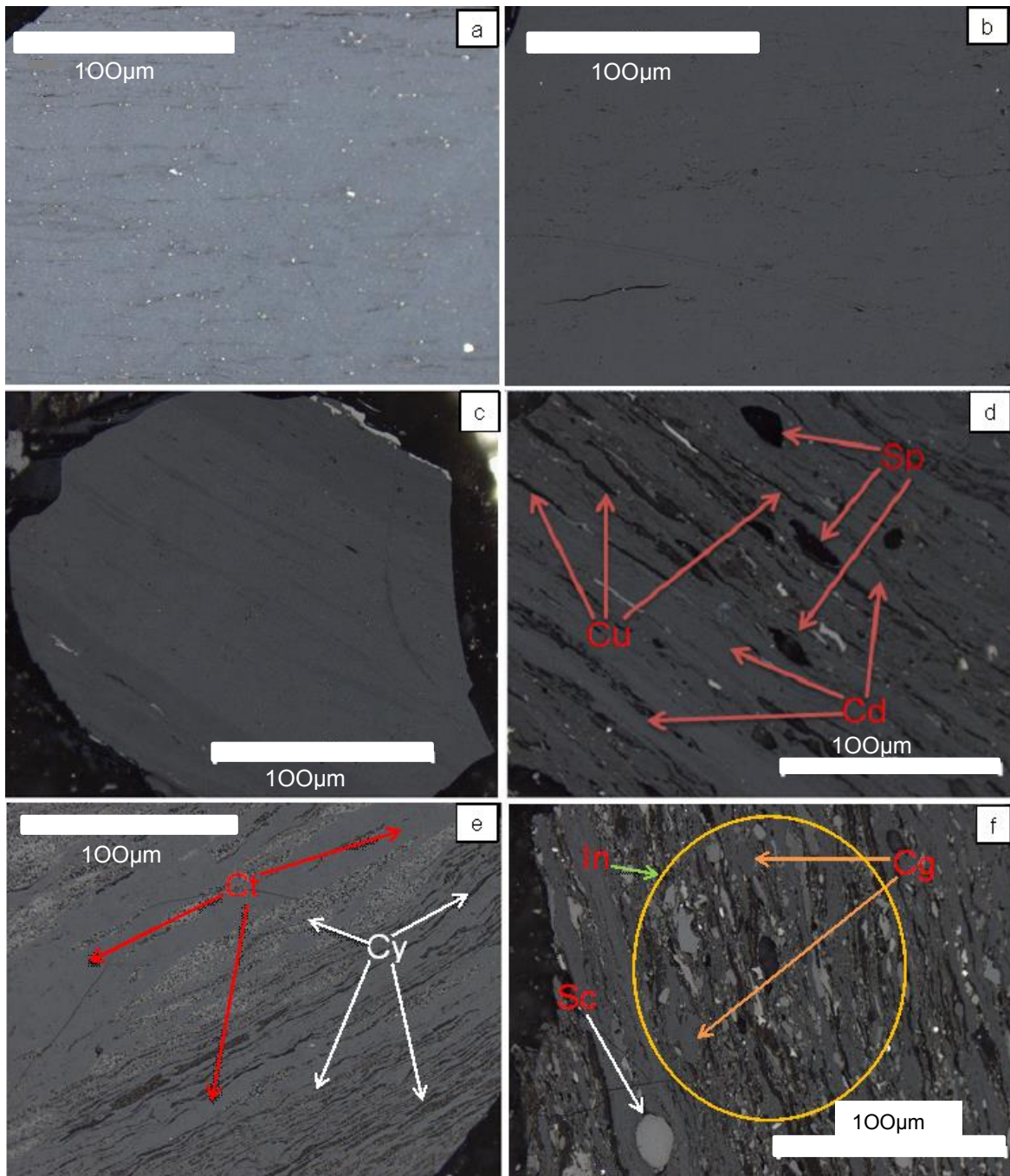


Figure 4.3: Photomicrographs of vitrinite group maceral from Moiyabana coal samples, under reflected polarised light in transparent oil immersion examination. **a)** Collotelinite with disseminated pyrite (white spots) and clay (grey/brown). **b)** Homogeneous band of collotelinite. **c)** Collotelinite showing banding as differentiated by colour and reflectance. **d)** Collodetrinite (Cd) forming groundmass in which sporinite and cutinite (liptinite) and inertodetrinite (inertinite) are embedded. **e)** Collotelinite (Ct) with clay (Cy). **f)** Corpogelinite (Cg) occurring in association with inertodetrinite (In) and secretinite (Sc).

4.1.2.2 Inertinite Maceral Group

The macerals identified in the inertinite group are fusinite, semifusinite, macrinite, micrinite, secretinite and inertodetrinite. Inertodetrinite (26.83 to 54.09 vol%) is the dominant maceral in this group (Table 4.2). It is observed as discrete small fragments of varying sizes, shape, structure and forms with high reflectance (Fig. 4.4f). However, it appears to have been derived mostly from fusinite and semifusinite macerals (mainly contain fragments of both).

Semifusinite maceral occurs in appreciable concentrations (9.98 to 24.45 vol%) with a reflectance between collotelinite and fusinite. It occurs as lenticels (in other samples bands) with poorly preserved cell structures and colour varying between light grey and white (Fig. 4.4a, c and e). Another inertinite maceral, fusinite, recorded values between 0.40 to 6.20 vol% and shows a well preserved cellular structure (Fig. 4.4b and d). The cell lumens are found to be filled with mineral matter in most of the fragments/bands, mostly argillaceous in composition (Fig. 4.4b).

Other macerals in this group include macrinite and secretinite. They both recorded very low values (0.00 to 0.60 and 0.00 to 1.00 vol%) respectively. Although rarely occurring in the Moiyabana coal samples, large fragments of secretinite have been observed as shown in Figure 4.4c. It occurs as large size oval to rounded bodies with un-oriented vesicles and very high relief (Fig. 4.3f and 4.4c). The funginite and micrinite macerals were not detected.

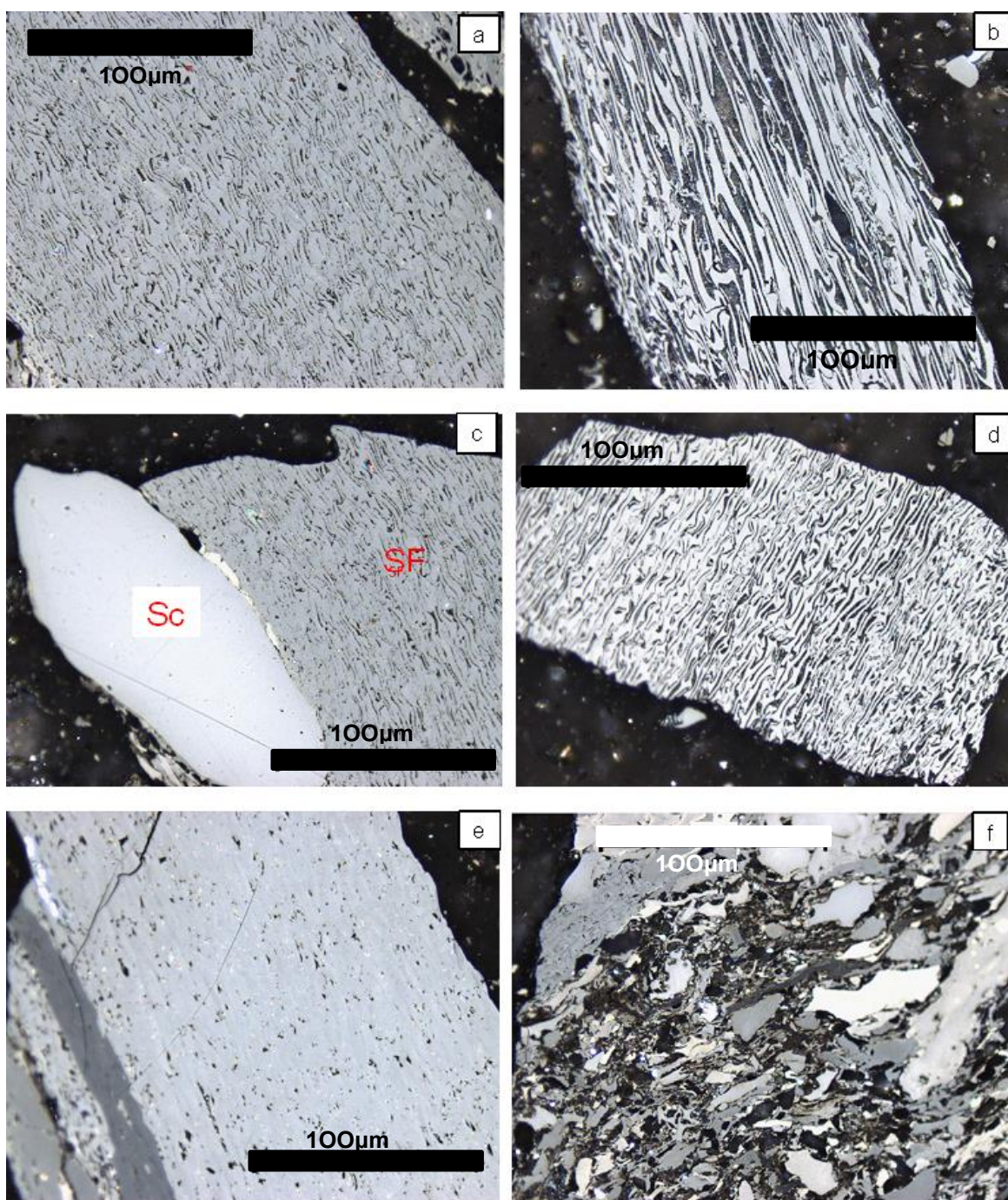


Figure 4.4: Photomicrographs of inertinite macerals from Moiyabana coal samples, under reflected polarised light in transparent oil immersion examination. **a)** Semi-fusinite. **b)** Fusinite. **c)** Secretinite(Sc) associated with semifusinite(SF). **d)** Fusinite. **e)** Semifusinite. **f)** Inertodetrinite.

4.1.2.3 Liptinite Maceral Group

The liptinite maceral group recorded the lowest amount in the maceral groups analysed. The identified maceral types in this group are sporinite, cutinite,

liptodetrinite, and resinite. Sporinite is the dominant maceral in the this maceral group (0.20 to 4.00 vol%). It was observed as elongated thread-like structures with serrated margins (Fig. 4.5b).

Although occurring in low amounts (0.00 to 0.80 vol%), cutinite is detected in most samples, except C and D. It occurs as elongated thread-like bodies or spindle-shaped dark grey coloured bodies (Fig. 4.5b). Resinite was recorded only in sample C. It is mainly observed to occur in the original cell cavities in vitrinite as oval-shaped structures (Fig. 4.5a). Liptodetrinite occurs in 4 samples and usually embracing the fragmental parts and detrital remains of structured liptinite macerals produced due to degradation and crushing (Fig. 4.5c and d). Alginite, suberinite and exsudatinite were not detected.

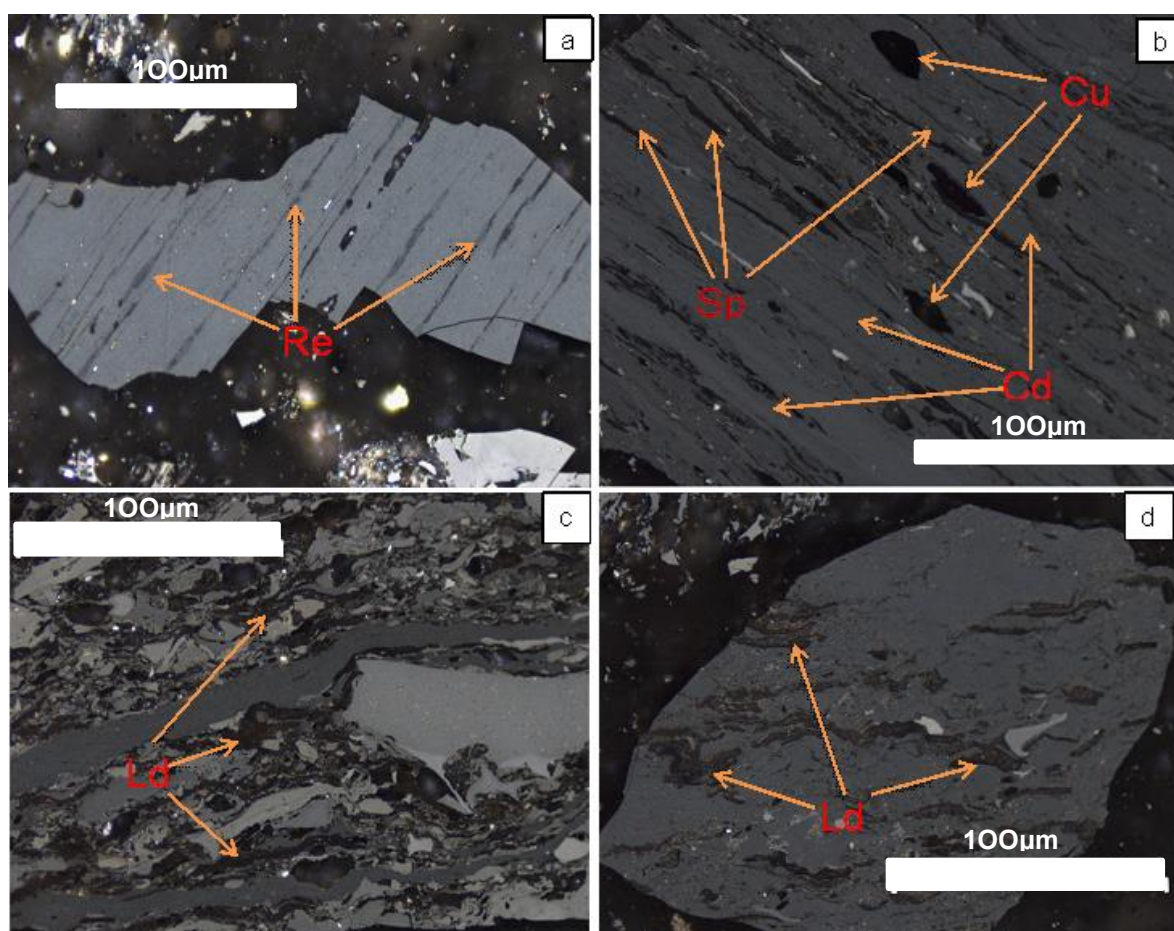


Figure 4.5: Photomicrographs of liptinite macerals from the Moiyabana coal samples, under reflected polarised light in transparent oil immersion examination. **a)** Resinite in vitrinite maceral. **b)** Sporinite (Sp) and cutinite (Cu) macerals types in colloditrinite (Cd) groundmass. **c)** and **d)** Liptodetrinite (Ld).

4.1.3 Vitrinite Reflectance Measurements

Table 4.3 presents results of the random vitrinite reflectance values with mean and standard deviation values generated in the petrographic analysis of the Moiyabana coal samples. The vitrinite random reflectance values range from 0.65 to 1.75%, while mean random reflectance values of these coal samples range from 0.84 to 1.04% with an average value of 0.95%. Standard deviation values range from 0.08 to 0.23. In Figure 4.6, results of random vitrinite reflectance (R_o %) are summarised in histogram format using data tabulated in Table 4.3.

The distribution of random vitrinite reflectance values in the form of a histogram illustrates the proportion of particles falling in the different reflectance intervals (half V-steps) (Fig. 4.6). Vitrinite reflectance analysis places the coals in the medium rank bituminous C borderline B category (ECE-UN, 1998) which is slightly higher than the other petrographic assessed coals from this area. Typical coals from Morupule Formation are reported to be sub-bituminous in rank (Hancox, 2016). It can be deduced from the plots that the samples A, B, C, G and to lesser degree F have their histograms clustered around the mean values, which signifies overall the same proportion of coal constituents (composition). The histograms of samples D, E and H have mixtures of different coals as shown by scattering of individual reflectance values within the same coal sample (Fig. 4.6).

Table 4.3: Random vitrinite reflectance (RoV) measurements with mean and standard deviation values from Moiyabana Coalfield, Botswana.

Specimen		Sample A	Sample B	Sample C	Sample D	Sample E	Sample F	Sample G	Sample H
Date		20/06/2016	15/06/2016	20/06/2016	21/06/2016	15/06/2016	15/06/2016	22/06/2016	15/06/2016
Count		124	110	72	85	131	114	120	130
Mean		0.84	0.93	0.95	1.04	1.04	1.00	0.84	0.94
Standard Deviation		0.08	0.10	0.09	0.18	0.23	0.17	0.10	0.11
Vitrinite Reflectance (%)									
From	To	Frequency (%)	Frequency (%)	Frequency (%)	Frequency (%)	Frequency (%)	Frequency (%)	Frequency (%)	Frequency (%)
0.65	0.70	2						6	
0.70	0.75	9	4	4	2	4	3	12	2
0.75	0.80	21	7	3	2	3	8	16	8
0.80	0.85	28	12	4	6	11	9	22	10
0.85	0.90	14	20	11	13	11	12	22	17
0.90	0.95	12	18	22	7	15	12	10	22
0.95	1.00	11	20	27	18	10	10	7	18
1.00	1.05	2	7	18	15	11	8	2	14
1.05	1.10	1	5	7	9	8	13	3	2
1.10	1.15		5	3	7	4	8		4
1.15	1.20		1	1	4	2	9		1
1.20	1.25		1		2	3			
1.25	1.30				4	2	3		
1.30	1.35					2	1		
1.35	1.40				5	2	1		1
1.40	1.45				4	2			
1.45	1.50				2	1			
1.50	1.55					5			1
1.55	1.60					1	2		
1.60	1.65					2			
1.65	1.70						1		
1.70	1.75					1			
Total Frequency (%)		100	100	100	100	100	100	100	100

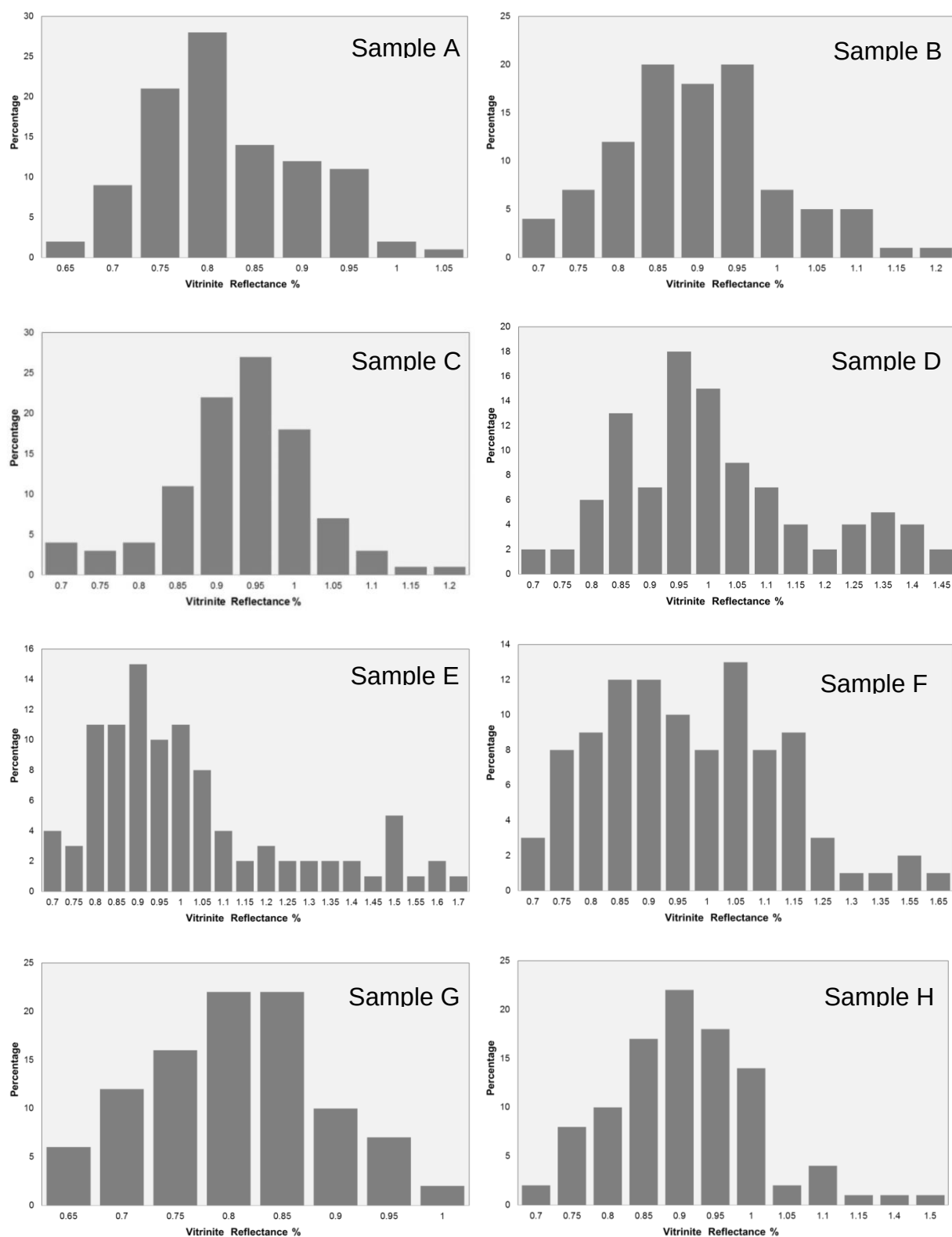


Figure 4.6: Histograms showing distribution of random vitrinite reflectance (Rom %) measured in half V-steps using values from Table 4.3.

4.1.4 Mineral Matter

The observable mineral matter content of the Moiyabana coal samples ranges from 5.21 to 57.21 vol% with an average value of 25.08 vol% (Table 4.4). The observable minerals include silicates (clay and quartz minerals), carbonates and sulphides (Fig. 4.7). The dominant fraction of the mineral matter is silicate minerals, occurring in concentrations ranging from 4.01 to 54.55 vol% with an average value of 19.49 Vol% (Table 4.2).

Table 4.4: Summarised maceral groups and mineral matter data of the Moiyabana coal samples, eastern Botswana.

	A	B	C	D	E	F	G	H
Vitrinite	12.6	9.0	3.3	5.0	6.2	8.2	17.0	13.8
Inertinite	78.4	78.4	37.7	70.4	74.5	64.2	36.5	60.1
Liptinite	3.8	4.4	1.8	0.4	3.2	2.6	4.0	4.0
Mineral Matter	5.2	8.2	57.2	24.2	16.2	25.1	42.5	22.0

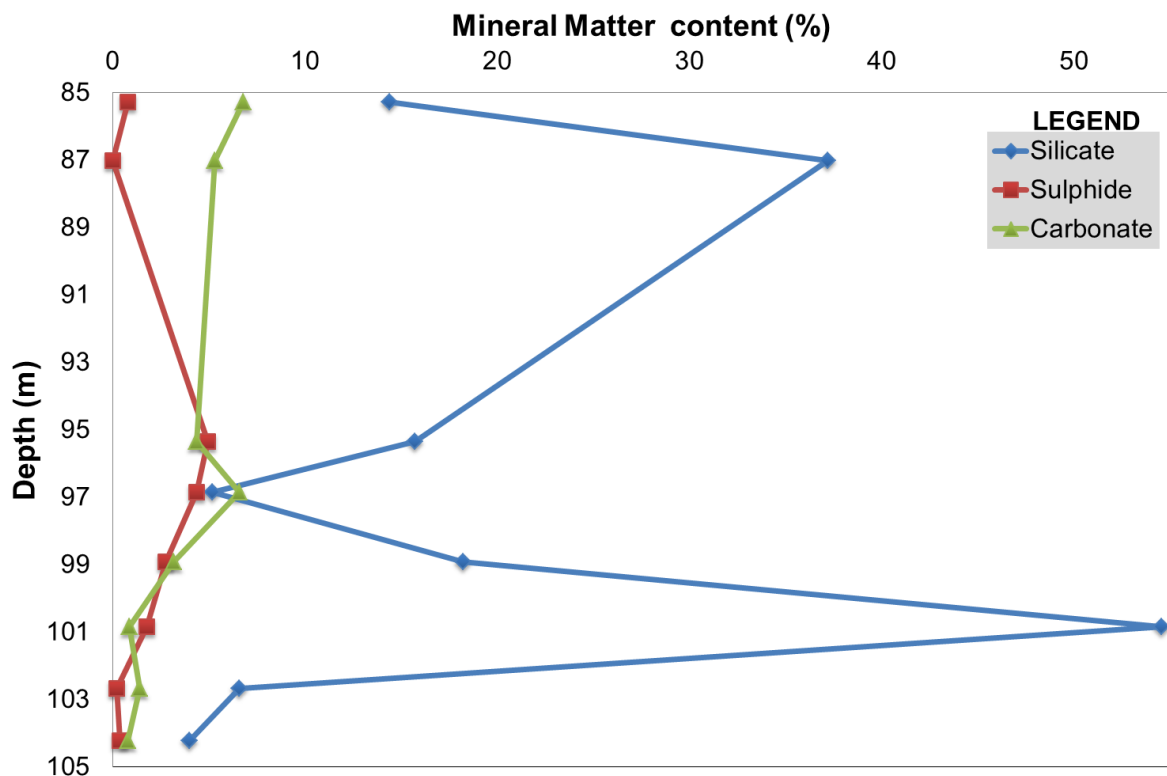


Figure 4.7: Petrographically observable mineral matter concentrations showing vertical variation.

Figure 4.8 shows some photomicrographs of clay occurrences in the Moiyabana coals taken under reflected polarised light with transparent oil immersion examination. It occurs as groundmass (Fig. 4.8a), thin bands (Fig. 4.8b), disseminated particles (Fig. 4.8c), cell filling (Fig. 4.8d) and groundmass (Fig. 4.9). Quartz is mainly observed to occur as individual grains (Fig. 4.9) and rarely observed to occur as cleat- and/or fracture-fillings in these coals.

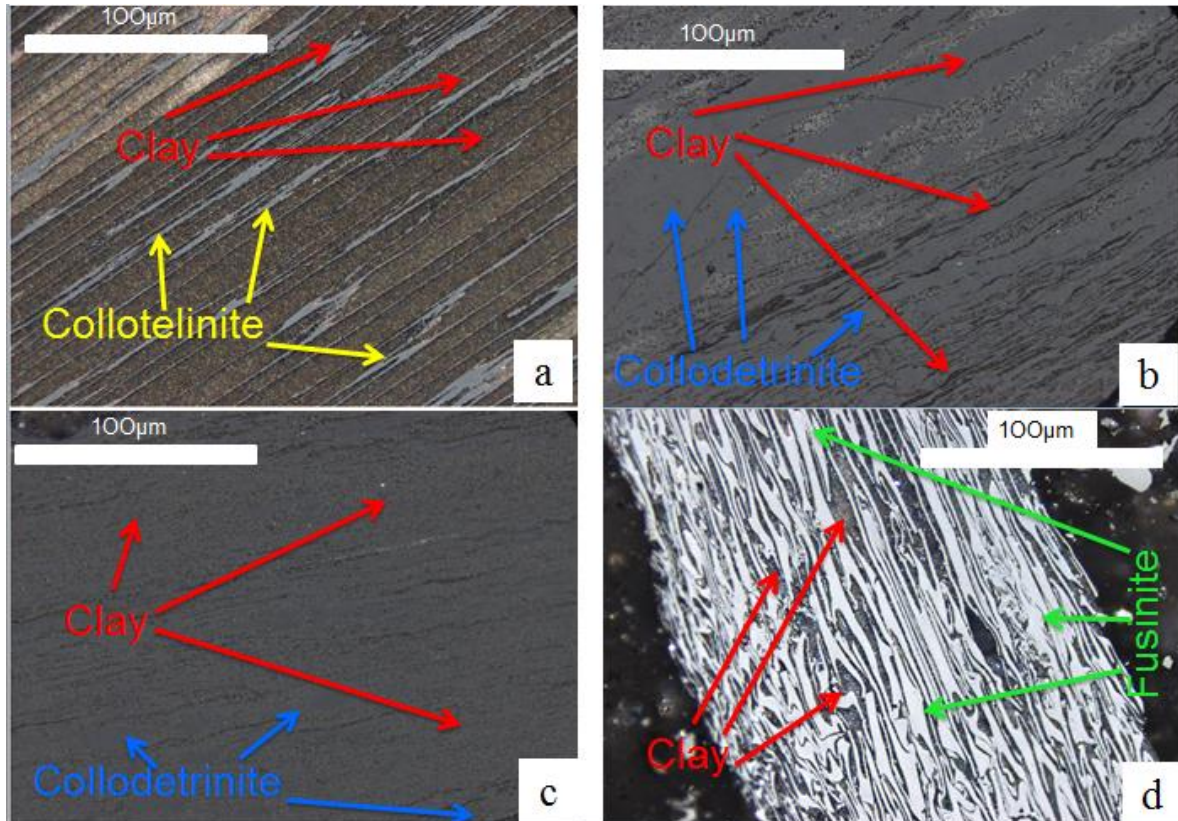


Figure 4.8: Photomicrographs of clay occurrences in the Moiyabana coal samples; under reflected polarised light in transparent oil immersion examination. **a)** Clay groundmass with stringers of collotelinite. **b)** Thin bands of clay in collodetrinite. **c)** Syngenetic clay-disseminated. **d)** Clay as cell-filling in fusinite.

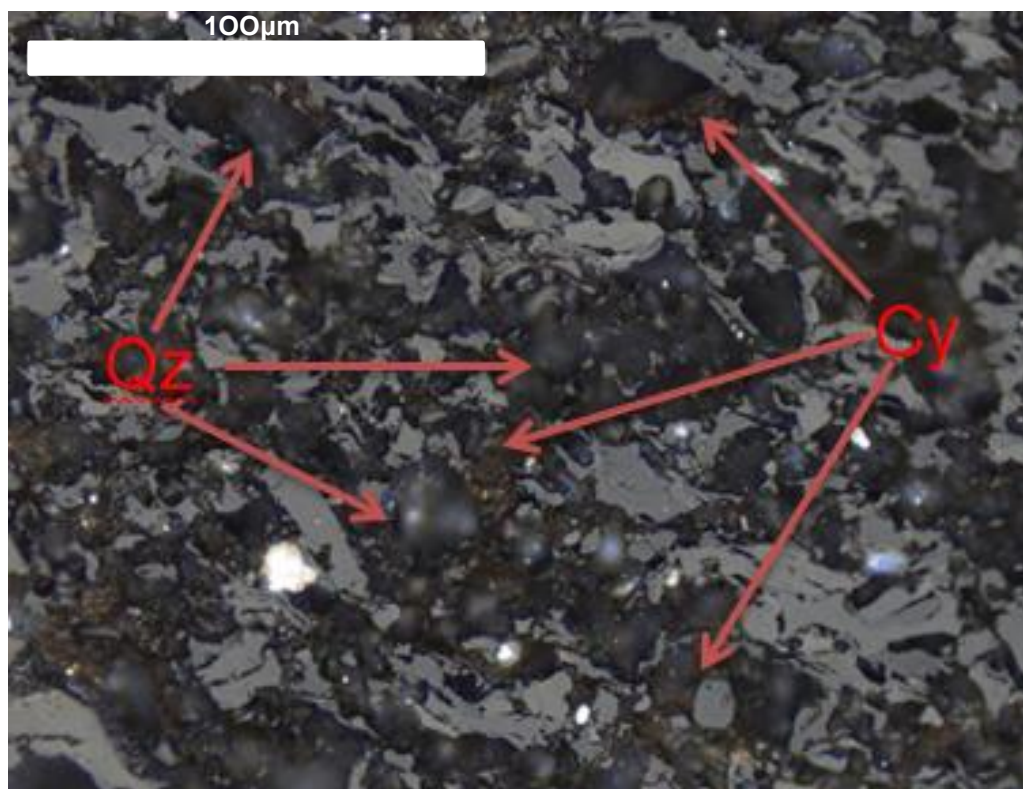


Figure 4.9: Photomicrographs of quartz (Qz) in association with clay (Cy) from sample G; under reflected polarised light in transparent oil immersion examination.

The carbonate fraction of the Moiyabana coal samples constitutes about 3.67 vol% on average of the mineral matter concentration. Calcite is the dominant carbonate mineral observed, occurring as epigenetic cleat-, fracture-, and/or cell-filling mineral in coals of the study area (Fig. 4.10). Another carbonate mineral, siderite, although hardly observed in study area coal samples, occurs as re-precipitated small irregular bodies or patches, and fissure and cell cavity fillings on vitrinite macerals.

The concentration of the sulphide fraction is lower in Moiyabana coal samples (0.20 to 4.97 vol%) with an average value of 1.91 vol%. Pyrite is the most abundant sulphide mineral. It occurs in several forms such as disseminated particles, discrete grains, framboidal bodies, massive replacement and fissure fillings (Fig. 4.11).

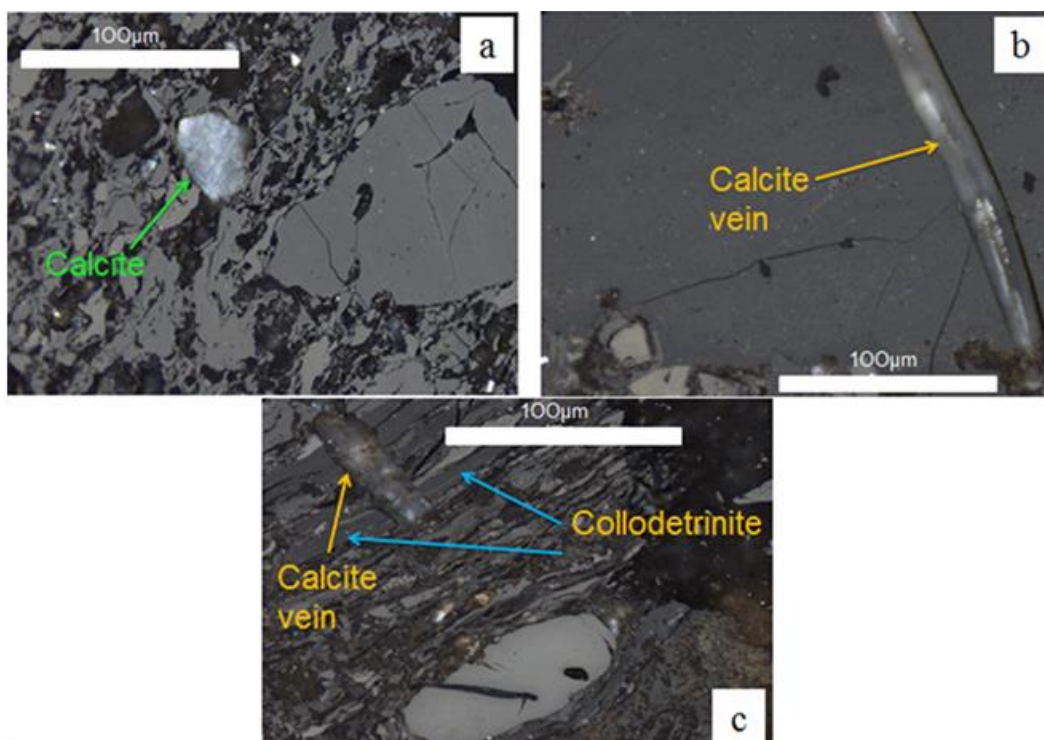


Figure 4.10: Photomicrographs of calcite occurrences in Moiyabana coal samples, under reflected polarised light in transparent oil immersion examination. **a)** Detrital calcite-restricted to occur in the organic matter. **b)** Fracture filling – occurring in veins and/or cleats. **c)** Epegenitic calcite vein cutting across collodetrinite.

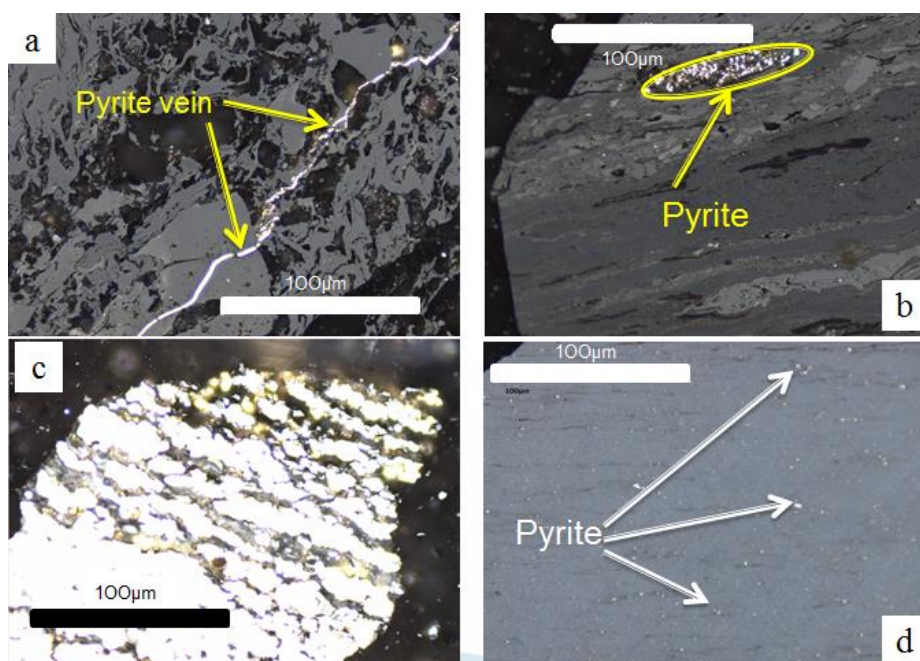


Figure 4.11: Photomicrographs of pyrite occurrences in Moiyabana coal samples, under reflected polarised light in transparent oil immersion examination. **a)** Epigenetic pyrite in cleats and veins. **b)** Syngenetic pyrite clustered together in vitrinite. **c)** Massive pyrite nodule as cell filling cavities. **d)** Syngenetic pyrite disseminated in vitrinite.

4.2 Proximate Analysis

Results of proximate analysis of the Moiyabana coal samples are presented in Table 4.5 and elaborated in Figure 4.12. The graphs in Figure 4.12 are based on the temperature intervals as explained in the methodology chapter in section 3.4.

The moisture content values of the Moiyabana coal samples range from 1.23 to 2.40%a.d.b with an average value of 1.67%, with the lowest values recorded in samples G and H (seam F2b) and the highest value recorded in sample D (seam F1c). The volatile matter content ranges from 13.29 to 19.31 % with an average value of 15.86%a.d.b. The highest volatile matter content value was determined in the bottom seam (F1a), whereas the lowest value determined in the seams immediately above it (F1b) (Table 4.5). The fixed carbon content ranges from 21.48 to 54.27 %, with an average value of 41.21%a.d.b. The lowest fixed carbon content value was located on an F1b seam (sample C) and the highest in the F1a seam (sample A), and lastly ash content varies from 24.47 to 63.94 % with an average of 41.26%a.d.b. Ash content values are inversely proportional to fixed carbon content values; highest where fixed carbon recorded the lowest value and lowest where fixed carbon recorded the highest value (Fig. 4.13).

No major variations are evident for the moisture and volatile matter content between the coal samples and the same parameters are generally low (Fig. 4.12). Conversely, variations can be spotted on the ash and fixed carbon contents between coal samples (Fig. 4.12). Qualitatively, the Moiyabana coal samples have moderate fixed carbon content and generally high ash content (Fig. 4.12).

Table 4.5: Results of the proximate analysis and Calorific Value (CV) of Air Dried Coal Seams.

Sample ID	Moisture Content (%)	Volatile Matter (%)	Fixed Carbon (%)	Ash (%)	CV (Mj/kg)
H	1.2	16.7	45.0	37.1	16.7
G	1.2	15.4	22.2	61.2	9.3
F	1.3	15.6	44.3	39.0	15.7
E	1.7	16.0	51.2	31.1	19.0
D	2.4	14.2	40.2	43.2	13.9
C	1.3	13.3	21.5	64.0	5.3
B	2.3	16.5	51.0	30.2	19.2
A	1.9	19.3	54.3	24.5	22.5

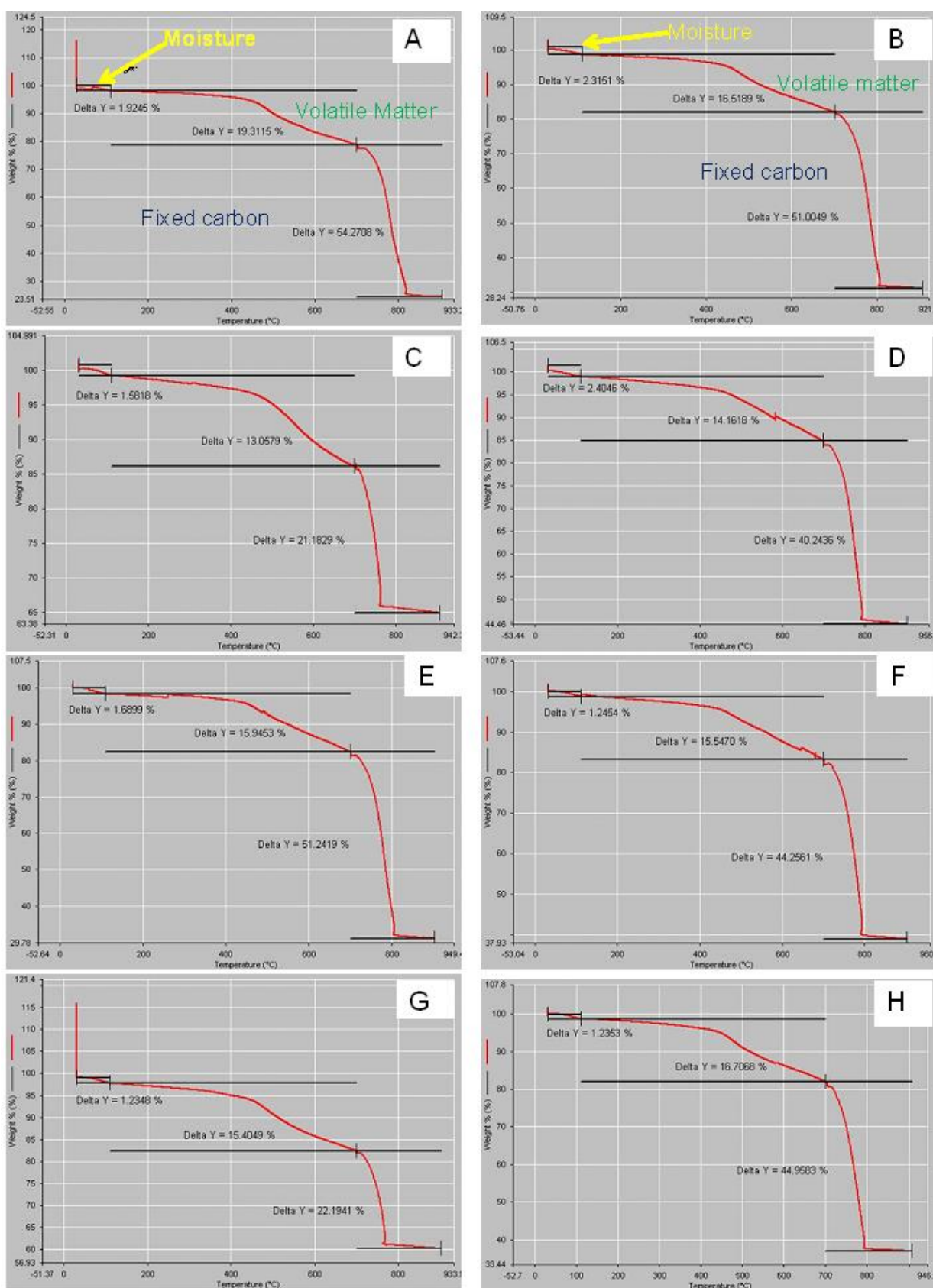


Figure 4.12: Proximate analysis graphs depicting weight (%) loss as temperature (°C) increases.

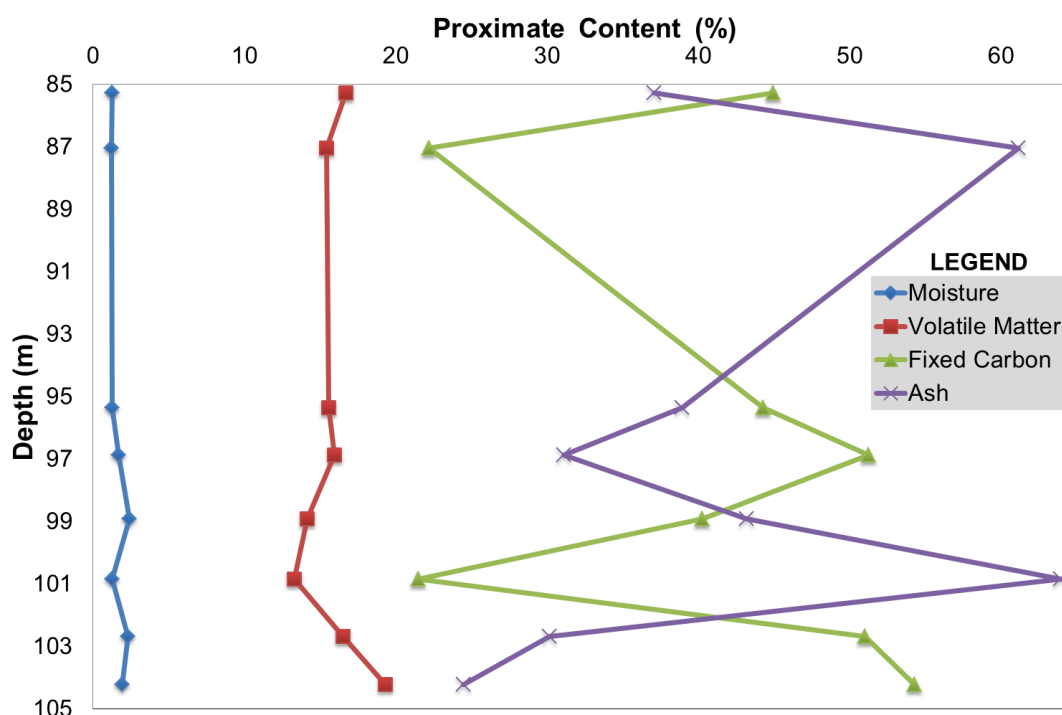


Figure 4.13: Plots of proximate component's variations with coal samples of Moiyabana Coalfield.

4.3 Ultimate and Sulphur Analysis

Results of ultimate and sulphur analysis of the Moiyabana coals are tabulated in Table 4.6. Total sulphur values vary from 0.41 to 2.04%a.d.b with an average value of 1.06%a.d.b. The highest value for sulphur was determined at sample F while the lowest value was recorded at sample G. These two values occur at the top seams of the study area. The carbon content values range from 18.10 to 57.70% with an average value of 39.16%. The carbon content is observed to be high in the basal seam (F1a) and lowest at seam F1b. The hydrogen content values vary from 1.46 to 3.10% with an average value of 2.19%. The highest hydrogen content value was recorded in the bottom seam at sample A while the lowest value was determined at the top seam (sample G). The nitrogen and oxygen content ranges from 0.35 to 1.19% with an average value of 0.81% and 6.88 to 9.27% with an average value of 7.58% respectively. The nitrogen and oxygen lowest values were recorded in the middle seam (sample C) and top seam (sample H) respectively, while the highest values of these elements were both determined in the middle seam (samples E and G respectively).

Table 4.6: Elemental composition (a.d.b) of Moiyabana coals, eastern Botswana.

Sample ID	A	B	C	D	E	F	G	H
C (%)	57.70	50.50	18.10	37.80	51.50	36.70	20.70	40.30
H (%)	3.10	2.65	1.50	2.01	2.55	2.02	1.46	2.25
N (%)	1.15	1.04	0.35	0.87	1.19	0.67	0.38	0.79
O (%)	7.86	7.10	7.35	7.42	7.48	9.27	7.25	6.88
S (%)	1.79	0.51	1.00	1.10	1.18	2.04	0.41	0.48
O/C	0.14	0.14	0.41	0.20	0.15	0.25	0.35	0.17
H/C	0.05	0.05	0.08	0.05	0.05	0.06	0.07	0.06

a.d.b-air dry basis

The total sulphur, hydrogen, nitrogen and oxygen contents in the Moiyabana coals exhibit no vertical variation from top to bottom seam (Fig. 4.14). Carbon is the only element in the ultimate parameters which show significant vertical variation between different seams across the Moiyabana coal seams (Fig. 4.14). Highest at the basal seam, it sharply decreases in the seam above (F1b) to record the lowest value in sample C, then records an increase through sample D to E, after which a gradual decrease through sample F to G. Lastly, an increase in carbon content occurs between sample G and H (Fig. 4.14).

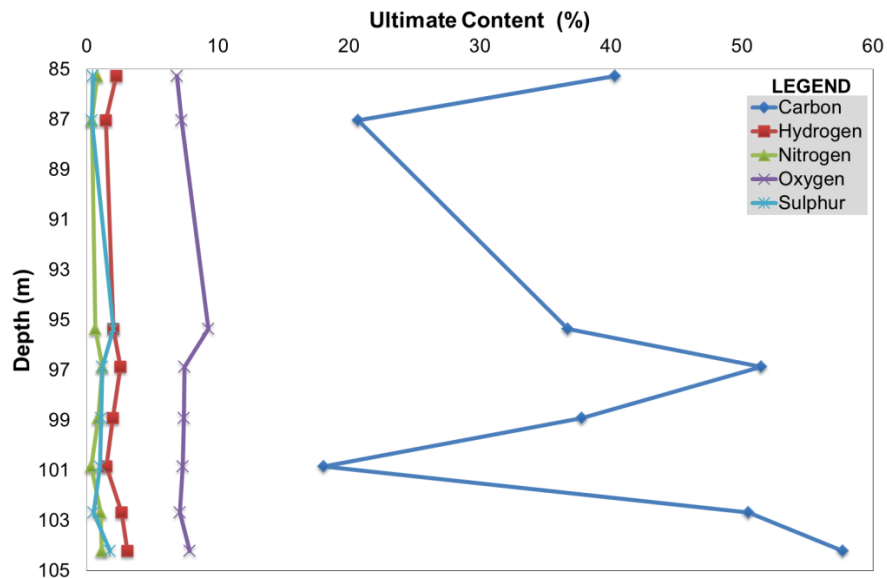


Figure 4.14: Plots of ultimate constituents content vertical variation with coal samples of Moiyabana Coalfield.

The carbon content is a very important component of ultimate analysis, mainly because it implies the total organic content (TOC) in a coal seam (Thomas, 2013). It is equally important to establish the relationship of carbon with other coal

components for better assessment of coal quality. The vertical trend variation is observed between carbon and sulphur as shown in Figure 4.14. A slight relationship exists between carbon and hydrogen, evidently where carbon is increasing a slight increase also occurs in hydrogen and similarly, where the carbon content is decreasing, hydrogen records a slight decrease (Fig. 4.14). Nitrogen and oxygen vary differently from carbon across all the seams in the study area. A linear relationship is observed between carbon and fixed carbon as their increase and decrease between coal seams perfectly correlate (Fig. 4.15 and Fig. 4.16d). As for carbon and ash content, an inverse relationship is observed between them (Fig. 4.16a).

Correlation coefficients (Fig.4.16) show varying relationships between carbon and proximate parameters. A very good/perfect correlation is observed between carbon and ash yield and fixed carbon both with a value of $R^2=0.99$ (Fig. 4.16a and Fig. 4.16d respectively). There is a poor correlation between C and moisture content ($R^2=0.24$) (Fig. 4.16b), whereas volatile matter content has a moderate correlation ($R^2=0.56$) with carbon. These findings agree with most of the coals in literature.

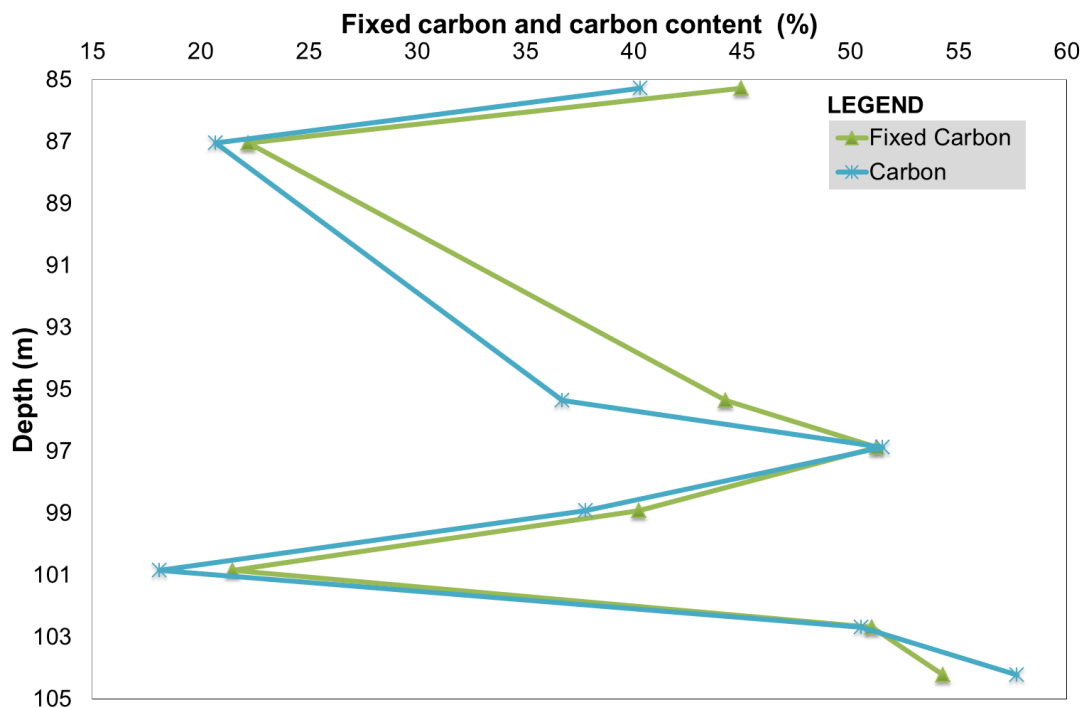


Figure 4.15: Fixed carbon compared to carbon on a vertical scale for Moyabana coal samples.

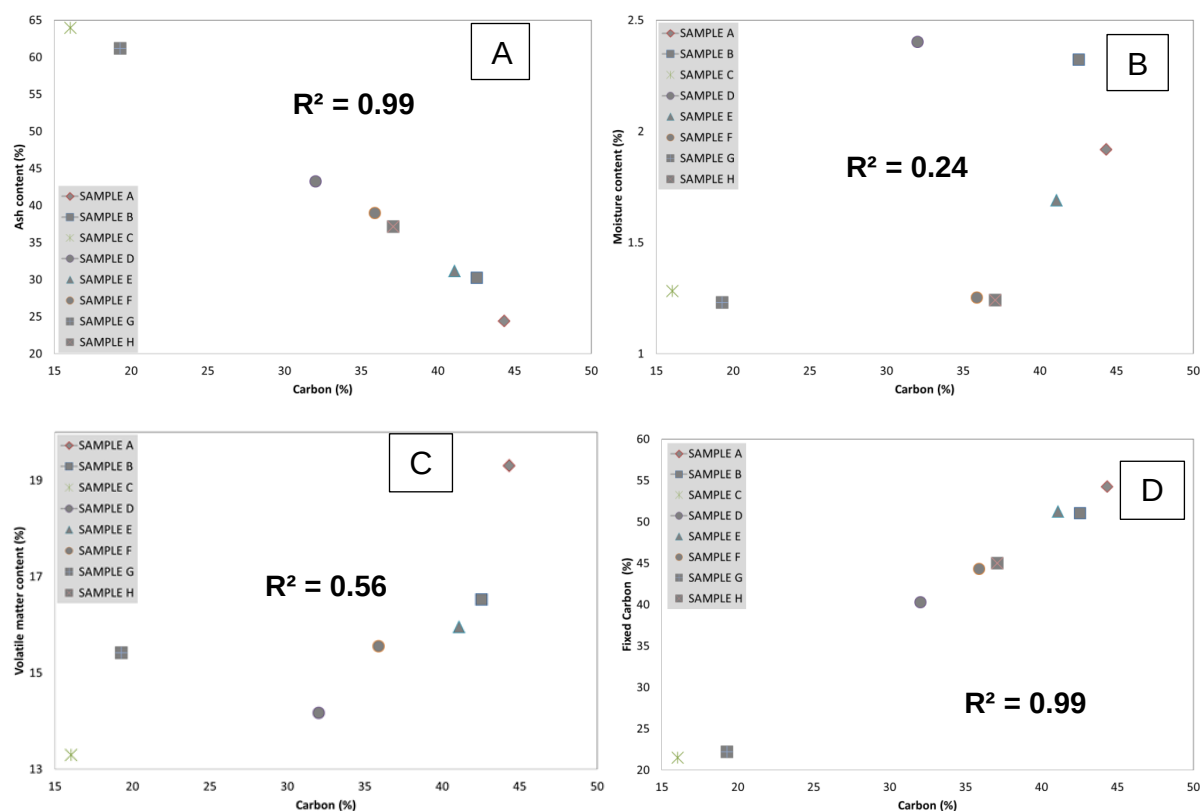


Figure 4.16: X-Y scattergrams proximate analysis components vs. carbon of Moiyabana coal samples, eastern Botswana.

4.4 Calorific Value (CV)

The CV results for Moiyabana coal samples are shown in Table 4.5. In these samples, CV values range from 5.32 to 22.50 MJ/kg with maximum value recorded in sample A (F1a) and lowest value recorded in sample C (F1b). As expected, a linear relationship between fixed carbon and CV is observed (Fig. 4.17). It is further observed that CV is slightly related to volatile matter content. A slight inverse relationship is observed between moisture content and CV. It is also observed that ash and CV are inversely related (Fig. 4.17 and Fig. 4.19). The coal samples with high ash content have been observed to yield minimum CV while coal with low ash content yields maximum CV (Fig. 4.17 and Fig. 4.19).

Another variation relationship is established for CV and ultimate constituents in Figure 4.18. It is observed that CV is directly related to carbon content (Fig. 4.18 and 4.19). An increase in carbon content similarly shows an increase in CV

content. No significant vertical variation is observed in the study area for other ultimate parameters (hydrogen, oxygen and nitrogen) with CV. As expected, very good correlation coefficients are observed between CV and carbon and ash (both $R^2=0.97$) (Fig. 4.19).

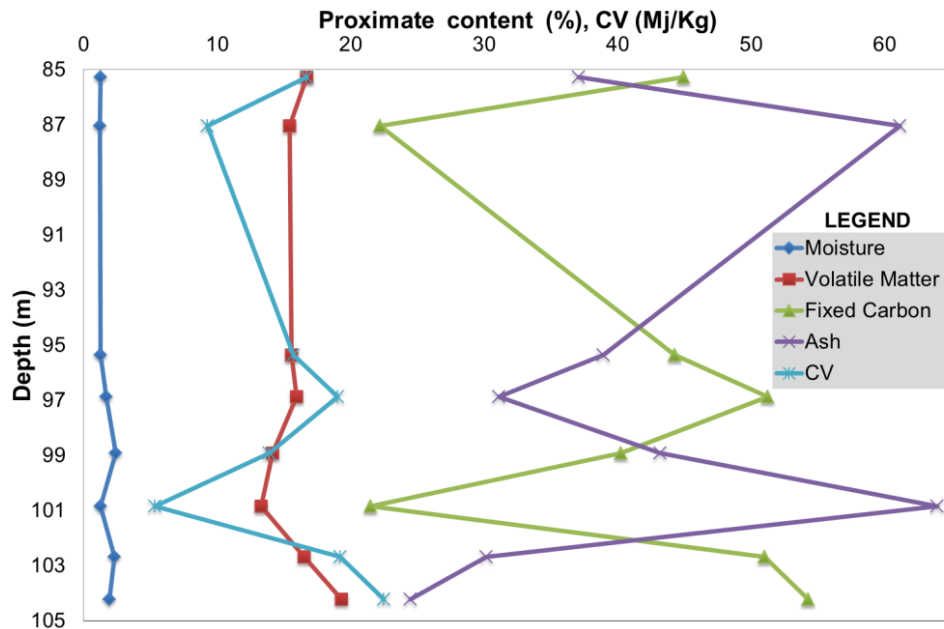


Figure 4.17: Variation relationship between calorific value and proximate parameters for coal samples of Moiyabana Coalfield.

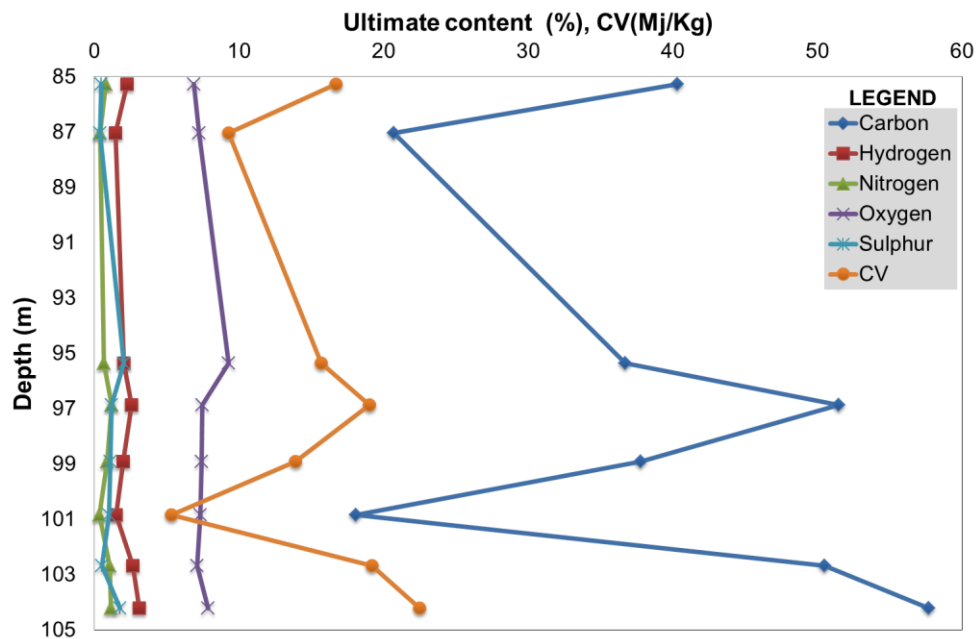


Figure 4.18: Variation relationship between calorific value and ultimate parameters in coal samples of Moiyabana Coalfield.

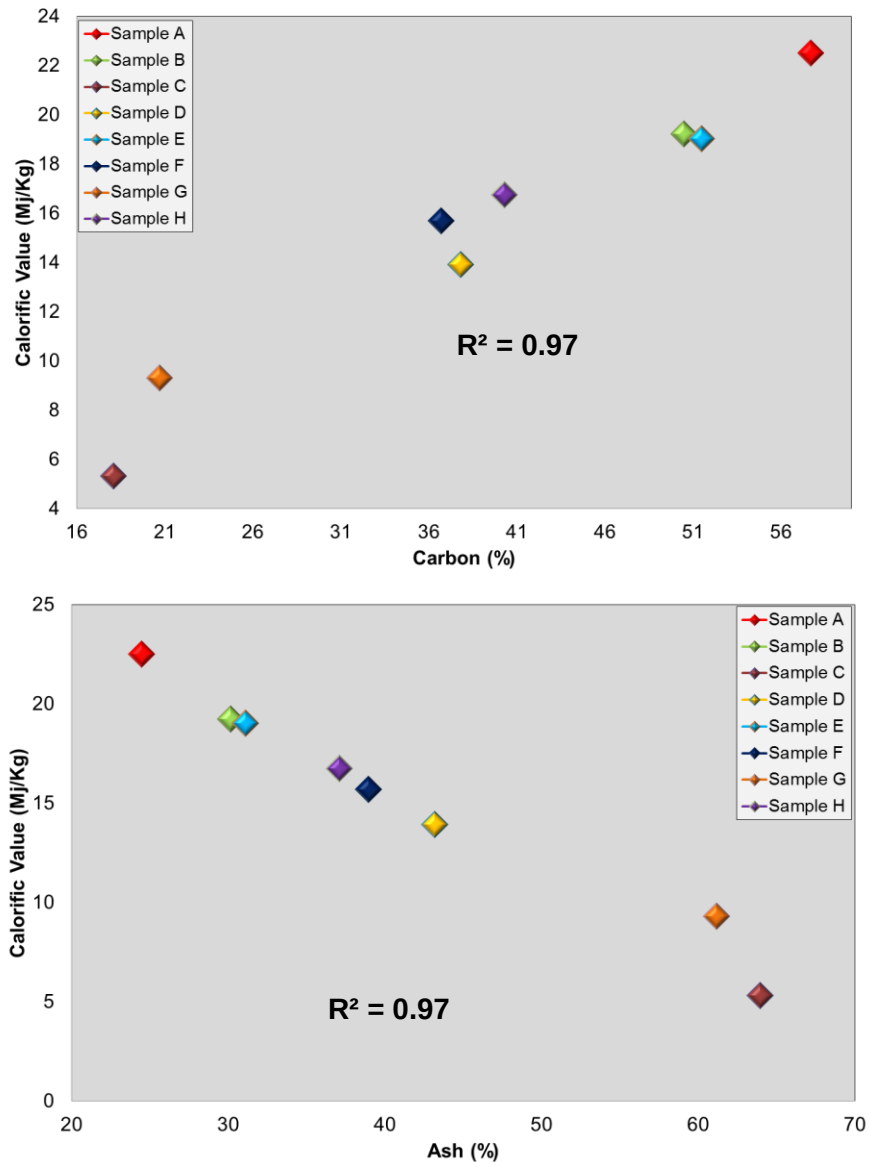


Figure 4.19: Plots of calorific value vs. carbon (top) and ash (bottom).

4.5 Geochemistry

4.5.1 Major elements

Table 4.7 shows the major elements of the Moiyabana coal samples determined from the ash. The SiO_2 and Al_2O_3 are the dominant components of the inorganic constituents and vary between 36.19 to 67.81% and 16.76 to 32.41 % respectively. The content of TiO_2 ranges from 1.51 to 2.11%, and the Fe_2O_3 content ranges from 2.79 to 12.68%. The MgO and CaO content vary between

0.83 to 4.63%, and 1.13 to 9.12% respectively. The Na₂O content varies from 0.11 to 1.45% K₂O, 0.51 to 1.21%, P₂O₅, 0.05 to 2.10 and 0.82 to 11.25 % SO₃. Lastly, MnO recorded the lowest amount ranging between 0.02 to 0.8%.

Table 4.7: Major elements analytical data (weighted %) of Moiyabana coal samples, eastern Botswana.

Sample ID	SiO ₂	TiO ₂	Al ₂ O ₃	Fe ₂ O ₃	MnO	MgO	CaO	Na ₂ O	K ₂ O	SO ₃	P ₂ O ₅
H	57.2	1.6	24.2	6.3	0.8	2.8	4.8	0.1	0.6	2.4	0.6
G	67.8	2.0	24.0	2.8	0.3	1.0	1.5	0.1	0.7	1.2	0.1
F	50.9	1.6	27.2	9.8	0.1	2.0	4.0	0.2	0.5	5.4	0.3
E	44.6	1.7	27.2	12.7	0.1	2.6	5.4	0.3	0.5	4.9	2.1
D	62.5	2.1	16.8	6.2	0.5	2.2	4.3	0.2	0.8	3.4	0.4
C	59.7	2.0	32.4	3.1	0.0	0.8	1.1	0.1	0.8	0.8	0.1
B	59.3	1.9	28.7	3.8	0.4	1.5	1.7	1.5	1.2	1.3	0.1
A	36.2	1.5	25.7	12.0	0.2	4.6	9.1	0.1	0.9	11.3	0.1

4.5.1.1 Comparison of vertical variation of major elements and ash content in borehole JVP20

In the Moiyabana JVP20 borehole, vertical distribution of ash content is systematically high in the areas where the coals were interbedded with carbonaceous mudstone and/or mudstone (e.g. sample C and G) and lower in samples which contained almost purely coal (e.g. sample A and B) as shown in Figure 4.20. Generally, the basal seam (F1a) has lowest ash content, whereas the seam overlying it (F1b) has very high ash content (Table 4.5 and Fig. 4.20).

4.5.1.1.1 SiO₂, Al₂O₃, TiO₂

As expected, the major oxide of Si varies vertically concordantly with ash yield (Fig. 4.20). They both show a general decrease from upper seam to middle seams where they record the lowest values. They are observed to increase in the seam above the basal seam (F1b) and dramatically dropping in the basal seam (F1a) (Fig. 4.20).

Aluminium oxide shows a steady increase from the upper seam to the middle seams where it follows a similar trend as the ash and SiO₂ (Fig. 4.20). The trends

for Si and Al oxides are consistent with most coals around the world that shows predominance of silicate minerals (clays and quartz) (Fig. 4.7).

There appears to be no relationship between ash yield and TiO_2 in Moiyabana coal samples (Fig. 4.20), and it is almost constant from the top to the basal seam.

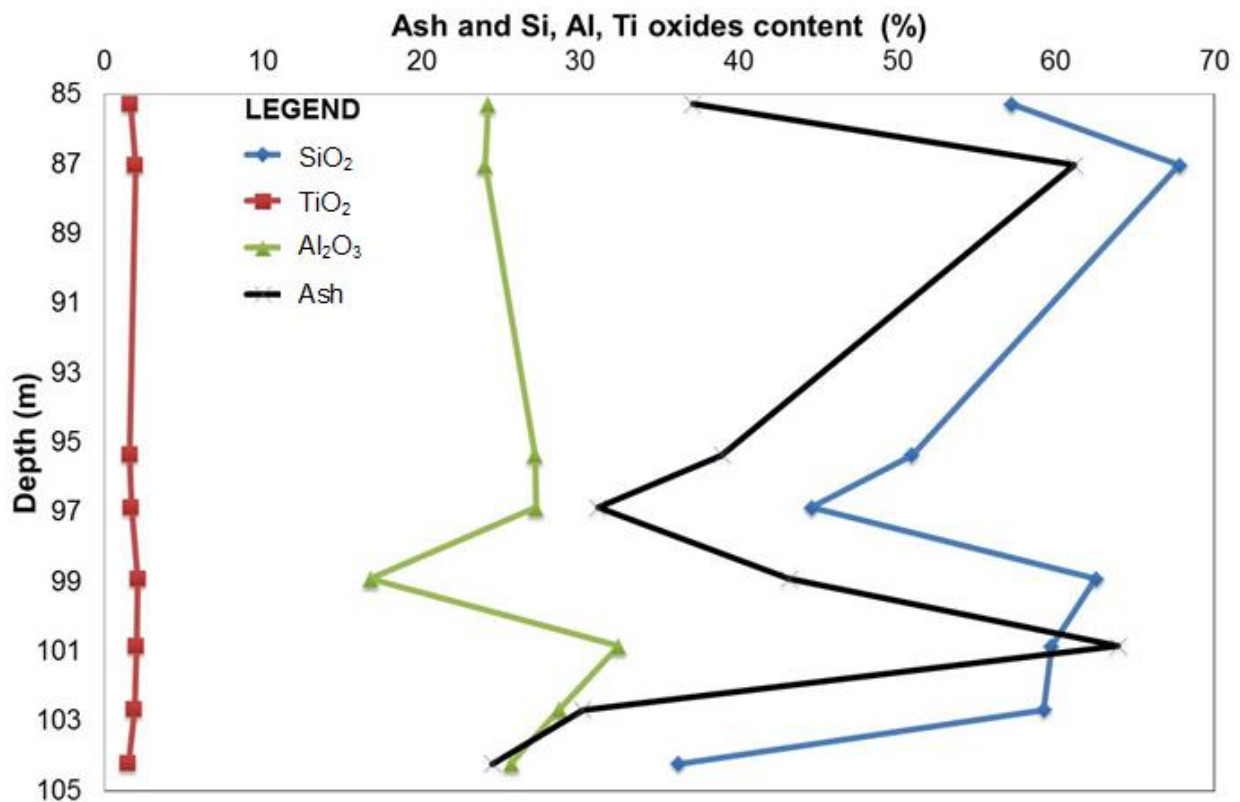


Figure 4.20: Plots of major elements content showing positive vertical correlation with ash.

4.5.1.1.2 Mg, Mn, Fe, Ca and S oxides

The Mg, Fe, Ca and S oxides show a different trend from that of the ash yield and SiO_2 (Fig. 4.20 and Fig. 4.21). Their trends show an inverse relationship when compared to the ash and SiO_2 ; where the oxides of Mg, Fe, Ca and S record the highest values, ash yield and SiO_2 record the lowest values and vice versa. These oxides show a similar trend across the profile with lowermost values observed in the upper seam. A general increase in the middle seams is evident, with highest values recorded in this portion of the coal strata. The Moiyabana coal samples are characterized by low contents of Ca, Mg and Mn which in turn reflect the low carbonate content (Table 4.7 and Fig. 4.21). Sulphur oxide contents in

these coal samples are considered to be generally moderate, with most values about 5% and less with only one exception at sample A (F1a) recording a very high value (11.25 %). When compared to the total S values from ultimate analysis and sulphide values from petrographic analysis, these values variation with depth is consistent across the three data sets (Fig. 4.22).

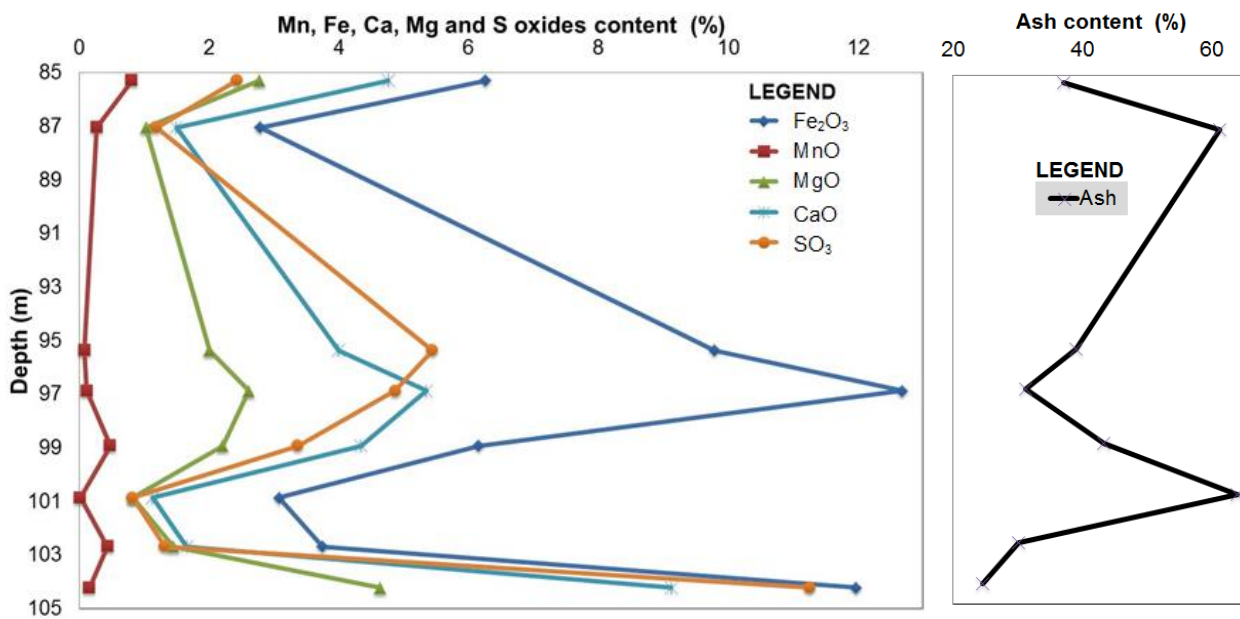


Figure 4.21: Plots of major oxides content compared to ash content in vertical scale showing negative correlation with ash yield.

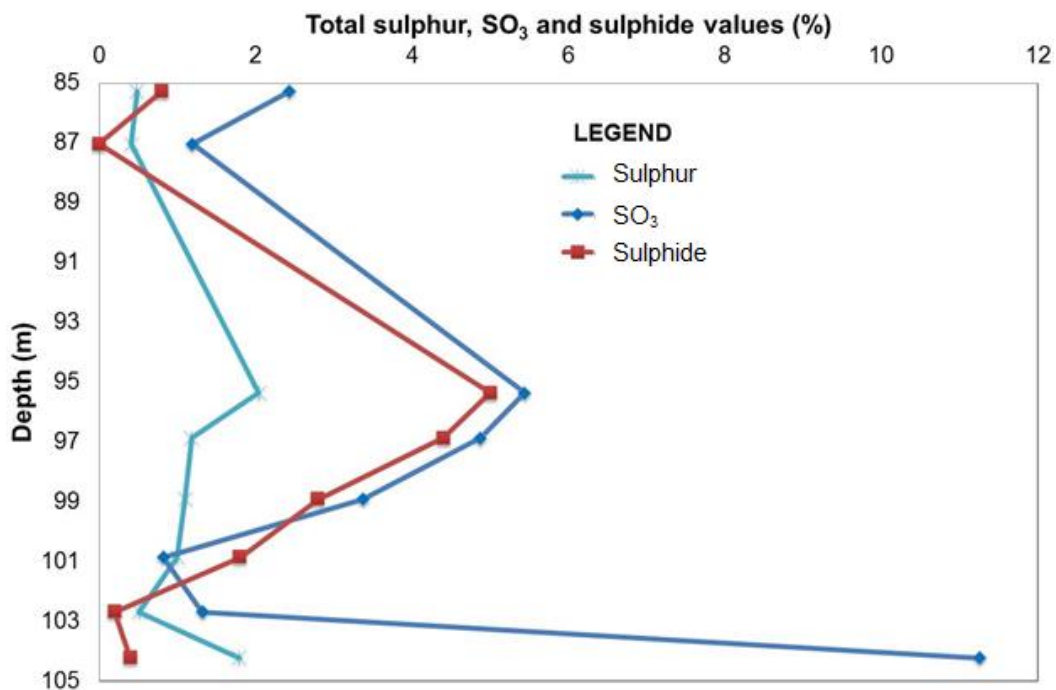


Figure 4.22: Plots of total S, SO₃ and sulphide values in vertical scale.

4.5.1.1.3 Na, K and P oxides

The oxides of Na, K and P occur generally in low quantity as compared to other oxides. They show no systematic vertical variation trend with the ash content (Fig. 4.23). The Na and K oxides are both observed to occur uniformly from the upper seam to the middle seam, where a slight increase in Na oxide is followed by a decrease down to F1b seam. At the same middle seam, K oxide recorded a gradual increase down to F1b seam. Na_2O and K_2O both recorded the highest values in the basal seams; the K contents are elevated more in the basal layers than in the top portions of the seams. This is interesting because K is one of the essential nutrients for plant growth and usually expected to be high in the top layers of peat due to plant bioaccumulation mechanisms. The P_2O_5 content is observed to decrease from upper seam to middle seam, where it records no variation with depth. A sharp increase is noticed towards the middle bottom portion to again suddenly dropping in the basal seam.

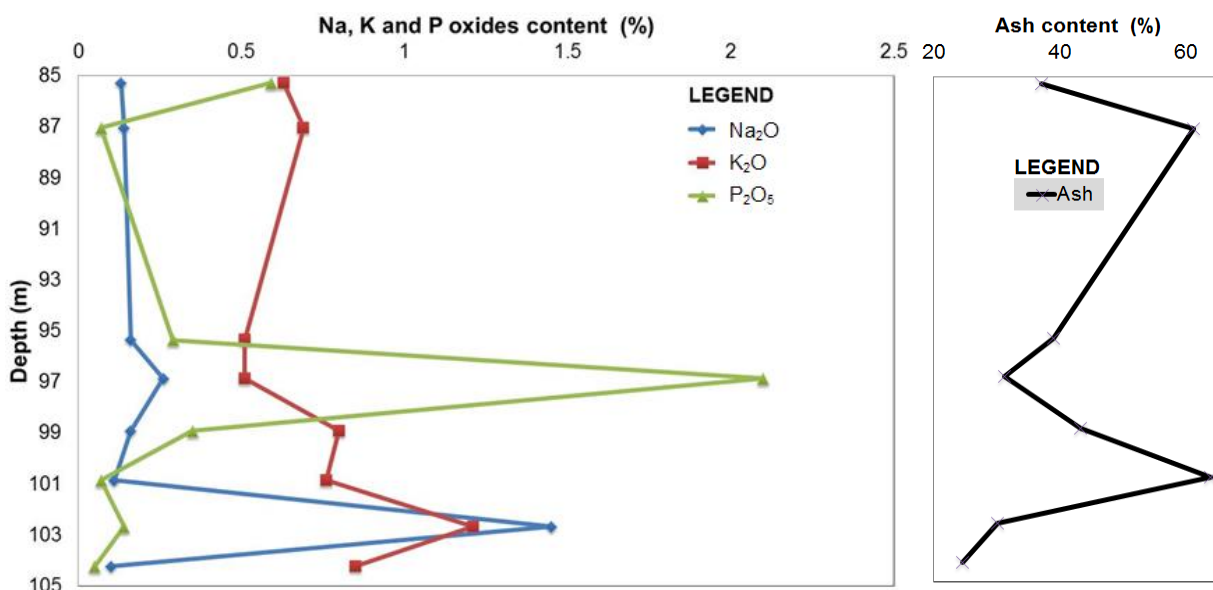


Figure 4.23: Plots of major oxides content showing vertical variation with ash of Moiyabana coal samples.

4.5.1.2 Correlations with ash contents and inter-element correlations

The results in Table 4.9 were obtained following Shao et al. (2004) system of oxides association in coal based on Pearson's correlation coefficients between oxides. The values were calculated using the formula below:

$$r = \frac{n(\sum xy) - (\sum x)(\sum y)}{\sqrt{(n \sum x^2 - (\sum x)^2)(n \sum y^2 - (\sum y)^2)}}$$

Where, n donates the number of samples analysed, x and y represent oxides being compared. An easy way of how to calculate the variable in the formula is given in an exemplary form in Table 4.8.

Table 4.8: Example of how to calculate values of Pearson's correlation coefficient for oxides in coal using SiO₂ and TiO₂ oxides (after Shao et al., 2004).

Sample I	x	y	XY	x ²	y ²
	SiO ₂	TiO ₂			
A	36.19	1.51	54.65	1309.72	2.28
B	59.25	1.89	111.98	3510.56	3.57
C	59.73	2.01	120.06	3567.67	4.04
D	62.54	2.11	131.96	3911.25	4.45
E	44.59	1.70	75.80	1988.27	2.89
F	50.92	1.59	80.96	2592.85	2.53
G	67.81	1.96	132.91	4598.20	3.84
H	57.20	1.63	93.24	3271.84	2.66
	438.23	14.40	801.56	24750.35	26.26

r= 0.80 (see Table 4.9)

Table 4.9: Values of Pearson's correlation coefficient for major elements in coal.

	SiO ₂	TiO ₂	Al ₂ O ₃	Fe ₂ O ₃	MnO	MgO	CaO	Na ₂ O	K ₂ O	SO ₃	P ₂ O ₅	Ash
SiO ₂	1											
TiO ₂	0.80	1										
Al ₂ O ₃	-0.21	-0.22	1									
Fe ₂ O ₃	-0.90	-0.74	-0.06	1								
MnO	0.35	0.06	-0.52	-0.28	1							
MgO	-0.83	-0.72	-0.26	0.78	0.10	1						
CaO	-0.85	-0.70	-0.29	0.84	0.01	0.99	1					
Na ₂ O	0.16	0.16	0.25	-0.29	0.20	-0.25	-0.35	1				
K ₂ O	0.18	0.33	0.08	-0.44	0.22	-0.09	-0.20	0.79	1			
SO ₃	-0.89	-0.71	-0.12	0.84	-0.28	0.91	0.93	-0.30	-0.14	1		
P ₂ O ₅	-0.35	-0.22	0.01	0.58	-0.06	0.17	0.24	-0.09	-0.5	0.08	1	
Ash	0.78	0.62	0.11	-0.75	-0.10	-0.79	-0.75	-0.31	-0.20	-0.71	-0.36	1

4.5.1.2.1 Si, Ti and Al oxides

By using the Pearson's correlation coefficient values calculated in Table 4.9, it is established that: oxides of Si ($r=0.78$), Ti ($r=0.62$) and Al ($r=0.11$) positively correlate with the ash yield (Table 4.9), although only Ti has a positive correlation ($r=0.8$) with Si. Al₂O₃ shows a negative correlation with both TiO₂ and Si ($r=-0.22$ and $r=-0.21$ respectively), clearly signifying no elemental relationship between these elements (Table 4.9). These correlations confirm, from a geochemistry perspective, that clay minerals and quartz are the controlling factors on ash composition and concentrations (Fig. 4.24). In Figure 4.25, it is evident that the silicate minerals plot varies vertically concordantly with an ash yield as expected.

4.5.1.2.2 Mg, Fe, Mn, Ca, Na, K, S and P oxides

The major oxides of Mg ($r=-0.79$), Fe ($r=-0.75$), Ca ($r=-0.75$), Mn ($r=-0.10$), Na ($r=-0.31$), K ($r=-0.20$), S ($r=-0.71$) and P ($r=-0.36$) all show a negative correlation with the ash yield (Table 4.9) signifying organic mode of occurrence for these elements. The strong positive correlation ($r=0.84$) between Fe₂O₃ and S suggest that these elements are associated with regard to their occurrence in the study area coal seams. This association probably reflects less organic S in the coal seams, rare occurrence of siderite and the common occurrence of pyrite. The

observed pyrite occurrence is consistent with petrographic result where it occurred in multiple forms. Another significant observation between total S and Fe_2O_3 is that total S increase and decrease are concordant with that of Fe_2O_3 , which is evident in both having a similar vertical trend (Fig. 4.21).

Another interesting observation is that CaO and MgO correlate well ($r = 0.93$ and $r = 0.91$ respectively) with the SO_3 (Table 4.9). This clearly indicates the same mode of occurrence of these elements in the study area coal seams, which probably suggest their sulphate occurrence. P_2O_5 and CaO show a weak positive correlation ($r = 0.24$) between these elements, probably suggesting a lower phosphate occurrence. Conversely, a strong negative correlation exists between S and oxides of Si and Ti with $r = -0.89$ and $r = -0.71$, respectively (Table 4.9), suggesting that these elements are not associated with sulphur.

K and Na oxides show positive correlation ($r = 0.79$), signifying the same host minerals in the study area coal seams. This probably suggests the presence of Na and K oxides in coal to mainly occur in the feldspar group of minerals (Baïoumy, 2009).

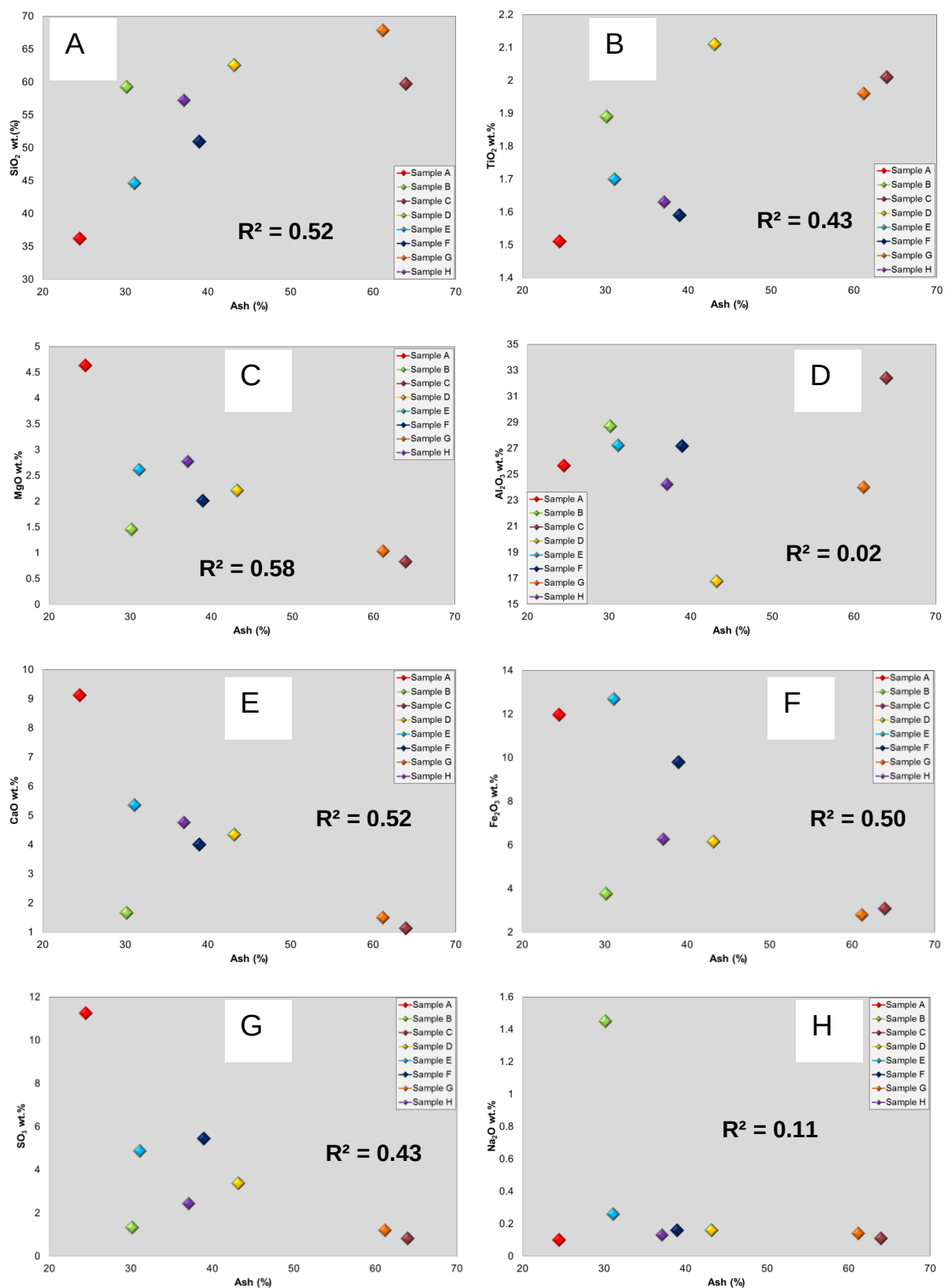


Figure 4.24: Major oxides showing relationship with ash content.

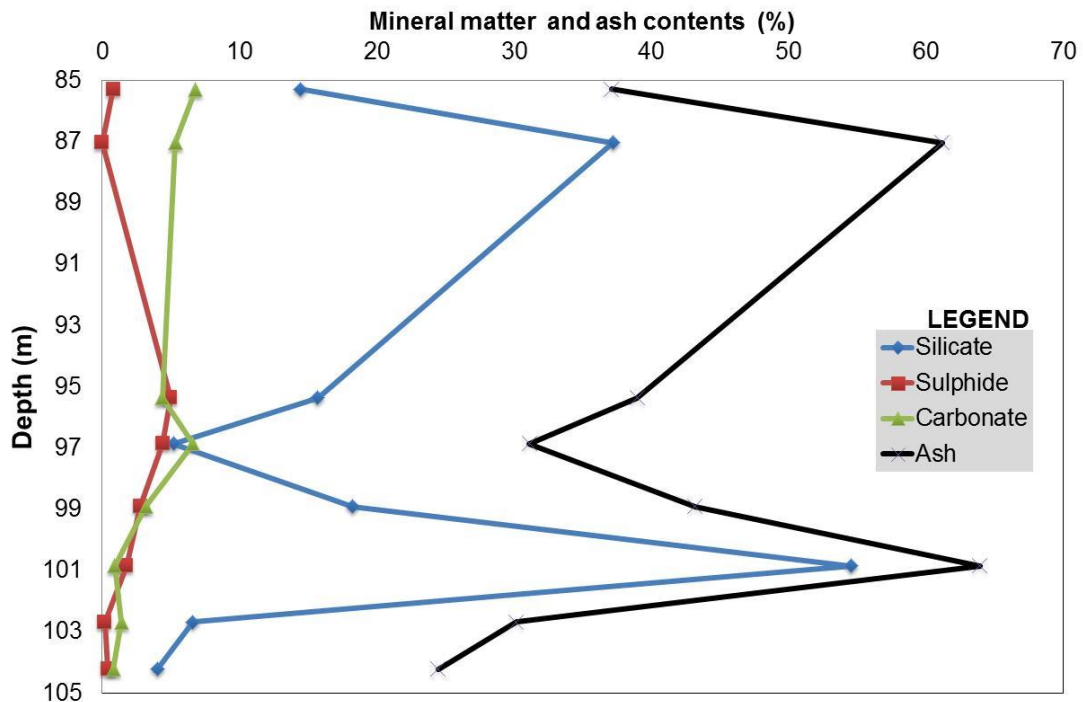


Figure 4.25: Plots of mineral matter concentrations and ash yield showing vertical variation in Moiyabana coal samples.

4.5.2 Trace elements

Very little is known about the trace element concentration and distribution in coals of Botswana. The main focus of this section is to fulfil some of the more important objectives of this study, which include: determination of the vertical distribution and concentrations of trace elements in the Moiyabana borehole core for JVP20, delineate their modes of occurrence, and discuss their potential environmental risks.

4.5.2.1 Trace element concentrations and vertical variation

The abundance of trace elements in the Moiyabana coals, in comparison to other coals is given in Table 4.10. For discussion purposes, the trace elements are categorised into three groups adopting US National Research Council (NRC) classification for trace elements based on environmental quality and health (Coal Trading Blog (CTB), 2011) as follows: trace elements of major (As, B, Cd, Pb, Hg, Mo, Se, Th and U), moderate (Cr, V, Cu, Zn, Ni and F) and minor (Ba, Sr, Mg, Co, Sb, Li, Cl and Ge) concern (Table 4.10).

Table 4.10: Trace elements data (ppm) of Moiyabana borehole core, JVP20 compared to other coals.

Concern level	Trace Elements	Sample A	Sample B	Sample C	Sample D	Sample E	Sample F	Sample G	Sample H	Coal Clarke's ^a	Highveld coals (Range) ^b	Average global values ^c
Major	As	2.0	2.1	2.2	3.6	2.5	5.0	2.9	2.0	9.0±0.7	1.0–4.7	5.0
	B	14.6	26.4	36.9	28.4	24.6	26.3	43.0	40.2	47±3	-	-
	Cd	0.1	0.2	0.3	0.3	0.2	0.2	0.2	0.2	0.20±0.04	0.05–0.51	0.6
	Hg	0.2	0.2	0.2	0.5	0.4	0.9	0.5	0.3	0.10±0.01	0.04–0.27	0.12
	Mo	3.2	1.4	1.6	1.8	1.4	2.3	2.6	2.0	2.1±0.1	0.4–<5	5
	Pb	21.1	21.1	36.1	42.8	25.6	47.3	23.4	30.6	9.0±0.7	4.2–11	25
	Se	0.4	0.4	0.6	0.6	0.9	0.4	0.4	0.9	1.6±0.1	<0.5–1.5	3
	Th	6.8	1.1	0.9	3.9	10.1	5.8	0.8	9.3	3.2±0.1	-	-
	U	2.9	4.7	4.9	5.7	5.1	5.1	4.7	6.3	1.9±0.1	-	-
Moderate	Cr	41.3	58.4	111.9	92.7	54.0	88.8	54.9	52.6	17±1	23–69	10
	Cu	18.2	24.1	30.8	27.1	19.9	24.3	36.5	26.6	16±1	9–16	15
	F	169.7	232.9	261.9	224.2	389.9	223.8	284.4	307.3	82±6	-	-
	Ni	16.2	13.8	33.6	23.6	16.4	28.1	16.2	17.1	17±1	12–23	15
	V	71.7	45.3	95.8	71.1	38.6	47.6	66.5	50.8	28±1	23–37	25
	Zn	94.6	45.3	39.9	51.6	62.2	87.1	32.2	26.9	28±2	8–19	50
Minor	Ba	69.4	180.5	291.6	349.2	484.0	383.2	347.8	490.9	150±10	-	-
	Cl	260.3	247.2	128.1	174.2	219.4	163.0	110.6	221.4	340±40	-	-
	Co	9.3	4.1	2.3	4.3	5.4	6.3	4.7	6.4	6.0±0.2	5–12	5
	Ge	2.4	2.3	3.21	2.5	2.6	2.8	3.3	3.4	2.4±0.2	-	-
	Li	17.8	16.6	89.7	36.3	16.8	37.6	44.8	27.1	14±1	-	-
	Mn	259.7	105.5	52.1	149.2	288.5	227.8	104.3	224.4	71±5	84–117	50
	Sb	0.2	0.3	0.4	0.6	0.4	0.8	0.5	0.4	1.00±0.09	<0.09–0.3	3.0
	Sr	69.4	158.6	144.8	246.7	396.3	205.5	107.7	251.7	100±7	-	-

^a Ketris and Yudovich (2009)^bWagner and Hlatshwayo (2005)^c Zhang et al. (2004)

4.5.2.1.1 Trace Elements of Major Concern

The trace elements As (2.02 to 4.98 ppm with an average value of 2.78 ppm), Cd (0.13 to 0.29 ppm with an average value of 0.22 ppm), Hg (0.18 to 0.88 ppm with an average value of 0.39 ppm), Mo (1.42 to 3.24 ppm with an average value of 2.05 ppm) and Se (0.38 to 0.93 ppm with an average value of 0.59 ppm) in the major concern category, vary slightly with depth in the Moiyabana coal samples (Fig. 4.26). Conversely, both B (14.57 to 43.00 ppm with an average value of 30.04 ppm) and Pb (21.10 to 47.26 ppm with an average value of 30.98 ppm) are the only elements in this category with significant variation trends with depth. The B concentration increases between sample H and G. This element records a decreasing trend between samples G, F to E, after which it increases again between samples E, D to C. Between samples C, B and A, the B content is observed to show a decreasing trend. Lead on the other hand is observed to decrease between sample H and G. It records an increasing trend between sample G and F, after which a sharp decrease is observed between sample F and E, which is followed by a sharp increase between sample E and D. The Pb content between sample D, C, B and A is observed to record a decreasing trend. The elements Th and U are included in this category because of their radioactive nature. The Th values range from 0.77 to 10.09 ppm, with an average value of 4.82 ppm. The concentration of this element is observed to increase with increasing depth (Fig. 4.26). The U content ranges from 2.86 to 6.28 ppm with an average value of 4.92 ppm. A general trend of decreasing from top to bottom seam of this element is observed (Fig. 4.26).

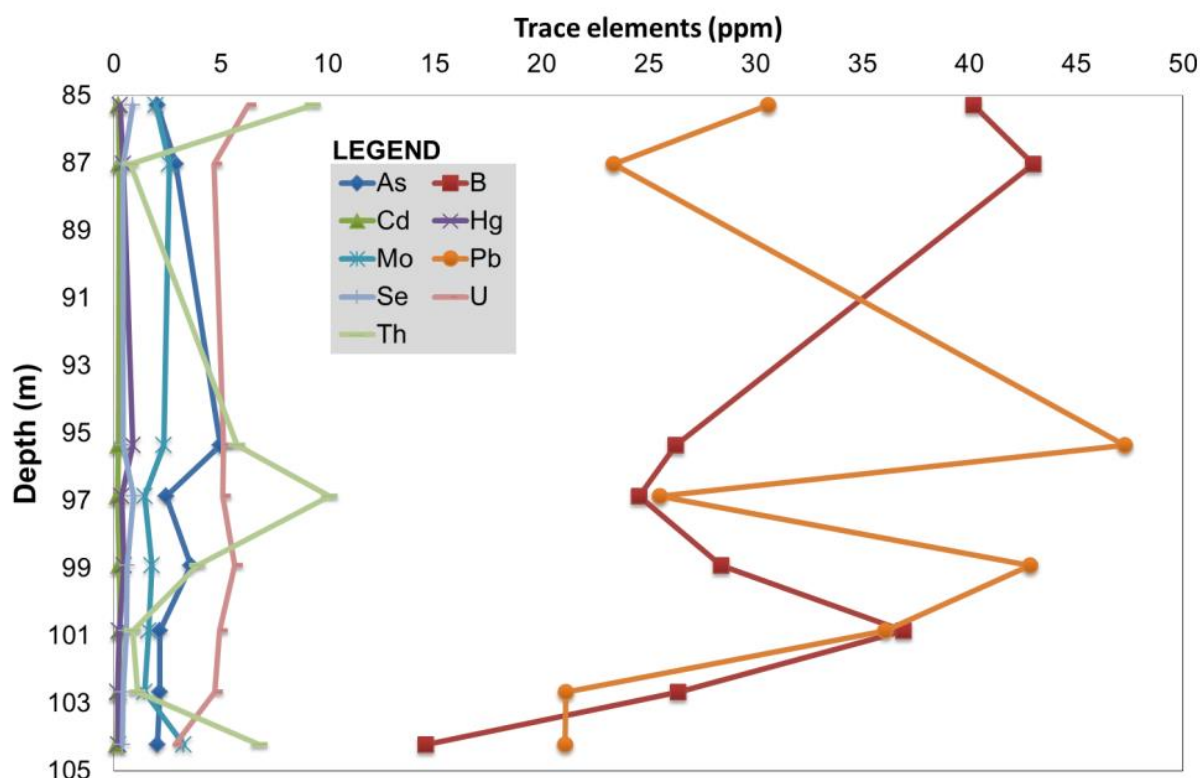


Figure 4.26: Plots of trace elements of major concern concentrations showing vertical variation in Moiyabana borehole core, JVP20.

4.5.2.1.2 Trace Elements of Moderate Concern

In this category, most trace elements have a similar vertical variation trend with depth (Fig. 4.27). These elements which include Cr (ranging from 41.29 to 111.90 ppm with an average value of 69.33 ppm), Cu (ranging from 18.19 to 36.52 ppm with average value of 25.94 ppm), Ni (ranging from 13.79 to 33.59 ppm with an average value of 20.62 ppm) and V (ranging from 38.60 to 71.67 ppm with an average of 60.93 ppm) have a general increase from sample H, G to F. They all record a decrease between sample F and E, after which an increase is observed from sample E, D to C. Most of these elements decrease between sample C, B to A, except for V which decreased between samples C to B and observed to increase between samples B to A.

A different trend is observed for only two elements (Zn and F) in this category. The Zn (ranging from 26.93 to 94.61 ppm with an average of 54.98 ppm) is observed to increase from sample H, G to F and decrease between samples F, E, D to C. This element is observed to increase from sample C, B to A. Fluorine

(ranging from 110.61 to 260.34 ppm with an average value of 190.54 ppm) on the other hand, decreases from the top seam to middle seam, where a dramatic increase and then decrease again in the middle seam is observed. Decrease in content is evident for F through the basal seam.

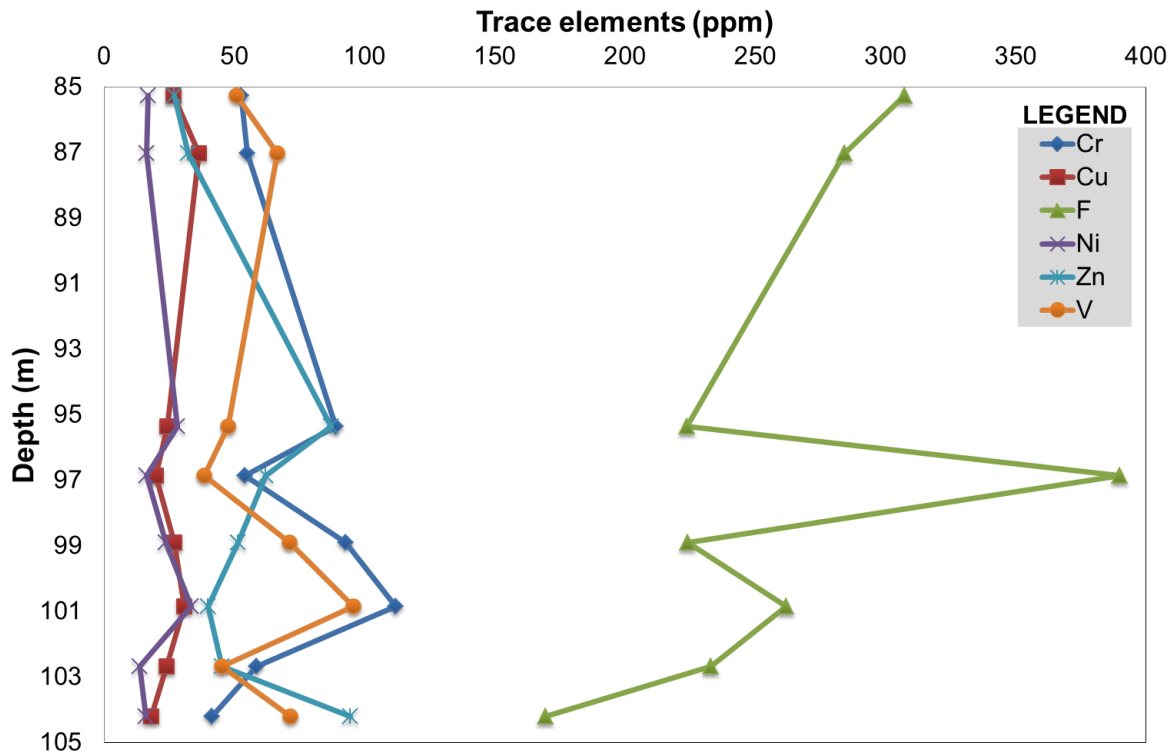


Figure 4.27: Plots of trace elements of moderate concern concentrations showing vertical variation in Moiyabana coal samples.

4.5.2.1.3 Trace Elements of Minor Concern

In this category, the values of Sb (0.18 to 0.81 ppm with an average value of 0.44 ppm), Co (2.30 to 9.25 ppm with an average value of 6.35 ppm) and Ge (2.31 to 3.39 ppm with an average value of 2.82 ppm) in Moiyabana coal samples do not show any systematic trend with depth (Fig. 4.28). The Li concentration level in Moiyabana coal samples ranges from 16.83 to 89.72 ppm with an average value of 35.84 ppm and it is observed to record almost same values in the top seams through the upper portion of the middle seam and basal seam (Fig. 4.28). The Li highest increase is evident in the lower portion of the middle seams (at sample C).

A concordant trend is observed in the elements Ba (69.38 to 490.88 ppm and an average value of 324.57 ppm), Cl (110.61 to 260.34 ppm with an average value of 190.54 ppm), Mn (52.11 to 288.46 ppm with an average value of 205.17 ppm) and Sr (69.38 to 396.28 ppm with an average value of 197.58 ppm) from the top seams down to the lower middle seam, whereupon Cl and Mn record an increase in the basal seam whereas Ba and Sr are observed to decrease in the same seam (Fig. 4.28).

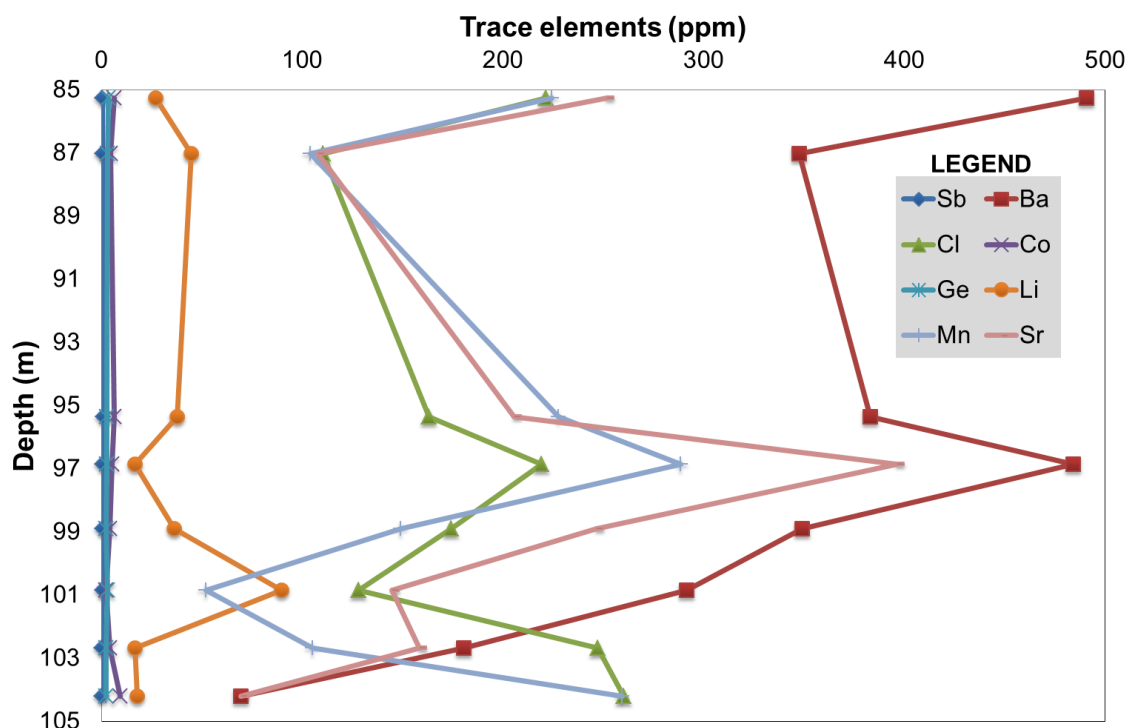


Figure 4.28: Plots of trace elements of minor concern concentrations showing vertical variation in Moiyabana coal samples.

4.5.2.2 Mode of occurrence of trace elements

The Pearson's correlation coefficient was also used for trace elements association with ash yield (Table 4.11). Moosavirad et al. (2012) categorised trace elements concentrations in coals based on the values of correlation coefficients between elements and ash yield, whereupon they divided the elements into four groups. Adopting the same criteria as Moosavirad, analysed trace elements in Moiyabana coal samples were grouped as follows: The first group of elements (B, Cd, Cu, Ge, Li and Mo) shows very high positive correlations with ash yield ($r > 0.7$). The second group of elements (Cr, Ni and V) shows medium positive correlations with

ash yields ($r=0.51-0.69$). The third group of elements (Ba, Pb, Sb and U) exhibits a weak positive correlation with ash yield ($r=0.21-0.50$) and fourth group of elements (As, Hg and F) shows the lowest positive correlation with ash yields ($r<0.20$). The variable positive correlations with ash content of the above trace elements affirm from a geochemistry perspective the inorganic affinity of these elements in Moiyabana coal samples.

All the other trace elements show negative correlation with the ash content, thus suggesting association to the organic affinity. Although showing organic mode of occurrence, they also vary insofar as the correlation coefficient with ash is concerned. Adopting almost similar criteria used above, trace elements associated with the organic fraction are grouped as follows: Group one elements (Cl, Co and Mn), those that have high negative correlation with ash ($r>-0.7$), group two elements (Zn and Th) show negative moderate correlation with ash ($r=-0.51-0.69$), group three elements (Sr) show weak negative correlation with ash ($r=-0.21-0.50$) and group four elements (Se) show very weak negative correlation with ash.

Table 4.11: Values of Pearson's correlation coefficient for trace elements in Moiyabana coal samples.

	Ash	As	B	Ba	Cd	Co	Cr	Cu	Ge	Hg	Li	Mn	Mo	Ni	Pb	Sb	Se	Sr	Th	U	V	Zn	Cl	F
Ash	1																							
As	0.2	1																						
B	0.8	-0.1	1																					
Ba	0.3	0.3	0.5	1																				
Cd	0.8	0.2	0.6	0.2	1																			
Co	-0.7	0.0	-0.6	-0.3	-0.9	1																		
Cr	0.6	0.4	0.2	0.1	0.8	-0.7	1																	
Cu	0.9	0.1	0.9	0.2	0.7	-0.6	0.4	1																
Ge	0.8	0.0	0.9	0.5	0.4	-0.2	0.2	0.7	1															
Hg	0.2	1.0	0.0	0.4	0.1	0.1	0.3	0.1	0.1	1														
Li	0.9	0.1	0.5	0.1	0.8	-0.6	0.8	0.6	0.6	0.0	1													
Mn	-0.7	0.1	-0.6	0.2	-0.9	0.8	-0.5	-0.8	-0.3	0.2	-0.7	1												
Mo	0.9	0.1	-0.2	-0.5	-0.5	0.7	-0.4	-0.1	0.1	0.2	-0.1	0.3	1											
Ni	0.6	0.4	0.2	0.1	0.6	-0.5	0.9	0.3	0.3	0.4	0.8	-0.4	-0.1	1										
Pb	0.3	0.8	0.1	0.4	0.5	-0.2	0.8	0.1	0.2	0.7	0.4	-0.1	-0.2	0.8	1									
Sb	0.4	0.9	0.2	0.5	0.4	-0.2	0.5	0.3	0.2	0.9	0.2	0.0	-0.1	0.5	0.8	1								
Se	-0.1	-0.3	0.2	0.8	-0.1	-0.1	-0.1	-0.1	0.3	-0.2	-0.1	0.4	-0.5	-0.1	0.0	-0.1	1							
Sr	-0.2	0.1	0.0	0.8	-0.1	-0.1	0.0	-0.3	-0.1	0.2	-0.3	0.5	-0.6	-0.1	0.2	0.2	0.8	1						
Th	-0.6	0.0	-0.3	0.4	-0.7	0.6	-0.4	-0.7	-0.1	0.1	-0.5	0.9	0.1	-0.3	0.0	-0.1	0.7	0.6	1					
U	0.4	0.2	0.6	0.8	0.5	-0.5	0.3	0.4	0.4	0.3	0.2	-0.1	-0.6	0.2	0.5	0.5	0.6	0.6	0.2	1				
V	0.6	-0.2	0.2	-0.4	0.5	-0.3	0.5	0.4	0.3	-0.3	0.8	-0.6	0.2	0.6	0.2	-0.2	-0.3	-0.6	-0.5	-0.3	1			
Zn	-0.6	0.4	-0.9	-0.4	-0.6	0.7	-0.1	-0.7	-0.6	0.4	-0.3	0.6	0.5	0.1	0.1	0.1	-0.4	-0.1	0.3	-0.6	-0.1	1		
Cl	-0.9	-0.4	-0.7	-0.3	-0.7	0.6	-0.6	-0.9	-0.6	-0.4	-0.8	0.6	0.0	-0.6	-0.5	-0.6	0.1	0.1	0.5	-0.3	-0.5	0.4	1	
F	0.1	-0.2	0.4	0.8	-0.1	-0.3	-0.2	0.1	0.3	0.0	0.0	0.2	-0.6	-0.2	-0.2	0.0	0.8	0.8	0.4	0.5	-0.4	-0.5	-0.1	1

4.6 Summary

The macroscopic results for Moiyabana coal samples are mainly banded showing predominance of dull (durain) coal over the other lithotypes, whereas the microscopic results show the high quantity of inertinite and mineral matter, and low proportions of both vitrinite and liptinite macerals. Collotelinite is the most abundant maceral of the vitrinite group, inertodetrinite is the dominant maceral in the inertinite group, whereas the liptinite group is dominated by sporinite macerals in the study area coal samples. Vitrinite reflectance analysis places the coals in the medium rank bituminous C, borderline B category (ECE-UN, 1998). Petrographically observed inorganic constituents include: silicates, carbonates and sulphides. As expected, silicates dominate the mineral content of these coal samples. Proximate results show generally low contents of moisture and volatile matter, moderate content of fixed carbon and high content of ash yield. The low to medium total sulphur values were observed for Moiyabana coal samples. The elemental proportions for these coal samples generated through ultimate analysis shows the low content of nitrogen and moderate to high contents of carbon, hydrogen and oxygen. A very good correlation was observed between carbon and ash and fixed carbon ($R^2=0.99$), whereas poor ($R^2=0.24$) and moderate ($R^2=0.56$) correlation were observed between carbon and moisture and volatile matter contents respectively. Low content of calorific values with very good correlations between CV and both ash and carbon are characteristics of these coal samples. The analysed major oxides show predominance of SiO_2 and Al_2O_3 . Oxides of Si, Al and Ti positively correlate with ash yield whilst oxides of Mg, Mn, Fe, Ca, S, Na, K and P negatively correlate with ash yield. The results of analysed trace elements in Moiyabana coal samples show positive correlation for elements B, Cd, Cu, Ge, Li, Mo, Cr, Ni, V, Ba, Pb, Sb, U, As, Hg and F with ash yield, suggesting inorganic affinity, and negative correlation of Mn, Th, Cl, Sr, Se and Zn with ash yield implying organic mode of occurrence for these elements.

5 DISCUSSION

In this chapter, the necessary interpretations are drawn based on the results presented in Chapter 4. In accordance with the aims and objectives of this dissertation; the important results obtained during this project will be discussed comprehensively. In Section 5.1, Lithotypes, macerals and mineral constituents and vitrinite reflectance are discussed extensively to objectively characterise the individual coal samples based on the rank. The economic potential of the coal with respect to its technological usage is mentioned. The deciphering of the environmental condition during the formation of these coals using petrographic faces forms part of this section. In Section 5.2, coal quality components which include proximate, ultimate and calorific value analysis results are discussed to establish which chemical properties aid in the ultimate usability and utilisation of these coal samples. Calorific value, carbon and ash content relationship with other chemical coal constituents are discussed in detail for correct categorisation of coal rank. Lastly, geochemical results (Section 5.3) are discussed to fruitfully delineate the vertical distribution of ash chemistry (major elements) and trace elements.

5.1 Coal Petrography

5.1.1 Macroscopic Character

The macroscopic descriptions of coal in the field help in the overall characterisation and classification of the coal samples under investigation. As outlined earlier, the geological log of JVP20 was produced following the Australian geological coal logging standard. It is established from the geological log (coal section or seam) that Moiyabana coals can be classified broadly as humic/banded coal based on the different megascopic types encountered in this coal samples. These different megascopic types, clearly suggest banded bright and dull coal lithotypes typical of humic coal.

5.1.2 Microscopic Character

5.1.2.1 Maceral Analysis

The Moiyabana coals are dominated by the inertinite group of macerals. Bustin et al. (1983) demonstrates, through the use of H/C and O/C atomic ratio, that during coalification the three maceral groups become enriched in carbon and each maceral group follows a distinctive coalification path. Figure 5.1 illustrates the coalification path taken by the Moiyabana coal samples using H/C and O/C atomic ratio plot with values of H+O obtained from Table 4.6. This plot resembles the inertinite coalification path demonstrated by very low content of both H/C and O/C atomic ratio (van Krevelen, 1961).

The high inertinite content (Table 4.4) in the study area is not surprising as most of the coals investigated in Botswana have been reported by many authors (e.g. Smith, 1984; Carney et al., 1994) to contain a high proportion of inertinite, which is typical of most southern Africa Gondwana coals (Cairncross, 2001; Hancox, 2016). The pre-dominance of inertinite group macerals is attributed to the prevalence of oxidizing conditions for a prolonged period of time, when accumulated vegetal matter was exposed to the surface (Cadle et al., 1993; Snyman and Botha, 1993). As the dominant maceral from this group is inertodetrinite, it is deduced that there was significant maceral reworking via floods leading to the inclusion of high content (>15%) of mineral matter plus deposition of the vegetation.

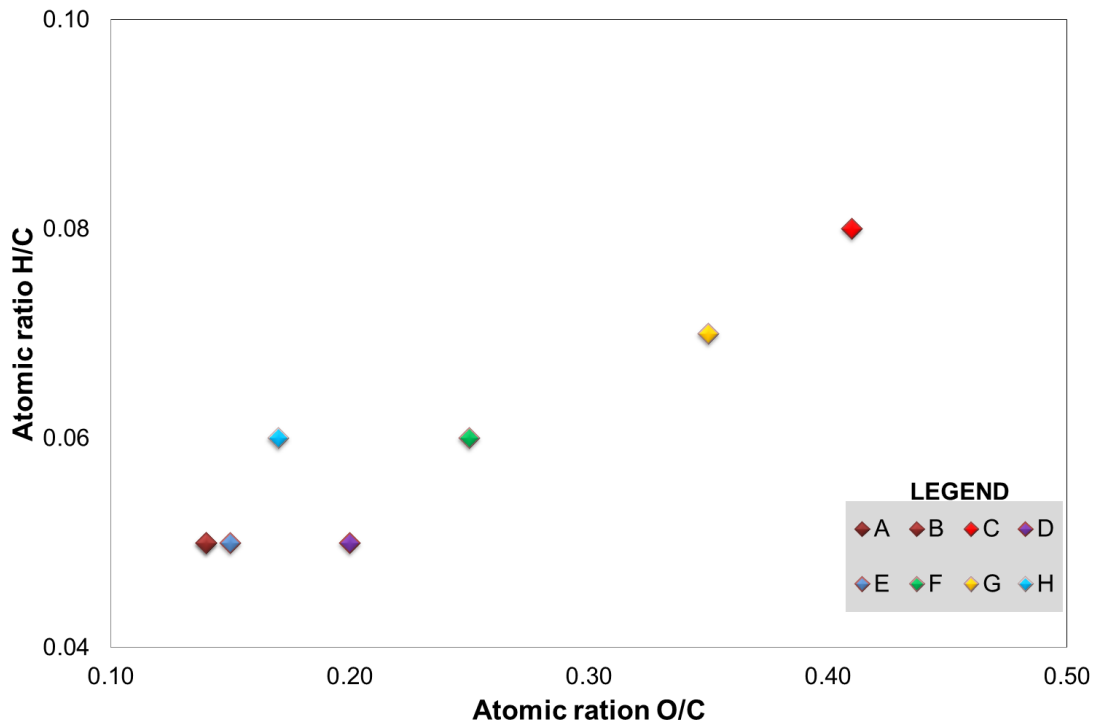


Figure 5.1: Plot showing the coalification path for Moiyabana coal samples (Bustin et al. (1983) based on van Krevelen, 1961). Values from Table 4.6.

5.1.2.2 Vitrinite Reflectance Measurements (Coal rank)

The vitrinite reflectance values are used to characterise coal seams in terms of rank. The use of the histograms (Fig. 4.6) helps to differentiate coal particles based on reflectance differences present within a coal sample. A lower standard deviation is preferable because the samples are less likely to cause problems during technological usage (Thomas, 2013) e.g. during coal combustion, a sample with similar coal constituent properties will respond consistently in the furnace, and hence smooth operation of the power station.

Vitrinite reflectance values are also used to derive conclusions as to the stage reached by coal during transformation of the vegetal material ultimately into graphite, through major stages like peat, lignite, bituminous coal and anthracite (Thomas, 2013). The mean reflectance values indicate a medium rank C bituminous coal, borderline B coal using ECE-UN rank classification, which are

suitable for the metallurgical industry in terms of rank. However, the vitrinite content is very low.

The increased reflectance values of Moiyabana coal samples might have been affected by surrounding dolerite dyke intrusions present within the vicinity of the coalfield as reported by Carney et al. (1994) and Ermanovics (1978). Based on Carney et al. (1994), suggestion of Pre-Karoo dykes during the formation of the coal seams in Botswana, it could imply that the dykes might have increased the temperature of the surrounding rock through contact metamorphism, hence directly increasing the temperature during coalification. Due to increased temperature, coal will lose volatiles, oxygen, methane and water leading to the local development of higher rank coal (Thomas, 2013). It must be stated, though that, due to lack of evidence to support the temperature increase resulting in deep burial of coal, Moiyabana coals can only be associated with direct contact heat from dolerite intrusions which is in agreement with earlier findings by Carney et al. (1994). Thus, heat induced by contact metamorphism during the dolerite intrusions could have subsequently led to the slight rank increase in Moiyabana coal samples.

5.1.2.3 Mineral matter

In most coals, the silicate fraction consists of clay minerals on average up to 60-80% of the total mineral matter associated with coal (Thomas, 2013; Ward, 2002). Clay minerals found in coal, are to some extent, associated with volcanic activity during Middle and Upper Ecca sub-group times. During these geologic times, thin tonsteins mudstones (kaolinite bearing mudstone) in coal horizon were formed (Carney et al., 1994). The earlier work by Smith (1984) recognised four tonsteins in the Morupule-Kgaswe Coalfield and similarly, Smith (1984) noticed tonsteins in various sectors of Mmamabuala Coalfield. Based on the previous investigation by different authors of the composition of tonsteins, i.e. Carney et al. (1994) they were found to be kaolinite rich rather than containing illite. The current study is in agreement with the above findings as kaolinite is mainly distributed along the bedding and in the cleats of the macerals (Fig. 4.8) and is probably derived from the weathering of detrital minerals such as feldspars, micas, and amphiboles

(Carney et al., 1994). The occurrence of kaolinite in the pores and the cell cavities of coal macerals is interpreted to be indicative of authigenic origin (Fig. 4.8).

The presence of high content of silica in the Moiyabana coals is due to the high quartz content observed in all coal samples. It is mainly of detrital origin (Thomas, 2013) based on the fact that it is dominantly occurring as individual grains (Fig. 4.9) as opposed to cleat- and/or fracture-fillings (Ward, 2002) which would otherwise suggest syngenetic alteration of aluminosilicate.

In the study area coals, calcite is the dominant carbonate mineral. Dai and Chou (2007) and Ward (2002) report that it is a common epigenetic cleat-, fracture-, and/or cell-filling carbonate mineral in coal, as is the case in the current study coals (Fig. 4.10b and c). Conversely, Dai et al. (2012) suggested that calcite rarely occurs as detrital grains, as it easily decomposes in the acidic environment of peat mire. As reported earlier, some samples from Moiyabana coal samples showed the presence of detrital fragments as shown in Figure 4.10a. The above observation suggests that detrital calcite particles in these coals are closely related to the kaolinite matrix, in which detrital calcite, quartz, occasionally, siderite are dispersed under freshwater (normal pH) environment which favoured the preservation of the detrital calcite particles after deposition. Some samples (E, F, G and H) show very high levels of carbonates.

Pyrite is the most abundant sulphide mineral in many coal samples (Dai et al., 2002; Kolker, 2012; Ward, 2002). It occurs in variable morphologies and is of different generations in the study area as evidenced by the multiple forms of pyrite in the Moiyabana coal samples. Of the observed pyrite forms, most occur as framboidal and epigenetic vein fillings (Fig. 4.11). Syngenetic pyrite is present as individual framboids disseminated throughout the vitrinite (Fig. 4.11b & d) in the study area coal seams, and clearly represents sulphide mineralization during or very shortly after peat accumulation (Ward, 2002). The epigenetic sulphide (Fig. 4.11a) occurrences observed may represent hydrothermal fluid input after coal formation (Thomas, 2013) and this form of pyrite is easily removed through beneficiation processes because it is easily dislodged from the coal surface. Although occurring in fewer amounts than the other two forms, euhedral to

subhedral pyrite occurs in veins or fills the pores and cavities in organic matter (Fig. 4.11c). There is also a noticeable trend of increasing pyrite to sample F, then almost no pyrite in samples H and G. This could possibly suggest a marine influence, but more facts are needed to substantiate this statement.

5.1.3 Peat-Forming Condition

5.1.3.1 Petrographic facies

The peat-forming conditions that prevailed during the formation of Moiyabana coal seams can be inferred based on the inherent coal constituents. The inference is based on the petrographic facies GI and TPI model by Diessel (1986) and mineral matter association with macerals in coals (Singh and Singh, 1996) as discussed in the literature review in subsection 2.3.2.5.1. The values of GI and TPI calculated from formula provided in subsection 2.3.2.5.1 are presented in Table 5.1.

Table 5.1: Values of GI and TPI ratios of Moiyabana coal samples.

	A	B	C	D	E	F	G	H
GI	0.17	0.12	0.09	0.07	0.09	0.13	0.46	0.23
TPI	0.69	0.71	0.49	0.37	0.37	0.42	1.40	0.63

Based on the GI-TPI plot (Fig. 5.2), Moiyabana coal samples are indicative of mainly low tree density except sample G, which shows increased tree density. It is also observed that most Morupule Formation coals were formed under open marsh conditions, except G which formed under dry forest swamp as evident in Figure 5.2. This observation suggests a different condition prevailed during the formation of sample G coal, possibly attributed to the differing water table level due to fluctuation of water influx to the depositional basin. The petrographic/chemical analysis also shows that sample G is different in terms of its mineralogical constituents.

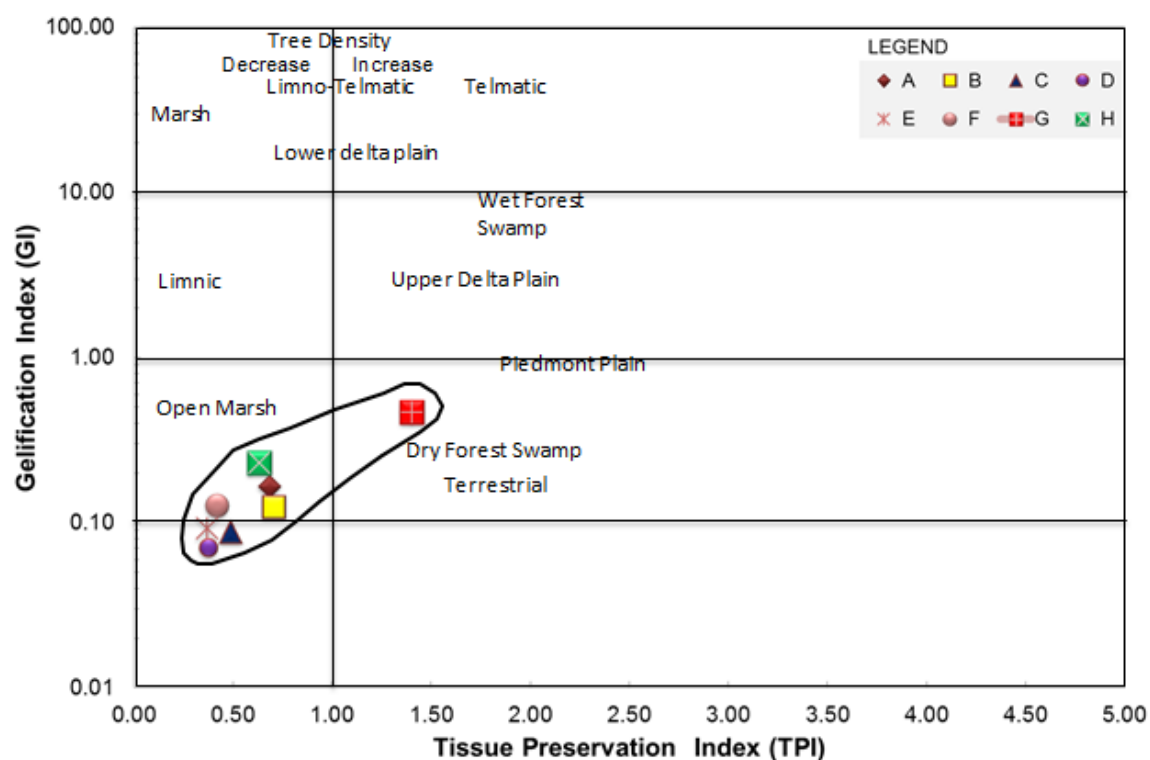


Figure 5.2: Coal facies diagram of TPI versus GI showing the peat-forming conditions of the Moiyabana coal samples.

Diessel (1986) concluded that wet conditions of peat formation are characterised by high GI and telo-vitrinite based high TPI, whereas dry conditions are characterized by low GI and telo-inertinite based low TPI. By using Figure 5.2 and Table 5.1, it is evident that Moiyabana coals were formed under dry condition in the peat formation stage as shown by low GI and telo-inertinite low TPI. Diessel (1986) went on to suggest that TPI (telo-vitrinite based TPI) values of 2.5 and more are indicative of the formation of coal seams under the influence of a lateral increase in subsidence, which is not the case with Moiyabana coal seams as all TPI values are less than 2.5.

Singh and Singh (1996) established that coals with high concentrations of inertodetrinite in association with mineral matter to be of allochthonous origin, which agrees with petrographic data of Moiyabana coals as shown in Table 4.2. This finding by Singh and Singh (1996), is supported by Carney et al. (1994) who concluded that most coal deposits in eastern Botswana are highly fragmented, suggesting fluvial transport prior to its deposition under anaerobic conditions, and only a few autochthonous coal seams have been identified. In agreement is also Falcon (1986b), whilst conducting study in the Transvaal Coalfield, reported that

coals with vitrinite contents as low as 5 - 10% (more typically between 10 and 40%) and high mineral matter contents (30 to 40%) citing No. 4 seam as example, are typical for the detrital or allochthonous coals which is true for Moiyabana coal samples (Table 4.4). Consequently, Moiyabana coal seams based on the above findings are inferred to have formed under open marsh with fluvially-dominated transportation and peat swamps that subsequently were buried following channel changes and basin subsidence.

5.1.3.2 Mineral matter concentration

As discussed earlier, the studied coal samples are rich in mineral matter content, hence the relevance of the model proposed by Singh and Singh (1996) in this project. According to this model, middle to top coal seams (F1b, F1c/d and F2b) represented by sample C to H (Fig. 4.1) were deposited under oxic (dry) moor with sudden high flooding whereas the bottom coal seam (F1a) represented by sample A and B (Fig. 4.1) were formed under alternating oxic and anoxic moor (Fig. 5.3).

This model has the shortcoming as it does not take into consideration the epigenetic minerals. The high carbonate contents observed for these coals are most likely epigenetic (e.g. cleats and veins), thus not related to the depositional environment.

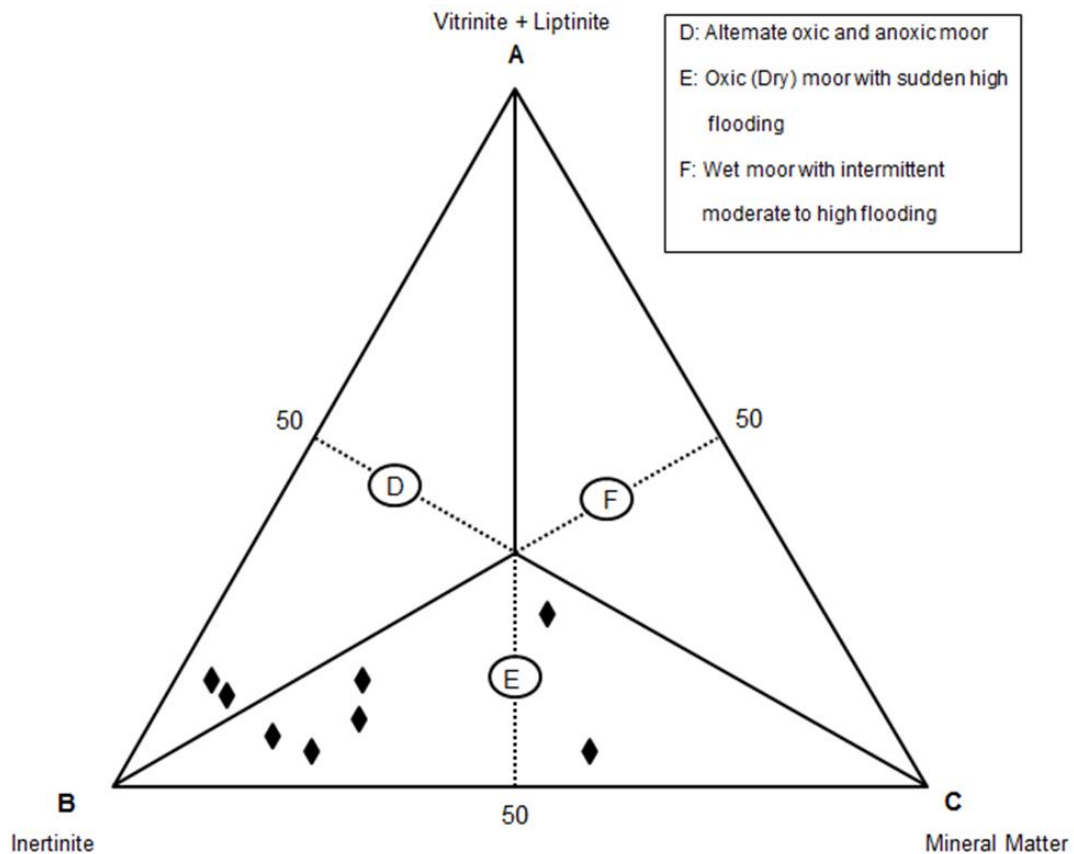


Figure 5.3: Ternary diagram illustrating depositional conditions based on the macerals and mineral matter content (after Singh and Singh, 1996).

5.2 Coal quality

Coal quality of the study area has been evaluated through proximate, ultimate and calorific value components, and compared to the petrographic findings.

5.2.1 Proximate analysis

The Moiyabana coal samples are characterised by moderately low volatile matter contents. Carney et al. (1994) associate volatile matter content in Botswana coals with change in macerals composition and rank. Carney outlined that Botswana coals with high content of volatile matter have low inertinite content and can be compared to the European coals, whereas coals with low volatile matter contents have high inertinite content. Carney et al. (1994) attribute the volatile matter variation to the palaeoenvironmental development from cold temperature climate with tundra peat swamps in a fluvio-deltaic sedimentary sequences (Smith, 1984),

to a warmer climate with the accumulation of more woody organic matter and less argillaceous material, under anaerobic conditions. On the other hand, Moosavirad et al. (2012) contend that the slight variation in the volatile matter contents is probably due to the compounds released from organic and mineral matter in coals. The conditions mentioned above are similar to the Carney argument, hence it can be inferred that volatile matter content in the study area are partly due to conditions that prevailed during Moiyabana coal formation. The low volatile matter content of Moiyabana coal samples, suggest to some degree, difficult to ignite coals.

Another very important component of the proximate analysis is ash content. Ash content plays a vital role in the overall quality and utilisation of coal, as it forms the inorganic fraction of coal which does not burn during coal combustion. By use of the Chinese coal quality classification standards (GB/T15224.1-2004, 2004), it categorises coal ash yields as follows; low, medium, and high ash coals denoted by <16%, 16–29%, and >29% ash yields, respectively. It categorises Moiyabana coal samples predominantly under high ash, except in sample A which falls in the medium category. The South African coal classification standard (SANS 11760-2007) also classifies Moiyabana coal samples similar to the Chinese standard (Table 2.7). Adopting the same classification standards for classification of Moiyabana coals using the ash yield parameter, it is evident that Moiyabana coal samples are predominantly containing high ash, except sample A which is in the medium ash category. The high ash content can be attributed to relatively slow burial of vegetative matter which is typical of Gondwana coal (Thomas, 2013). Insofar as coal trading is concerned, high ash content has detrimental effects on the overall commercial grade of coal (South African Classification, 2009) and also directly affects the heating value of coal (Fig. 4.19). Consequently, the Moiyabana coals need to be beneficiated before utilisation in power stations due to their high impurity content.

5.2.2 Ultimate analysis

Chou (1990) classified coal into three types based on the sulphur content: low (<1 %), medium (1– 3 %), and high (>3%) total sulphur. Based on this classification,

the Moiyabana coals may be classified as low to medium sulphur coals. It is imperative to note that the abundance of sulphur in coals is related to the depositional environment of coal seams (Chou, 1997; Liu et al., 2007; Zheng et al., 2008). Sulphur is thought to originate within the precursor peat during coal formation (Chou, 1990), or via marine transgressions. It is known that the sulphur content of marine water, where sulphur bacteria had a special role, is much higher than that of freshwater. Chou (1997) further suggests that the peats which are formed under marine influenced condition possess higher sulphur contents. In the absence of evidence to indicate any marine influence in the study area, the presence and the amount of sulphur in the Moiyabana coals is taken to reflect possible freshwater conditions during peat deposition. Although the increase in pyrite through the sequence is evident, possibly suggesting a marine transgression, more factual evidence is needed to substantiate this idea.

As stated earlier, oxygen, carbon and hydrogen can collectively be used to classify coal based on H/C and O/C atomic ratios. The Moiyabana coals have H/C and O/C atomic ratios ranging from 0.05 to 0.08 and 0.14 to 0.41 respectively. The H/C and O/C atomic ratios determined for the Moiyabana coal samples are indicative of the humic nature of the coals and are in agreement with the nature and origin of these coals.

Nitrogen data in coal analysis are mainly used for comparison purposes. With regard to coal quality, coals which are low in nitrogen are preferred by the industry due to the release of flue gases which pollutes the atmosphere. Speight (2005) advises that coal should not, as a rule have nitrogen contents of more than 1.5-2.0% (d.a.f) because of these NO_x emissions. Moiyabana coal samples have nitrogen content ranging from 0.35 to 1.19% (d.a.f) (Table 4.6), falling within Speight's suggested acceptable limit.

5.2.3 Calorific Value (CV)

The South African Classification (2009) used in domestic commercial trade of coal, grade coal based mainly on the ash and calorific value content. In the study area coal seams, higher grade coals are characterised by low ash content and

higher calorific values, whereas low grade coals have high ash and low calorific values as depicted in Figure 4.19. The basal seam (F1a) in Moiyabana and other coalfields in the SE belt of the Central Kalahari Sub-basin have been correlated by Smith (1984). Cairncross (2001) notes a CV value of 24.4 MJ/kg for Morupule Coalfield basal seam (Morupule Main), whereas in the current study the two samples (A and B) analysed from the basal seam have calorific values of 22.50 MJ/kg and 19.22 MJ/kg respectively. Although having a CV less than that of Morupule Coalfield, Moiyabana basal seam CV is close to the acceptable value and can be upgraded during beneficiation to meet the commercial coal grading.

5.3 Geochemistry

The Moiyabana coal seams and the rest of the south-eastern belt in the Central Kalahari sub-basin contain very high amounts of ash. Cairncross (2001) reports ash content of 21.1% for the Morupule Main seam in the Kgaswe Coalfield, whereas the Morupule Main seam equivalent in Moiyabana area recorded between 24.5 to 30.2% under the current study. Segwabe (2008) notice a slightly higher ash content value of 25.4% for Morupule Main seam than that provided by Cairncross. All other seams analysed in the present study have significantly higher ash contents.

5.3.1 Major elements

Elements exhibiting positive correlation with the ash yield suggest that these elements are occurring in the inorganic fraction of coal whereas elements with positive correlation with the organic carbon contents indicate their organic association in coal (Baïoumy, 2009). The indirect method used by Nicholls (1968) and later by Shao et al. (2004), was used in this study to determine the organic/inorganic affinity of elements in coal. The major elements in coal generally occur in mineral matter (Liu et al., 2001) rather than in organic matter (Pike et al., 1989). Therefore, major elements analyses can be used as a tool for discriminating element–mineral associations (Moosavirad et al., 2012).

The quartz, clay minerals (mainly kaolinite) and pyrite are the carriers of the dominant major elements of SiO_2 , Al_2O_3 and Fe_2O_3 respectively (Zhao et al., 2013). The concentrations of SiO_2 and ash in this study have almost the same vertical variation trend. The above statement is further supported by the strong positive correlation coefficient ($r=0.78$) between SiO_2 and ash content which could possibly suggest that SiO_2 is the controlling factor for the ash proportion. The generally high content of both ash and SiO_2 in the upper and lower seams to lowest in the middle seams is reported by Liu et al. (2001) to be attributed to two possible processes. In the first processes, they contend that an abrupt change of environmental conditions (e.g. pH, Eh) favoured the precipitation of silica in the swamp during that period of time. This abrupt change in condition could possibly have been brought about by the short time high water influx and long dry periods during deposition of coal seams as evident from Singh and Singh (1996) model. Secondly, they opined that some unusual process (e.g. flooding or subsidence) occurred to let the swamp gain access to inorganic input.

The abundance of Al_2O_3 has a similar trend to that of silica (Fig. 4.20) but has no inter-elemental relationship with Si oxide as established in the correlation coefficient of $r= -0.21$ (Table 4.9) between them, suggesting they do not exist together in mineral form for the study area coal seams. Positive correlation observed between elements Si, Ti and slightly weak positive correlation of Al_2O_3 with ash content suggests that these elements mode of occurrence is inorganic.

The negative correlation observed between Fe and ash content (Table 4.9) suggests an organic mode of occurrence of this element. The vertical distribution trend for Fe_2O_3 and SO_3 is the same in Moiyabana coal samples (Fig. 4.21). The strong positive correlation coefficient between Fe_2O_3 and SO_3 ($r=0.84$) suggest occurrence in the same mineral e.g. possibly in the finely disseminated pyrite/marcasite in macerals. According to Baioumy (2009), the correlation between oxides of Fe and total S confirms that iron sulphides (mainly pyrite) control the Fe_2O_3 content in the coal samples. The above evidence confirms that Fe and total S oxides exist together in sulphides for the Moiyabana coals. Some exceptionally high content of Fe oxides suggests the presence of iron oxides

(hematite) and Fe-bearing clay minerals (Ward, 2002) finely dispersed in the organic matter in addition to the presence of variable types of pyrite occurrences.

Although not having the same vertical distribution trend as SiO_2 and ash, TiO_2 has a strong positive correlation with both SiO_2 ($r=0.80$) and ash ($r=0.62$) as shown in Table 4.9. According to Liu et al. (2001), this suggests the inorganic fraction for this element due to its positive correlation coefficient with ash, which in turn is attributed to rutile (TiO_2) as common inclusions in clay mineral (Liu et al., 2001). In sedimentary settings, rutile is known to form during the reconstitution processes in clays and shales, and can also occur as a common detrital mineral (Moosavirad et al., 2012). Rutile is present in concentrations close to 2% in most Moiyabana coal samples. Moosavirad et al. (2012) further suggests that this range of Ti oxide contents is high when compared with the usual Ti content in coal elsewhere (0.05–0.2 %). Based on Moosavirad et al. (2012) suggestion for Ti/Al ratios, range of 0.06 to 0.13 for the coals of the study area could be regarded as a moderate variation in the Ti/Al ratios which implies that the detrital material supplied to the site of deposition had moderate values of Ti/Al ratio, which in turn signifies weak association of both Ti and Al oxides in the coal seams, as evident by $r = -0.22$ (Table 4.9).

There exists a strong positive correlation between oxides of Ca and Mg ($r=0.99$) suggesting the occurrence of both in dolomite mineral. The CaO, MgO, MnO and P_2O_5 vertical distribution trend vary differently from ash and SiO_2 and the same oxides have negative correlation with both ash and SiO_2 . For the above to happen, Moosavirad et al. (2012) suggest an organic affinity of these oxides. The weak positive correlation between P_2O_5 and CaO ($r=0.24$) signify very weak association of both P and Ca oxides in apatite mineral of the Moiyabana coal samples.

The presence of Na and K in coal is mainly due to the feldspar group of minerals (Baïoumy, 2009) albite and orthoclase respectively as opposed to common Na/K-rich illite (Zhao et al., 2013). The negative correlation observed for both Na_2O and K_2O with ash content ($r = -0.31$ and $r = -0.20$ respectively) suggest an organic mode of occurrence for this elements. As for Na, the negative correlation with ash

and positive correlation between Na_2O and Cl ($r=0.43$) further suggests Na to additionally occur in pore water as reported by Moosavirad et al. (2012). Consequently, the distribution pattern of Na for Moiyabana coal samples is partly inferred to be controlled by water fluctuation during formation and deposition of the peat.

5.3.2 Trace elements

Trace elements determination in coals is becoming more important because of restrictions on the emission of air toxics and the disposal of waste ash after coal combustion. In many countries legislation has been introduced limiting the discharge of environmentally sensitive trace elements (Clarke and Sloss, 1992). This legislation necessitated every coal user to prioritise knowing the trace elements concentration in coal prior to mining, to avoid stringent penalties of failing to comply with environmental laws. Özbayoglu (2012), notes that the amount of trace elements emission from coal combustion is strongly dependent on the modes of occurrence in coal. Therefore, it is imperative to establish concentration and trace elements association with their source mineral matter in coals.

5.3.2.1 Trace elements concentration

The analysed data for trace elements (Table 4.10) of the study area coal samples when compared to Coal Clarke's values (Ketriss and Yudovich, 2009) show high enrichment in trace elements of Ba, Cr, Cu, F, Hg, Li, Mn, Pb, Sr, U, V and Zn (all samples higher than the Coal Clarke level), normal concentration for trace elements of Cd, Co, Ge, Mn, Ni and Th (near to the Clarke level) and impoverished in trace elements of As, B, Sb, Se and Cl (all samples depleted than the Coal Clarke level). Trace elements of major concern are generally within the Coal Clarke's limit (Ketriss and Yudovich, 2009) except for Hg and Pb and the two radioactive elements, whereas all the trace elements of moderate concern are enriched except Ni and lastly, trace elements of minor concern are distributed across all the three concentration level.

In comparison to South African Highveld coals (Wagner and Hlatshwayo, 2005), trace elements concentration of Cr, Cu, Mn, Pb, V and Zn show enrichment and trace of As, Cd, Hg, Mo, Se, Ni, Co and Sb are normal/comparable to Moiyabana coal samples. Using data of average global values (Zhang et al., 2004) in comparison to Moiyabana coal samples, trace elements concentration levels of Cr, Cu, Mn, V and Zn are enriched whereas trace elements concentration values of As, Cd, Hg, Mo, Pb, Se, Ni, Co and Sb are normal to depleted.

The trace elements grouping based on the elements affinity to sulphur, iron and silicon are grouped into sulfophile, siderophile, and lithophile elements respectively (Babu, 1975; Goldschmidt, 1954; Wagner and Hlatshwayo, 2005). In explanation of the above groups, Goldschmidt (1954) described siderophile elements as most likely occurring in their native state, sulfophile elements combine with sulphur as sulphides, whereas lithophile elements occur primarily as rock forming minerals. Although different geological processes are attributed to enrichment/depletion of trace elements in coals, the enrichment of most sulfophile elements (Cu, Zn, Hg and Pb) is reported by Ketris and Yudovich (2009) to have been caused by hydrothermal input related to coal metamorphism. As for most Botswana coals, these fluids are reported by Carney et al. (1994) to have been heated by dolerite sills/dykes when they intruded the host rock during the formation of coal seams. The Stromberg lava basalt which was emplaced after the formation of coal (Smith, 1984) could also have played a part in increasing the temperature of the surrounding fluids. However, enrichment in litho- and siderophile elements such as (Ba, Cr, Mn, F, Li and Sr) for Moiyabana coals may partly be due to hydrothermal input, but primary coal-forming flora could have acted here as an actual factor of difference (Yudovich, 1978).

The enriched trace elements with significant importance insofar as the environmental and health aspect is concerned are further discussed individually as follows:

Mercury (Hg): During the early work carried out on coal regarding Hg concentration in coal, Dvornikov (1981) found out that mercury is present in coal

in three forms, i.e. mercury sulphide (HgS), metallic mercury, and organic mercury. In review of most technical works dealing with Hg in coal, Swaine (1990) concluded that these forms of mercury are mainly found in Russian coals near areas of Hg mineralization. Swaine (1990) went on to suggest that Hg is mainly associated with pyrite for most coals which is also the case with Moiyabana coal seams as evidenced by the relationship between Hg and sulphide minerals (Fig. 5.4). Work by Finkelman (1994) also supported earlier findings by Swaine that mercury is found in solid solution with pyrite. He further alluded that epigenetic pyrite probably has more Hg than syngenetic pyrite. In the current study, most pyrite occurs mainly as epigenetic which support the above finding by both Swaine and Finkelman. Based on the above facts, it is safe to assume that Hg occurs in sulphides (mainly pyrite) which is supported by positive correlation coefficient with ash ($r=0.17$) which in turn suggest inorganic mode of occurrence of this element.

Lead (Pb): Finkelman (1994) states that Pb occurs mainly as sulphides or associated with sulphide minerals, with galena being the most common form of lead in coal. Swaine (1990) also suggests that an organic association may be possible, most likely in the lower rank coals. In the current study, Pb is clearly associated with galena as evidenced by the similar vertical distribution trend between sulphur/sulphide and Pb (Fig. 5.4). The positive correlation coefficient between Pb and ash ($r=0.33$) further suggest Pb occurs in an inorganic fraction of coal. Therefore, this element can be easily removed with the heavy fraction during coal beneficiation.

Uranium (U): High amounts of trace elements in solutions of the percolating water in the peat bog are requisite to the enrichment of trace elements in coal (Ketriss and Yudovich, 2002). The coal-forming bog conditions influenced by the percolating water in the coal bed determine the solubility of minerals present in the peat allowing for a possible ion exchangeability based on the pH and Eh of water. In the study area, U distribution moderately correlates with MnO and weakly correlates with MgO (Fig. 5.5), which imply U enrichment due to co-precipitation of U with Mg- and Mn-hydroxides under oxidising conditions, as suggested in Schönwiese (2007). The above contention is supported by the

positive correlation of ash with U ($r=0.37$), suggesting an inorganic mode of occurrence. The other possibility for the enrichment of U could be influenced by volcanic ash (tonsteins) occurring possibly in zircon above the coal seams (Dai et al., 2011a). The fluid movements up and down the coal horizon would allow for the exchangeable ions to be leached from the tonstein layers above to their ultimate fixation in coal below, in that elevating the U content in coal seams.

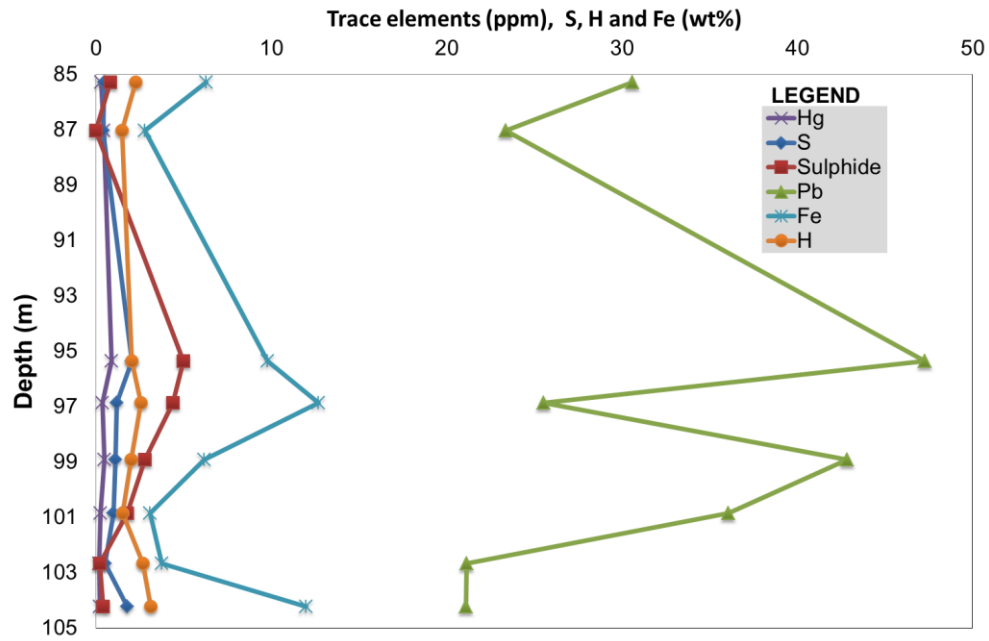


Figure 5.4: Vertical relationship between trace elements.

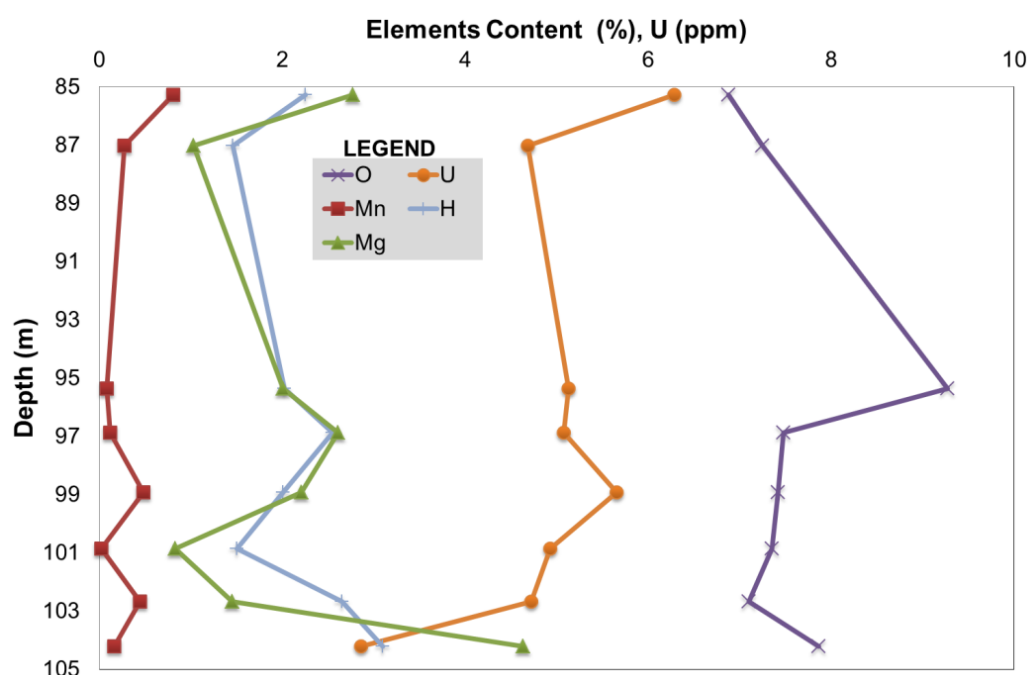


Figure 5.5: Vertical relationship between elements.

5.3.2.2 Trace Elements Association with Mineral Matter in Coal

Every trace element in coal is associated to some degree with minerals found in coal. With the previous section successfully discussed trace elements of major concern with higher concentration than the Coal Clarke levels in Moiyabana coal seams, it would be inappropriate not to discuss trace elements relationship with mineral matter in coal. Only trace elements of significant importance will be discussed in this section to aid in beneficiation processes prior to mining and utilisation of coal. Selected elements to be discussed include:

Antimony (Sb): Although occurring in small amount, Sb is classified by the US National Research Council (NRC) as one of the hazardous air pollutants (CTB, 2011). Finkelman (1994) suggested that there is no direct evidence to support any particular mode of occurrence of Sb in coal. Under the same research he however, indicated that Sb may be found in solid solution in pyrite or as minute accessory sulphides dispersed through the organic matter. Conversely, Gluskoter et al. (1977) indicated that Sb is organically associated with coal. Swaine (1990) supports earlier findings by Gluskoter et al. (1977) by asserting that Sb is apparently organically bound in the coal.

In Moiyabana coal samples, Sb is high in concentration where sulphur content is high (all sample F) and low where sulphur content is low (all sample B). This observation could be interpreted to indicate S and Sb occur in soluble sulphides (stibnite) in Moiyabana coal samples. Finkelman (1994) also concluded that Sb may be associated to some degree to sulphide minerals but noted only pyrite mineral in solid solution. In agreement is also Özbayoglu (2012) who categorised Sb under the classification of US NRC for trace elements based on their distribution in coal with iron sulphide (pyrite), clearly associating Sb with inorganic affinity. This argument can be further supported by the positive correlation of Sb with ash content ($r=0.37$), which also is in agreement with the idea that Sb is associated to the inorganic fraction (pyrite, stibnite and silicates mainly finely dispersed clays in macerals).

Arsenic (As): There is direct evidence and a substantial amount of indirect evidence for the association of As mainly with pyrite and to a lesser degree with arsenopyrite in coal (Finkelman, 1994). Arsenic is mainly associated with the inorganic fraction in coal, with small amounts that may be organically associated (Swaine, 1990). Finkelman states that As is associated with the pyrite for most U.S. coals. Gluskoter et al. (1977) state that As showed an inorganic association for all of the samples that they studied. It is interesting to note that As is one of the few elements that are found in coal at a higher concentration than it is found in the earth's crust (Gluskoter et al., 1977). In the current study, a weak positive correlation ($r=0.2$) exist between As and ash content indicating inorganic fraction (pyrite, clays and carbonates) of this element. The weak correlation of As with ash content also suggests possibly that some As could be associated with organic matter.

Cadmium (Cd): Many authors associate Cd predominantly with sulphide minerals in coal, particularly sphalerite (Swaine, 1990; Finkelman, 1994). Gluskoter et al. (1977) report data that supports the conclusion that Cd is strongly inorganically associated in coal. In Moiyabana coal seams, Cd is clearly associated with the inorganic fraction as evident by strong positive correlation with ash content ($r=0.81$). Although Cd has no known biological function, it is known to accumulate

in the kidneys and liver. It has been established also that Cd is carcinogenic and when present in excess amounts can cause emphysema and fibrosis of the lungs.

Chromium (Cr): Finkelman (1994) assumes Cr to either be associated with the organic matter in coal or with the clays in coal. Swaine (1990) conclude that Cr mode of occurrence in coal is not clear. Rimmer (1991) found strong indications that Cr is associated with the clays. Conversely, Mukherjee et al. (1988) found out that Cr was predominantly associated with organic matter. Gluskoter et al (1977) classifies Cr as an intermediate element, which means it is neither strongly associated with the organic or inorganic material. In Moiyabana coals, Cr correlates positively with the ash content ($r=0.59$) suggesting an inorganic mode of occurrence. This finding is in agreement with Finkelman and Rimmer that Cr is associated with clay. The slightly higher values of Cr in Moiyabana coals are attributed to high amount of clay observed in these coal samples. It is also possible that Cr is added to the coals during crushing and milling due to the metal used in the equipment.

Cobalt (Co): The investigation carried out on Co to date, suggests that Co is associated with the sulphide minerals (pyrite/marcasite) present in coal. According to Swaine (1990), Co is associated with the mineral matter in coal. Finkelman (1994) asserted that Co shows some degree of association with the organic matter in coal and the clays present in coal. Data analysed by Mukherjee et al. (1988) is in agreement with Finkelman. They found that small amounts of Co may occur as organic chelates. In Moiyabana coal samples, Co is associated with an organic fraction as evidenced by negative correlation coefficient with ash content ($r=-0.68$). The small amount of Co present in the current study could be inferred to occur as organic chelates, which is in support of Mukherjee et al. (1998) findings.

5.3.2.3 Trace elements as indicators of peat-forming facies

The boron concentration will be used as an indicator of paleosalinity and manganese as an indicator for peat forming conditions during formation of Moiyabana coal seams.

Boron (B): Many researchers have used B as an indicator of paleosalinity. Curtis (1964) and Eagar and Spears (1966) both examined the same coal measures and came up with two contradicting findings. Curtis found that B can be used as a paleosalinity indicator, whereas Eagar and Spears found little correlation with B and other paleosalinity indicators. Earlier studies by Swaine (1971) and later by Goodarzi and Swaine (1994) successfully used total B content to develop fresh/brackish and brackish/marine indices. Goodarzi and Swaine (1994), determined the B contents (in ppm) related to specified coal-forming environments: Less than 50—freshwater, 50–110—mildly brackish, and more than 110—brackish.

In the Moiyabana coal, the B concentration ranges between 14.57 to 43.00 ppm (db) with an average value of 30.04 ppm (db), indicating a freshwater indice. There is a strong positive correlation ($r=0.84$) between B and ash content, suggesting that B has a detrital origin and associated with the inorganic fraction (tourmaline) in coal. By using the B content to infer depositional environment which prevailed during coal formation, the low B content is attributed to a freshwater environment during the time of deposition for coals of Moiyabana Coalfield.

Manganese (Mn): Ketris and Yudovich (2002) suggest that Mn can reveal valuable information about paleo-peat bog types based on the Mn content, since the dispersion of the Mn contents in coal, caused by variable contribution of the Mn (organic form), reflects different facies in the peat-formation area. Ketris and Yudovich (2009) report that Mn contents in peat depend strongly on peat pH and Eh and they demonstrated that in oligotrophic peat (which may be the precursor for low-ash coal), the conditions for the bonding of Mn by the peat did not exist, and opposite, intensive leaching of the primary vegetative Mn occurred. They further suggest that if large inputs of the terrigenous particles to running-water peat bogs, an alkalization of peat water occurred, and conditions for Mn (organic) formation were created. However, they determined that subsequently, diagenetic processes of carbonate and pyrite formation could bring about a destruction of the weak-bonding Mn (organic) compounds, and to the redistribution of Mn in carbonatic (siderite and calcite) and sulfidic (pyrite) forms.

In the study area coal seams, Mn vertical distribution trend varied with depth (Fig. 4.21). This variation could possibly imply different facies for the peat-formation of Moiyabana coals. It has been established earlier in this report through the use of Pearson's correlation coefficient that Mn negatively correlates with ash ($r = -0.10$), implying an organic affinity. Taking into account the higher Mn concentration than the Coal Clarke level in these coal samples, there exists a possibility that large inputs of terrigenous particles rich in Mn in the running water peat bog could have caused the alkalization of peat water, making a perfect condition suitable for the formation of Mn (organic) as suggested by Ketris and Yudovich (2009). The correlation of the triad Mn-Fe-Ca in coals is noted by Ketris and Yudovich (2002) to indicate the presence of these elements together in phosphate minerals. Conversely, in the study area coal seams, these two elements do not correlate with Mn, signifying a different mineral occurrence of these elements.

5.4 Summary

The macroscopic results reveal that Moiyabana coal samples can be classified broadly as humic/banded coals based on the bright and dull lithotypes. The microscopic assessment of these coals resembles an inertinite coalification path. The pre-dominance of inertinite maceral group is attributed to the prevalence of oxidizing conditions for a prolonged period of time, when accumulated vegetal matter was exposed to the surface. According to the mean random reflectance (Rom) values which range from 0.84 to 1.04%, the study area coals samples can be classified as bituminous medium rank C, borderline B coals, which comparatively are high for Botswana coals. The increased rank values are ascribed to the surrounding dolerite dyke intrusions present in the vicinity of the coalfield, as reported by Carney et al. (1994). The inorganic content of these coals is dominated by the silicate fraction which consists mainly of about 60-80% clay minerals. Peat forming conditions based on the model by Diesel (1986) using the petrographic facies diagram reveals mainly open marsh conditions with low tree density. A model by Singh and Singh (1996), based on earlier work by Goodarzi (1985), used the proportions of both macerals and mineral matter to understand

the depositional environment. According to this model, top coal seams were deposited under oxic (dry) moor with sudden high flooding whereas the bottom coal seams (basal) were forming under alternate oxic and anoxic moor. It has been established that, as the rank increases, the volatile matter decreases hence lower volatile matter for these coals makes it difficult to ignite. The high mineral matter content of these coals can be attributed to relatively slow burial of vegetative matter which is typical of Karoo hosted coals. The sulphur contents of Moiyabana coal samples categorises these coals as low to medium sulphur coals suggesting possible freshwater condition during peat deposition. The increase of pyrite through the sequences could reflect a possible marine transgression into the deposit. The calorific values of samples in the basal seam (F1a) are comparable to most equivalent seams in Botswana. The strong positive correlation coefficient ($r=0.78$) between SiO_2 and ash content possibly suggests that SiO_2 is the controlling factor for the ash proportion. The generally high content of both ash and SiO_2 in coals is reported by Liu et al. (2001) to be attributed to two possible processes. In the first processes, they contend that an abrupt change of environmental conditions (e.g. pH, Eh) favoured the precipitation of silica in the swamp during that period of time. Secondly, they opined that some unusual process (e.g. flooding or subsidence) occurred to let the swamp gain access to inorganic input. The strong positive correlation coefficient between Fe_2O_3 and SO_3 ($r=0.84$) suggest the occurrence in the same mineral (pyrite/marcasite). The analysed data indicate that most trace elements of major concern are within global acceptable limits, except Hg, Pb, U and Th. The enriched trace elements are thought to have been caused by hydrothermal input related to coal metamorphism. The boron concentration level indicates a freshwater indice for these coals whereas strong positive correlation ($r=0.84$) between B and ash content suggesting that B has a detrital origin. In the study area coal samples, Mn variation with depth possibly implies different facies for the peat-formation of Moiyabana coal seams.

6 CONCLUSION

The petrographic, chemical and geochemical analyses were performed on coal samples from a single borehole core including different seams of the Morupule Formation from the Moiyabana Coalfield located in the south-eastern Kalahari sub-basin, Botswana. Macerals, coal rank, coal quality, and major and trace elements concentrations, as well as the type of peat-forming conditions using critical petrographic facies and mineral concentration were studied. The following conclusions are made:

- Petrographic investigations indicate that the Moiyabana coal seams are inertinite rich and depleted in terrestrially derived organic debris (vitrinite and liptinite). The predominance of inertodetrinite in the inertinite group indicates fluvial transportation before / during / immediately after peat mire accumulation.
- According to the mean vitrinite reflectance values (0.84-1.04 RoVmr%) from the studied coals, they are classified as medium C rank bituminous coal, borderline medium B, based on the ECE-UN in seam coal classification system.
- Petrographic identifiable minerals include: silicates (clay and quartz) dominate in the inorganic fraction, and carbonates (dominated by calcite), sulphides (mainly pyrite) and sulphates are less abundant.
- The H/C and O/C atomic ratios categorise these coal samples as humic coal.
- Petrographic facies models employed in this study suggest the Moiyabana coal seams were deposited under freshwater environment, oxic (dry) moor with sudden high flooding to alternating oxic/anoxic moor depositional conditions and peat-forming condition mainly under open marsh.

- The chemical component of these coal seams reveals that Moiyabana coals are characterised by high ash, low moisture and low volatile matter. The high ash content of these coals is attributed to the relatively slow burial of vegetative matter.
- The assessed samples indicate a low to moderate heating value, suggesting that these coals are suitable for electrical power generation.
- The Moiyabana coals generally are comparable in quality with the extensively developed coal deposits in the eastern flank of Botswana. The moderately low sulphur content indicates that the coals were formed in a freshwater environment.
- The geochemical component of Moiyabana coals is characterized by high contents of Si, Al, Ti and Fe reflecting high content of silicate minerals. The low content of Ca, Mg, Mn and P values suggests low carbonate and phosphate contents. Na occurs mainly in pore water space than as a solid-bounded phase.
- Elements As, Al, B, Ba, Cd, Cu, Cr, F, Ge, Hg, Li, Mo, Ni, Pb, Sb, Si, Ti, U, V and Zr exhibit positive correlation with ash yield, suggesting these elements are derived from inorganic materials. All the remaining elements show varying negative correlation with ash yield, suggesting an organic affinity of these elements. The boron concentration suggests freshwater environments for these coal seams
- The environmentally hazardous trace elements (As, B, Cd, Mo and Se) are mostly under acceptable limits, except a few (e.g. Pb) which are associated with the inorganic fraction. Therefore, they can be removed with the heavy fraction of the coal seams during grinding and crushing process before coal combustion.

- Based on the overall trace elements content of these coal seams, they should not hinder the utilisation of this coal.

7 Recommendations

In future the following researches are recommended:

- Lateral distribution of coal seams using more than one borehole core is needed to improve the assessment of the Coalfield.
- The characterisation of all the seams including the non-coal units such as roof and floor and the partings between the coal seams in the study area need to be considered to aid in the trace elements mode of occurrence.
- The identification of the geochemical controls which govern the distribution of inorganic (mineral matter) in the coal will help going forward insofar as the trace elements movement is concerned.
- An assessment of the dolerite intrusion in the area to explain the higher than expected reflectance values.
- Beneficiation and utilisation tests on the coals to predict performance.

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