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A DISSERTATION SUBMITTED IN FULFILMENT OF THE REQUIREMENTS FOR THE DEGREE OF MASTER OF TECHNOLOGY IN EXTRACTION METALLURGY

Correlation of spontaneous combustion liability indices and basis for coal analysis for reported Witbank coal, South Africa

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DECLARATION

I hereby declare that this dissertation submitted in the partial fulfilment of the Degree for Master of Technology to “University of Johannesburg” is my own, unaided work. Where use has been made of the work of others, it has been duly acknowledged. It has not been submitted to any other University/institution for the award of any degree.

Signed:

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Abisola Risiwat Gbadamosi

This ________ day of ________________ 2019

ABSTRACT

The self-heating of coal due to oxidation potentially leading to its ignition is called “spontaneous combustion”. The characterisation of the physical and chemical properties of coal on a standard provides an understanding of its characteristics towards spontaneous combustion. The liability of coal to undergo spontaneous combustion for selected coal samples obtained from the Witbank Coalfields was examined using different spontaneous combustion tests. The simple indices [crossing point temperature (XPT) and Stage II Slope] obtained from differential thermal analysis and composite indices [FCC (Feng, Chakravorty, Cochrane) and Wits-Ehac Indices] were used to examine the liabilities of 30 coal samples. The test results are examined and presented according to the readily recognisable characteristics known to be indicators of self-heating liability. Two characteristics obtained from a differential thermogram (XPT and Stage II Slope) are evaluated as indicative of the liability of coal to self-heat. The simple indices provide inconsistent predictions of spontaneous combustion liability, while the composite liability indices provide more reliable results.

The coal properties (moisture, volatile matter, fixed carbon, ash, carbon, hydrogen, nitrogen, oxygen and sulphur contents) on different basis [% air dried (ad), % dry (db) and % dry ash free (daf) basis] were determined from the proximate and ultimate analyses in compliance with the American Society for Testing and Material (ASTM) standard. The results obtained from these coal properties were related to different liability indices used in this study to develop trends of linear relationships using regression analysis. This study indicated that the ad basis indicates a higher correlation coefficients than the db and daf basis for the XPT and FCC Index, while the daf shows a higher correlation coefficients than the ad and db basis for the Wits-Ehac Index. It was found that the trend of linear relationships of these coal properties differs from one liability index to the other. The XPT show a better trend followed by the Stage II Slope on the coal properties among the spontaneous combustion liability indices evaluated.

The results of the experimental tests were used to evaluate the characteristics known to be an indicator of the liability of coal to undergo spontaneous combustion and it was found that the coal properties influence self-heating and vary from one sample to the next.

LIST OF PUBLICATIONS

The following were originated from this work:

Publications (ISI/IBSS Journals)

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Published Conference Proceedings

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DEDICATION

To my late father, my beloved mother, husband and siblings who have always supported my academic life and journey.

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LIST OF SYMBOLS USED

% = Percentage

g = Gram

mm = Millimetre

^{o}C = Degree celsius

^{o}C/min = Degree per minute

μm = Micron meter

LIST OF SUBSCRIPTS USED

%ad = percentage air dry basis

%db = percentage dry basis

%daf = percentage dry ash free basis

W_1 = Initial weight

W_2 = Final weight

W_3 = weight of ash and crucible

LIST OF ACRONYMS USED

A% = Percentage of ash content

ANN = Artificial neural network

ASTM = American society for testing and materials

C% = Percentage of carbon content

CFD = Computational fluid dynamics

DSC = Differential scanning calorimetry

DTA = Differential thermal analysis

FC% = Percentage of fixed carbon content

FCC = Feng, Chakravorty, Cochrane

FT = Flammability temperature

IPT = Ignition point temperature

M% = Percentage of moisture (%)

MS Excel = Microsoft Excel

MSR = Mixture surface regression

N% = Percentage of nitrogen content

O% = Percentage of oxygen content

r = Correlation coefficient

S% = Percentage of total sulphur

t = Time (min)

TGA = Thermogravimetric analysis

PFC = Pulse flow calorimetry

Rmax = Mean reflectance

VM% = Percentage volatile matter content

WOE = Wet oxidation potential

XPT = Crossing point temperature

1 INTRODUCTION

1.1 Chapter overview

Coal is a combustible material comprising of various organic and inorganic constituents. When coal is subjected to the air, it undergoes self-heating which leads to spontaneous combustion (Banerjee, 2000; Liu & Zhou, 2010). During self-heating, reactions of coal and oxygen provide sufficient energy to aid combustion without the effect of an external heat source. Recent studies reported that the spontaneous combustion of coal-shale, highwalls, spoil heaps and waste dumps is similar to coal oxidation, which produces carbon monoxide and carbon dioxide (Onifade et al., 2019). The self-heating of coal with a possible alteration to endogenous fires is a threat in underground and open mines and has an unfavourable impact on the mine environment. Self-heating of coal is a well-known problem in coal mining operations, which commonly results in the loss of valuable and non-renewable resources, and has significant economic and environmental impacts (Liu & Zhou, 2010; Wessling et al., 2008). The occurrence involves a variety of complex physical and chemical processes, resulting from the interaction between coal and oxygen and the associated heat liberated to the surrounding areas (Wessling et al., 2008). If sufficient oxygen is available in the air but the energy generated is not dissipated sufficiently by conduction, convection or radiation, a further increase in temperature will promote the oxidation and self-heating of coal (Yuan & Smith, 2011).

Studies have indicated that the intrinsic (proximate, ultimate and petrographic properties) and extrinsic (geological, mining and atmospheric condition) factors impacting the advance of self-heating are the source of complexities in understanding the mechanism of coal spontaneous combustion (Onifade & Genc, 2019; Beamish & Blazak, 2005; Panigrahi & Sahu, 2004; Falcon, 1987). All the experimental techniques to estimate the liability of coal to undergo spontaneous combustion primarily depend on these factors. A number of different techniques such as the Wits-Ehac Index, Wits-CT Index, R70 tests, XPT, FCC Index, Flammability Temperature (FT), Differential Thermal Analysis (DTA), MR Index, Wet Oxidation Potential (WOE), Ignition Point Temperature (IPT), Russian U index, Stage II Slope, Olpinski Index, thermogravimetry analysis (TGA), etc. have been designed and used for years to estimate the liability of coal (Genc et al., 2018; Mohalik et al., 2008; Kucuk et al., 2003; Kaymakci & Didari, 2002; Beamish et al., 2001; Nugroho et al., 1998).

There have been many investigations to estimate the liability of coal to undergo spontaneous combustion both experimentally and computationally (Said et al., 2020; Stracher & Taylor, 2004; Ren et al., 1999), but limited studies have compared the results of different spontaneous combustion tests for the assessment of the liability of coal and proved their validities. Furthermore, limited study has been done to explain the characteristics of coal self-heating taking into account the coal recording basis such as air dry (ad), dry basis (db), and dry ash free (daf) basis.

In this study, the spontaneous combustion liability indices of thirty coal samples from the Witbank Coalfields were evaluated using four spontaneous combustion test methods (XPT, Stage II Slope, Wits-Ehac Index, and FCC Index). The results of the coal properties (moisture, ash, volatile matter, fixed carbon, total carbon, nitrogen, hydrogen, oxygen and sulphur contents) of these samples on % ad, % db and % daf basis were reported. The trends of linear relations between the coal properties and the liability indices were established using regression analyses. The outcome of this study would allow for a detailed understanding and interpretation of coal recording basis and their liability indices.

1.2 Background of the study

Spontaneous combustion is a known problem influencing the environment, valuable and non-renewable natural resources, the economy, life, property and human health (Onifade & Genc, 2019; Avila et al., 2014; Zhang et al., 2013). The concern is about the emission of large volumes of greenhouse gases, which due to global warming has become the main focus of study (Yuan & Smith, 2008). The impact is more immediate on the local or regional level with consideration to natural resources and human life (Pone et al., 2007; Stracher &Taylor, 2004).

The factors that control the liability of coal to undergo spontaneous combustion have been the subject of many investigations. The factors include and are not limited to pyrite, particle size, mineral matter, petrographic properties, volatile matter, sulphur, moisture, etc. (Sujanti & Zhang, 2001; Kaymckci & Didari, 2002; Singh & Demirbilek, 1987). The characteristics of coal has been related to the intrinsic properties (petrographic properties, oxygen, ash, moisture, volatile matter, sulphur, calorific value, pyrite, porosity and etc.) and extrinsic properties (particle size, environmental, mining methods and geological condition) (Chandralal et al., 2014; Arisoy & Akgun, 2000; Falcon, 2004). Many studies have reported the relations between liability indices and coal properties (Onifade & Genc, 2018; Beamish et al., 2001, 2000).

Banerjee et al. (1985) suggested a technique to assess the liability of coal using the XPT value. Although, the study indicated that the XPT is generally not reliable for coal with high moisture and recommended the assessment of such coal by using both the XPT and rate of temperature increase. Mahidin et al. (2002) indicated that the XPT decreased with a rise in the quantities of volatile matter, oxygen and moisture. Beamish & Blazak (2005) identified that the values of R70 (liability index) reduced with a rise in the quantity of mineral matter.

Researchers globally have examined the spontaneous combustion liability of coal by using several methods (Said et al., 2020 Riebeiro, 2016; Beamish et al., 2012; Mohalik et al., 2008; Ren et al., 1999). The crux of their work is the use of a laboratory experiment to measure the liability of different coal to self-heat. A number of experimental methods have been reviewed and a decision was made to carry out experimental tests using the XPT and DTA. Some characteristics of coal thermograms thought to be indicative of spontaneous combustion liability such as XPT, Stage II Slope and low transition temperatures from Stage II to Stage III referred as simple indices, are identified to be inconsistent in the assessment of the tendency of coal to spontaneously combust (Gouws et al., 1991; Wade et al., 1987). The concept of composite indices, comprising of two or more simple indices were evaluated and compared with the results obtained from the simple indices in order to overcome these difficulties.

Considering the values obtained from each of the liability indices, coal has been grouped based on their liability to undergo spontaneous combustion as high, moderate and low liability in most cases. However, out of these testing methods, no comparisons have been established from the results obtained from each technique to validate the liability of different coal to self-heat. In this work, an attempt was made to evaluate 30 coal samples from the Witbank Coalfields and to conduct laboratory tests to examine their liability to undergo spontaneous combustion by using established simple and composite indices. Furthermore, selected properties of theses samples which contribute to spontaneous combustion are determined. The results of the different spontaneous combustion tests are compared with each other and related to the selected coal properties. The results of the experimental tests are used to evaluate the characteristics known to be an indicator of the liability of coal to self-heat.

1.3 Description of the Study Area

The research area is in Witbank Coalfields, South Africa. The Witbank Coalfields was the center of coal mining in South Africa since mining commenced in the 1890s. In the past, many collieries in the Witbank area used room and pillar methods, typically with low coal recovery, leaving behind a significant amount of coal in the pillars, roofs and floors. The rate of airflow into the mine workings results into self-heating which later leads to spontaneous combustion. The impacts of self-heat and spontaneous combustion in the Middleburg colliery was documented by Bell et al. (2001). When the mining operations were suspended in 1947, the phenomenon of spontaneous combustion was first observed. Self-heating of coal was first discovered when old workings in the middle seam were subjected to atmospheric conditions (Bell et al., 2001). The need to extract water from the old operations arose due to large amounts of water beyond the normal limit, thereby resulting in extreme spontaneous combustion in the new seams. The mine faced serious difficulties in managing spontaneous combustion as a result of the problem of finding the old workings and recognising the bords when drilling. The mine also faced the issue of spontaneous combustion in highwalls, waste dumps, stockpiles, etc.

The Boschmanskrans pit in the Middleburg colliery also experienced spontaneous combustion in the highwalls, spoil heaps, stockpiles, and the No. 2 seam due to earlier underground operations. There was an inadequate sand problem in the mine to clad the highwalls undergoing self-heating. The high liability in the Arthur Taylor colliery for spontaneous combustion of the No. 3 seam and its quantity of high sulphur was the purpose why the coal seam was not always mined often for commercial purposes. Kleinkopje Colliery faced severe spontaneous combustion in coal seam, blast-holes and stockpiles once subjected to atmospheric conditions and when oxygen in the atmosphere enters into old workings from the points of contact between two ramps. The regular occurrence of self-heating in Landau colliery reduced in 2003 due to division between the two seams in the mine at its thickest. This is because the coal mining activities had to be carried out in two stages. There was a vital and long-term risk of coal self-heating in the mine and effective practices were implemented to mitigate the incidence. Figure 1.1 shows the various Coalfields in South Africa.

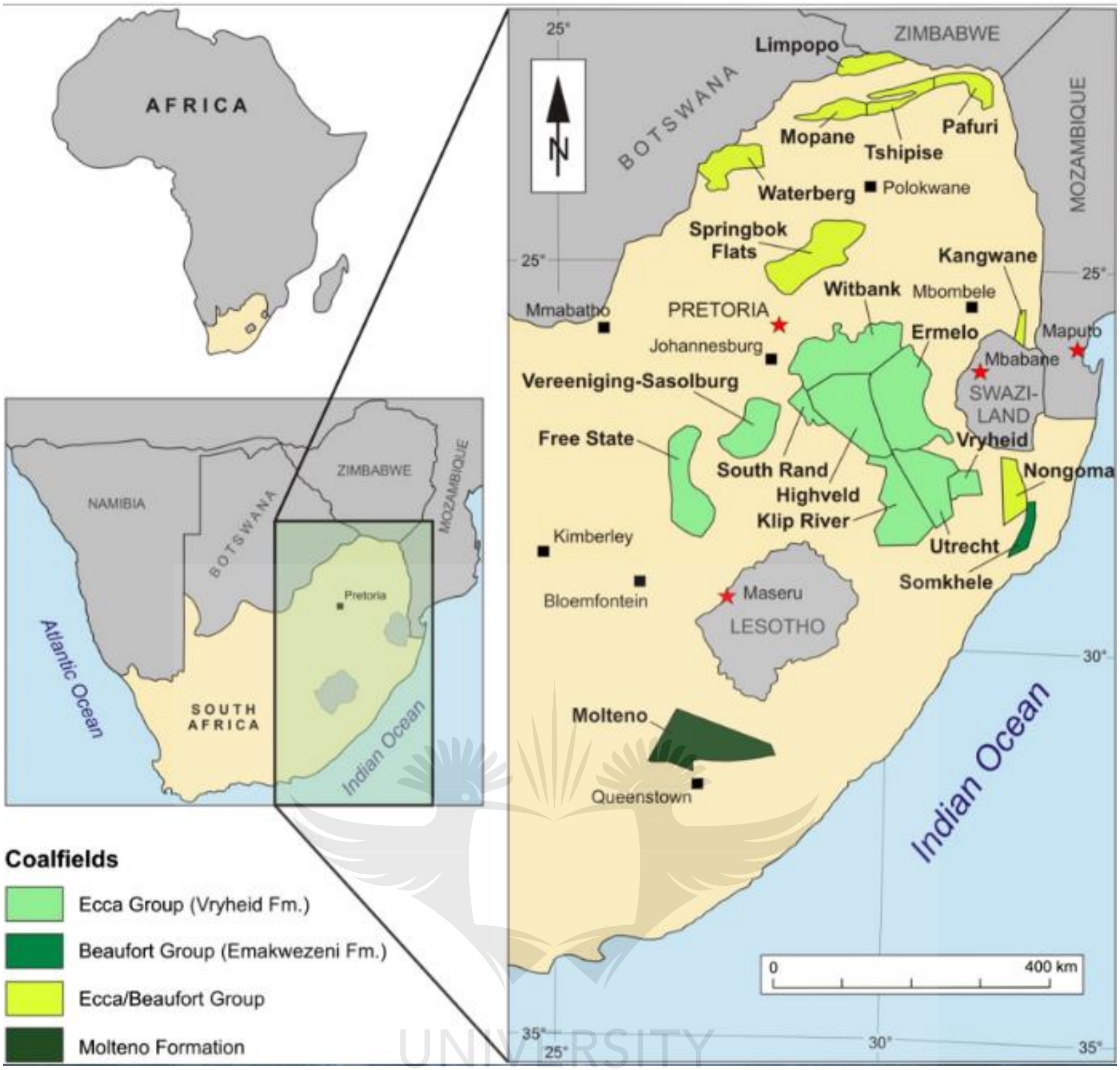


Figure 1.1 Coalfields of South Africa (Snyman, 1998)

1.4 Problem Statement

Spontaneous combustion of coal is one of the problems faced by both coal producers and users. For a period of time, the contact of open-pit walls with atmospheric conditions contributes to instability in highwalls, discontinuities and finally leads to spontaneous combustion. The degree of ventilation and amount of coal that accumulates in old workings accumulate to provide ideal conditions for self-heating. Wind direction provides airflow into the old workings, hence leading to a differential pressure from the top and bottom of the highwalls. The prediction of the liability of coal and carbonaceous shale to undergo spontaneous combustion involved the use of different spontaneous combustion tests (Onifade, 2018). However, the reliability and comparisons of the spontaneous combustion tests used in this study are yet to be established to date. Furthermore, the trend of linear relationship between coal recording basis and liability indices are yet to be reported in any studies. This research is necessary in view of the concerns raised and the fact that spontaneous combustion in coal mines is a long-standing issue.

1.5 Justification of the Research

Spontaneous combustion of coal is a type of combustion that result from self-heating without external heat application. The reason being due to variation in the microscopic organic and inorganic constituents existing in coal. This problem occurs anywhere in coal mining operations. Most of the coal fires in coalfields usually results from spontaneous combustion despite the different control measures in place (Onifade, 2018). The liability of coal to undergo self-heat differs in various coal seams. Therefore, when taking precautionary actions against the incidence of coal fires, it is important to determine their degree of propensity to prevent the loss of valuable and unrenewable resources acquired by the mine. Most studies examined the characteristics of coal on the ad basis but not taking into account other basis such as db and daf to determine the trend of linear relationships with spontaneous combustion liability indices (Nimaje & Tipathy, 2016; Eroglu, 1992). However, limited studies in the past have been carried out to compare results of different spontaneous combustion tests, evaluate the trend of linear relationship between coal recording basis and the spontaneous combustion liability indices. In addition, providing reliable prediction techniques to estimate the liability of coal and their trend of relationships with different coal recording basis can give a better evaluation of these properties.

1.6 Research Question

The coal industry has made progresses in many ways over the years, including anticipating, avoiding, tracking and managing spontaneous combustion in coal mines. Previous works to assess the liability index of coal both experimentally and computationally are well documented. However, up to date no comparisons and relationships have been established between the different liability indices and coal recording basis. Additionally, in understanding the trend of linear relationship of the self-heating characteristics of coal with consideration to various coal recording basis, no studies have been reported. Due to the mentioned concern, this raises the primary question *"Is there comparisons and relations between the results of different liability indices and the coal recording basis using R-squared value and correlation coefficients?"* This raises the following sub-questions;

- *Will the physicochemical properties of coal determined using different basis have an influence on the liability index of the coal sample; and*
- *What is the trend of linear relationship between the physicochemical properties of the coal samples and liability indices?*

1.7 Objectives of the Research

This study aims to study different spontaneous combustion tests methods and coal recording basis and examine the trend of linear relationship between them using regression analysis. The following objectives are set on the basis of the purpose of this study:

a) Evaluation of the status of current research on the topic;
b) Investigation of selected properties of coal (volatile matter, ash, fixed carbon, moisture, carbon, nitrogen, hydrogen, sulphur and oxygen contents) on ad, db and daf basis according to the ASTM standards;
c) Assessment of the spontaneous combustion liability indices of coal samples using different spontaneous combustion tests and comparisons of the results; and
d) Evaluation of the trend of linear relationships between (b) and (c) using regression analysis.

1.8 Scope of research

Simple research opportunities are provided by presenting suitable solutions to the research question, which is the basis of this study. The study focussed on the evaluation of different spontaneous combustion tests methods and coal recording basis for coal samples from the Witbank Coalfields. The results from these liability indices have been examined and compared. The trends of linear relations between the data of selected coal properties and liability indices using regression analysis were established and discussed.

1.9 Structure of thesis

The research comprises of six chapters:

Chapter 1: Introduction

This chapter presents the research topic including the background of study, the areas of study, the problem statement, the objectives of the research and research questions, the scope of the work and the structure of dissertation.

Chapter 2: Literature Review

This chapter offers a detailed review which focuses on national and international research on related study conducted on coal spontaneous combustion and coal fires.

Chapter 3: Experimental methods

This chapter explains the experimental approaches and detailed information on the coal sample testing procedures. Proximate analysis, ultimate analysis, total sulphur and spontaneous combustion tests were involved in the experimental investigation. The spontaneous combustion tests were determined using the Stage II Slope, XPT, Wits-Ehac Index, and FCC Index.

Chapter 4: Results and discussions of spontaneous combustion and coal properties test

This chapter presents and explains the results and interprets the findings of the research achieved from the experimental investigations.

Chapter 5: Relationship between coal properties and liability indices

In this chapter, statistical analysis (linear regression) was used to establish trends of linear relations between the coal properties and the liability indices.

Chapter 6: Conclusions and Recommendations

This chapter presents an overall review of the research based on the findings gained from the study. The findings of the study and their recommendations have been clarified.

1.10 Summary

This chapter presents some background information to the reader on the dissertation research topic, motivation, and structure. It also explained why it was important to compare the results of different spontaneous combustion test methods in order to better assess the risk of spontaneous combustion in coal mines. The next chapter sets out the literature survey findings on topics in-line with this study.

2 LITERATURE REVIEW

2.1 Introduction

This section focuses on the global role of various research institutes and industries conducting research on spontaneous combustion and coal mine fires on previous studies. The mechanism and possible factors that influence the incident and various approaches used to assess the risk were reported.

2.2 Mechanism of self-heating of coal

Coal self-heating happens when there is enough oxygen in the air to sustain the low-temperature reaction between coal and oxygen. Coal interaction with oxygen at low temperatures is exothermic as a whole, though some reaction sequences may be endothermic (Phillips et al., 2011). Through conduction or convection, the heat generated by coal oxidation is not dissipated sufficiently, causing a net temperature increase in a coal mass. Exothermic processes such as microbial metabolism, coal-to-water interaction and pyrite oxidation leads to self-heating within a coal mass if the heat generated by coal oxidation is not properly dissipated by conduction, convection and radiation into the atmosphere. The conditions for coal spontaneous combustion, excluding the influence of external factors are:

(a) Reaction between coal and oxygen;

(b) Exothermic reactions followed by formation of heat; and

(c) The produced heat should exceed the dissipated heat.

When coal exposed to the surface reacts with oxygen in the air, there is an adsorption/ absorption reaction that simultaneously forms heat and gaseous products. The chemical reaction is as follows in Equation 2.1 (Stracher & Taylor, 2004).

$$\text{Coal} + \text{Oxygen} \rightarrow \text{Oxygen} + \text{Heat} \rightarrow \text{Gas product} \quad (2.1)$$

The incident of spontaneous combustion is not limited to coal, but it is understood that the phenomenon occurs in other products such as spoil heaps, coal waste, coal shale and pyritic black shale (Restuccia et al., 2017; Rumball et al., 1986). In all cases, the mechanism governing spontaneous combustion are identical, regardless of the material involved. Due to the degree of metamorphism, the rate of oxygen absorption in coal depends on the rank of coal (Falcon, 1987). The heat generated by coal is the reason of coal oxidation and depends on factors such as the amount of various organic and inorganic matter present in the coal (Onifade, 2018; Beamish et al., 2001). When coal is exposed to the atmosphere for a long time, it is less reactive than fresh coal (Ren et al., 1999). According to Brooks et al. (1988), due to its low permeability, fine coal particles require limited oxygen flow, while coarse coal is highly reactive due to its high permeability and specific surface area. Oxygen can enter from the surface into coal seams and exposed some parts of the coal to oxygen in the air. The reactivity level for a coal seam depends on the time of exposure and the amount of oxygen content (Kaitano et al., 2007). When the rate of exposure increases, more heat will be generated at an increased depth, making the distribution of heat within the coal seam difficult. Figure 2.1 shows the places where there was spontaneous combustion.

Figure 2.1 (a) Self-heating of stockpile and (b) symptoms of self-heating within coal seam, Witbank Coalfields, South Africa (Onifade, 2018)

2.3 Factors affecting spontaneous combustion liability of coal

The self-heating of coal arises due to the cumulative impacts of varying amounts of intrinsic and extrinsic factors (Fierro et al., 2000; Banerjee et al., 1985). The intrinsic factors are associated with the coal inherent properties, i.e. its physical and chemical characteristics, petrographic and mineral properties. The extrinsic factors are related to the prevailing geological, mining and environmental conditions during coal mining activities, and these are largely site-specific and difficult to evaluate. In this research, an analysis of the role of the intrinsic factors that influence the liability of coal to undergo spontaneous combustion were discussed. The factors affecting the liability of coal to undergo spontaneous combustion are grouped as follows:

a) Geological factors;

b) Seam factors; and

c) Mining factors.

The mining factor is extrinsic while the seam and geological factors are intrinsic (Phillips et al., 2011; Bhattacharyya, 1982).

2.3.1 Geological factors

The geological factors that influence the spontaneous combustion of coal involves factors such as seam thickness and seam gradient, faults, bedding plane, coal friability, coal bursts, geothermal gradient, faulting, caving characteristics, surrounding strata conditions, etc. A better understanding of the impacts of some of these variables are summarised below:

- ***Seam thickness:*** In a case where the thickness of the seam is greater than what could be completely mined in a section, the area is likely to be exposed to self-heat as the unmined area appears to be subjected to slow airflow. It is understood that self-heating of coal depends on the physical factors involved in seam thickness, working methods, ventilation type, and the coal rank. Between the bands of thick coal seams, some segments could be more prone to spontaneous combustion than the others. The thinner the layer of coal, the harder it becomes in the goaf area to avoid leaving relatively high-risk coal (Phillips et al. 2011; Eroglu, 1992).
- ***Seam gradient:*** Room and pillar or longwall mining methods are commonly used in flat seams which are less likely to be combusted spontaneously. In an inclined seam, it becomes more difficult to prevent spontaneous combustion, as the convection current caused by the temperature difference tends to create air currents within the goaf (Onifade & Genc, 2019; Eroglu, 1992).
- ***Coal friability:*** The more friable the coal, the greater the surface area exposed to oxidation, which tends to generate more heat per unit volume of coal. The surface area also increases significantly as coal breaks quickly after extraction. As the surface area of the coal is increased, the atmosphere will be exposed to more surface area. The spontaneous combustion thus increases as the surface area increases (Bhattacharyya, 1982).
- ***Coal outbursts:*** In the harder formations and higher rank coal, this usually takes place rather than in the softer and lower rank coal. However, care must be taken where there

is a possibility of coal explosion and spontaneous combustion at the same time, as the risk of explosion is very high (Eroglu, 1992; Bhattacharyya, 1982).

- ***Geothermal gradient:*** This does not affect the self-heating directly; though anywhere the geothermal gradients are high, the strata temperature of the workings can rise faster with a rising working depth than with a low geothermal gradient. If a seam is in a higher geothermal gradient area, the possibility of spontaneous combustion is high (Eroglu, 1992; Banerjee et al., 1985).
- ***Fault:*** Coal seams with many discontinuities such as fault commonly allow the ingress of oxygen in the air which promote the occurrence of self-heating. Any drilling on the fault plane that results in the forming of the fine coal and the ventilating air with which the fault enters the seam and oxidises the fine coal can lead to spontaneous combustion. A fault usually slows the rate of face advance to a reasonable minimum, with the incidental possibility of heat generation (Bhattacharyya, 1982).

2.3.2 Seam factors

Seam factors such as petrographic composition, mineral content, sulphur and sulphur types, coalification, porosity, volatile matter, surface area, particle size distribution, inherent moisture etc. have been well documented to influence the liability of coal to undergo spontaneous combustion (Onifade & Genc 2019; Kaymakci & Didari, 2002; Beamish et al., 2000; Akgun & Arisoy, 1994a; Falcon, 1987). According to Nugroho et al. (2000), coal mass with a small particle diameter (+ 0.18 - 2.67 mm) can experience self-heating faster than those with a fine particle size (- 0.18 mm) because large particles have a reduced total surface area and bulk density effect. A study reported by Bhat & Agarwal (1996) showed that the size of coal particles affects heat loss through convection and the heat generated by the mass transfer coefficient which controls the degree of condensation of moisture. Kim (1977) stated an inverse relationship between the size of coal particles and the spontaneous combustion liability index.

Studies have been reported on the effects of moisture and oxygen content on the liability of coal (Arisoy & Akgun, 2000). Beamish & Hamilton (2005) found that as a result of adsorption, irregular wetting and drying of coal on stockpile surfaces facilitates spontaneous combustion. Akgun & Arisoy (1994b) suggested that moisture is extracted from the coal when dry air flows over wet coal, resulting in a decrease in temperature. While misty air reaches dry coal, temperature increases due to moisture adsorbed from the atmosphere. Dry coal requires temperature changes that affect the heat balance during coal oxidation and cause self-cooling, while wet coal is an exothermic reaction that releases heat to aid self-heating (Akgun & Arisoy, 1994b).

A number of studies have indicated that as the ash content increases, the liability of coal decreases due to a decrease in the amount of inert and organic material serving as a heat sink (Nimaje & Tripathy, 2016; Panigrahi & Sahu, 2004). Beamish & Blazak (2005) reported a negative relationship between R70 (Australian test for predicting spontaneous combustion of coal) and ash content for low-to-high rank coal.

The amounts of mineral matter present in coal have been reported in previous studies (Mastalerz et al., 2010; Suarez-Ruiz & Crelling, 2008). Studies indicated that due to the presence of a number of varying organic and inorganic matter in coal, the rate of self-heating can be delayed (Beamish & Arisoy, 2008; Beamish et al., 2005).

The presence and distribution of sulphur content in coal have been well documented (Mastalerz et al., 2010; He et al., 1999). Excessive quantity of sulphur in coal accelerates self-heating, which eventually aids spontaneous combustion. Sulphur in coal is classed as organic and inorganic sulphur. Organic sulphur and the macromolecular composition are difficult to distinguish. Inorganic sulphur in coal is in the form of pyrite and sulphate. The volume of pyritic and sulphate sulphur varies with coal. Pyrite, as the main component of inorganic sulphur, has important effects on the spontaneous combustion of coal (Mastalerz et al., 2010). The reaction of pyrite with oxygen in the presence of water produces hydro-peroxide (H_2O_2), which eventually causes low-temperature oxidation. Pyrite with a concentration of more than 2% has been documented to aid oxidation of coal (Onifade, 2018). Beamish & Beamish (2012) suggested that the type of pyrite present in a coal dictates the rate of self-heating, but not the quantity of pyrite. The chemical reaction for pyrite oxidation is stated as follows in Equation 2.2 (Lain, 2009):

$$Fe_2S + 7O_2 + 16H_2O = 2H_2SO_4 + 2FeSO_4.7H_2O + 1321kJ \qquad (2.2)$$

The above reaction is exothermic and occurs at low temperatures. The heat produced from the reaction is double that of coal with the same oxygen (Martinez et al., 2009). The presence of pyrite is therefore a primary factor in the spontaneous combustion of coal.

The effects of petrographic composition on the liability index of coal have been reported by Falcon (1978); Onifade (2018) and Nimaje & Tripathy (2016). The amounts of the liptinite content in South African coal is very small compared to the other macerals. Macerals such as inertinite, liptinite and vitrinite are subject to self-heating and weathering in terms of time, temperature and environmental conditions, but the effects of each maceral on spontaneous combustion are different. Studies published by Beamish & Blazak (2005) and Ivanova & Zaitseva (2006) have verified that vitrinite is more vulnerable to spontaneous combustion compared to the other two macerals due to the relationship between coalification and low-temperature oxidation. The amount and type of macerals and the degree of coalification have been reported to be reliable variables for estimating the liability of coal to undergo spontaneous combustion (Avila, 2012; Morris & Atkinson, 1988).

2.3.3 Mining factors

In open cast mines, the amount of coal left on the coaling bench, discontinuities within the benches and coal seams influences self-heating (Onifade & Genc, 2019; Phillips et al., 2011). In underground mines, parameters such as ventilation system and airflow, pillar and roof conditions, rate of advance, multi-seam working, mining methods, waste material in abandoned areas, heat from machines, worked-out areas, etc. affects the liability of coal (Bhattacharyya, 1982). The mining factors affecting the liability of coal are summarized below:

- ***Ventilation system and rate of airflow:*** The rate of air flow is a complex variable since air supplies enough oxygen while also transporting the heat created. There is a sufficient amount of air that provides enough oxygen to allow coal to oxidise, but it is not enough to stop the heat from accumulating (Kim, 1977).
- ***Leakage:*** This happens where leakage path flows through cracks at air passages, in and around regulators and doors, and other similar areas where the outflow is a high-pressure gradient and a tendency for air to flow through solid coal. Ideally, it is not necessary to depend on impermeable stops because, if they were fully impermeable, a dangerous fire-amp pressure would be seen (Kim, 1977).
- ***Pillar conditions:*** The size and strength of the pillar directly influences the liability of coal. Ideally, to prevent collapse, the pillars should be of an appropriate size. This size depends on the coal strength, the thickness of the cover, and the impact of other workings within the panel. Increased in methane emissions are an indicator of pillars being crushed, resulting in self-heating (Phillips et al., 2011; Bhattacharyya, 1982).
- ***Roof conditions:*** Poor roof conditions allows a simple passage of shock waves and increases crack developments within the roof, potentially leading to spontaneous combustion. Poor roof conditions may likely increase the tendency of coal towards

spontaneous combustion. Roof fall leaves openings that must be protected and filled with timber on a regular basis. Such areas are always filled with methane because of their origin and often result in spontaneous combustion (Phillips et al., 2011; Eroglu, 1992).

- ***Rate of advance:*** The rate of advance in the course of mining could be a source of spontaneous combustion during coal mining. Ideally, when a working face operates normally, any single piece of coal passes through the region at a pace equivalent to the rate of advance of the working face. The time taken to enter or leave the area for any single piece of coal is very crucial. If the time is excessive, an unacceptable degree of oxidation may result in spontaneous combustion (Eroglu, 1992; Bhattacharyya, 1982).
- ***Multi seam workings:*** Spontaneous combustion may develop in cases where one seam is mined above or below the other seam due to roof conditions and air leakage. Where there is a multi-seam case, there may be an event of spontaneous combustion for the seam currently being worked and any other seam above or below it (Phillips et al., 2011; Eroglu, 1992).
- ***Mining methods:*** During underground mining, partial pillar removal, where most of the coal seam remains in the goaf and pillars, facilitates spontaneous combustion. This is important during the mining of old workings, as surface mining methods expose areas where crushed coal has been left over a period of time, thereby leading to spontaneous combustion. Longwall mining leaves areas removed between the entries as the areas of workings. The pressure difference in the ventilation system allows air to flow through these areas, leading to spontaneous combustion (Onifade & Genc, 2019; Bhattacharyya, 1982).
- ***Heat from machines:*** Generally, heat energy from machinery is dissipated in the ventilating air stream and the average increase in surrounding temperature of the air can be very high. If the impact of heat energy from machinery is secondary, additional air may need to be circulated, which will require increased ventilation pressure resulting in an increased risk of leakage (Phillips et al., 2011; Eroglu, 1992).
- ***Worked-out areas:*** Potential causes of spontaneous combustion are worked-out areas that are not adequately protected by ventilation stoppings. Suspension in the ventilation system is expected due to roof lifts or floor lifts that appear to hinder the condition of the rib and the accumulation of broken coal that leads to possible combustion (Phillips et al., 2011).

2.4 Methods for predicting the spontaneous combustion liability of coal

The prediction of the liability of coal to undergo spontaneous combustion involves the identification of different coal based on their properties and utilizing different spontaneous combustion tests methods. The various test methods used to estimate the liability of coal or self-heating characteristics of coal are briefly described.

2.4.1 Differential scanning calorimetry (DSC)

This method is used to calculate changes in the energy inputs provided to a sample and a reference material with respect to temperature when this material is held at a fixed temperature (Mohalik et al., 2009). Mahajan & Walker (1971) used this method to evaluate the self-heating characteristics of four Indian coal samples. This approach suggested that the variables obtained from DSC were not consistent based on different studies published (Mohalik et al., 2009). Mohalik et al. (2010) proposed a correction of the modified experimental variables used. The approach does not provide a clear understanding of how two coal samples are responsible for spontaneous combustion.

2.4.2 Thermogravimetric analysis (TGA)

In the power industry, this method has been in use to analyse coal reactivity, obtain kinetic parameters of fuel and to measure physical characteristics such as the quantity of ash, volatile matter, fixed carbon and total moisture (Vaan Graan & Bunt, 2016; Avila, 2012). Avila et al. (2014) suggested the possibility of achieving reliable results with the use of different heating ramps during low-temperature oxidation, the calculation of absorbed and released oxygen, the amount of moisture and volatile matter, and the estimation of reactive and non-reactive coal components. This process measures the weight loss of coal at high temperatures as a result of self-heating. A particular weight of coal is heated and plotted against time/temperature by means of a fixed heating ramp. The results obtained are referred to as thermogravimetric (TG) curves (Vaan Graan & Bunt, 2016; Avila et al., 2014; Avila, 2012).

2.4.3 Russian U index

This method calculates the volume of oxygen consumed over 24 hours by a coal. By determining the gas composition, the gasses collected under experimental conditions can be quantified. Banerjee (2000) claimed that the oxygen consumed during the test was directly proportional to the liability of the coal. The drawback in this process is that the amount of oxygen consumed by a coal during testing is not reproducible when the experiment is carried out on the same coal.

2.4.4 Crossing point temperature (XPT)

Several studies have used this test to assess the liability of specific coal to undergo spontaneous combustion in relation to its ignition temperature (Gouws, 1987; Banerjee, 1985; Humphreys, 1979). The system involves heating coal in an oxidised state that causes oxidation either at an automatic heating level or at a particular temperature from the ambient temperature to the ignition point temperature of the coal. Studies documented using this approach showed a lower liability index for coal with a higher XPT and vice versa (Gouws, 1987; Banerjee, 1985).

2.4.5 X-Ray Diffractometer (XRD)

Research have been carried out to evaluate the quantity and identification of various minerals using X-ray diffraction methods in a particular coal to undergo spontaneous combustion (Gong et al., 1998). The results obtained from this process were compared with the results from petrographic analysis and other thermal methods to check the validity of this procedure. It was proposed that changes in iron minerals in coal can result from the catalytic reactions from iron minerals during the oxidation process (Gong et al., 1998). This method indicated that pyrite, among other minerals, can definitely be altered during coal oxidation, with pyrite oxidation varying with the other forms of sulphur and mineral composition (Ribeiro et al., 2016).

2.4.6 Olpinski index method

In this method, a pellet of fine coal is heated up at a constant rate in a quinolone steam bath, while air is discharged to the sample. Time versus the temperature curve is measured until a temperature of 235 ^{o}C is reached. At this point, the increase in temperature of the coal is used to determine the liability (Szb Index) of the coal (Banerjee, 1985). This approach has been used for Indian coal to create relationships with proximate and ultimate analysis (Nimaje & Tripathy, 2016; Nimaje et al., 2013).

2.4.7 Adiabatic calorimetric method

This method is commonly used in many countries to replicate the self-heating characteristics of coal (Ren et al., 1999; Cliff et al., 1996). This method involves putting a coal in a reaction vessel using an adiabatic oven or oil bath to the degree that the heat is not released from the vessel. The temperature of the coal is controlled in the reaction vessel at particular intervals relative to the increase in coal temperature and air/oxygen is allowed to pass through the reaction vessel and hence, determining the liability of the coal to spontaneous combustion. A study documented by Cudmore (1988) explored the impact of inherent moisture and air humidity under adiabatic conditions, but not the degree to which it influences the liability of coal to undergo spontaneous combustion. Some limitations have been found in the use of this method; such as time to complete an experiment, the configuration of reaction columns and the necessary quantity of samples required, the exact particle sizes and the correct air/oxygen flow rate.

2.4.8 Wits-Ehac tests

The Wits-Ehac testing method has been in use since the 1980's to estimate the spontaneous combustion liability of coal and coal-shale (Genc et al., 2018; Genc & Cook 2015; Eroglu, 1992). The method combines the XPT and DTA calculated from the coal at periodic temperature measurement to generate a self-heating liability index. The index takes advantage of the fact that coal, which is more liable to self-heating, has a steeper stage II slope and a lower XPT than coal, which is less liable to self-heating. A full description of this testing method is well reported by Wade et al. (1987).

2.5 Analysis for the prediction of spontaneous combustion

Previous works carried out with consideration into intrinsic and extrinsic factors and spontaneous combustion tests methods are classified into mathematical modelling, experimental and statistical analyses. A brief descriptions of various approaches used by academics/researchers/industries for analysing the mechanism of spontaneous combustion is described below:

2.5.1 Mathematical modelling

Self-heating, which leads to spontaneous combustion of coal, is caused by a number of factors such as intrinsic and extrinsic factors. A number of mathematical models are established to assess the characteristics of coal and the influence of different factors leading to spontaneous combustion (Yuan & Smith, 2012; Krishnaswany et al., 1996). Nelson & Chen (2007) explained how a single Arrhenius formula could determine the rate of oxygen adsorption in self-heating of coal. For instance, some assumptions are used in mathematical modelling that the self-heating of coal which could lead to spontaneous combustion is caused by the absorption of insignificant amounts of oxygen. It is also commonly expected that all coal has similar activation energy, “t’’. This assumption has resulted in a method that is ineffective for determining safety in shipping coal (Nelson & Chen, 2007). In addition, Nelson & Chen (2007) reported the risk caused by the self-heating characteristics of coal using the Frank-Kamenetskii self-heating analysis.

Brooks et al. (1988) developed a mathematical model to calculate the propensity of coal stockpiles to undergo spontaneous combustion. By reducing the lumps of coal into three parts, the model was developed. As used by Brooks & Glasser (1986), where reaction flow and heat transfer take place, the model integrated a-one-dimensional chimney area. It is assumed that the gas reaching the bottom of the chimney was depleted by the oxygen stream through the whole coal bed. The influences of particle size on coal spontaneous combustion was studied by Akgun & Arisoy (1994b). The research examined coal with different particle sizes which were oxidised in a cylinder 3 m long and 0.3 m in diameter. There was a critical particle range where self-heating resulted in spontaneous combustion. Bhat and Agarwal (1996) established a mathematical model that explores the deposition and elimination of moisture in a coal. The model established studied the impact of moisture migration on coal oxidation. By reducing the impact of moisture in a coal stockpile, Krishnaswamy et al. (1996) developed a two-dimensional model to estimate the liability to undergo spontaneous combustion. The study indicated that the main source of airflow within a stockpile is wind-driven forced convection (Krishnaswamy et al., 1996).

Rosema et al. (2001) established a numerical simulation model called "COAL TEMP" to investigate the oxidation and potential for coal spontaneous combustion under the influence of atmospheric conditions. It was found that the study area has a low liability for spontaneous combustion and that self-heat can be identified as a hot spot at the initial heating stage using thermal infrared viewing systems (Rosema et al., 2001). Fierro et al. (2001) used a one-dimensional model to create model on stockpiles containing lignite coal samples that was considered unsafe due to spontaneous combustion. The mathematical model developed showed that in spontaneous combustion of coal stockpiles, the porosity and wind flow played an important role. Arisoy et al. (2006) used a dimensional test column to examine the spontaneous combustion risk of coal stockpiles both theoretically and experimentally. A model was established to simulate a one-dimensional full-scale test column. To simulate large-scale storage conditions, predictions from this model were used. The experiment conducted closely emulated the prediction model for moisture transfer characteristics. Humphreys (2005) identified a study that integrates the data on the mechanisms of spontaneous combustion and heat loss. Numerical modelling was employed to simulate the coal's experimental results and self-heating characteristics. The study developed relations between the results of the experimental study and the rate of self-heating (Humphreys, 2005). Zhang et al. (2016) established a two-dimensional mathematical model for the assessment of the liability of coal stockpile to undergo spontaneous combustion. The developed model examined the kinematics parameters and various factors that impact the spontaneous combustion of coal stockpiles under different conditions.

Arisoy & Beamish (2015) used an adiabatic oven experiment to analyse the liability of coal based on the data obtained from the reaction kinetics of coal oxidation. The time versus temperature impact on the reaction kinetics were estimated based on repeated tests on the coal at different temperatures (20 to 57 ^{o}C). The initial reaction rate at low temperatures was mainly non-Arrhenius, but crosses at temperatures above 70 ^{o}C with Arrhenius behaviour. A new rate model equation was developed which took into account variations in the kinetic characteristics of the low-temperature zones. Yuan & Smith (2008) modelled the influences of coal properties on coal spontaneous combustion in longwall gob areas using a CFD analysis. The results of the model showed that mine ventilation and coal properties can affect coal spontaneous combustion during long-wall mining. The concentration of oxygen and temperatures in coal stockpiles was calculated by Zhang et al. (2009). During the experiment, a BP-ANN (artificial neural network) with 150 training samples were created. The temperature of the coal column and concentration of oxygen are chosen as output variables. To calculate the trend on the untried experimental data, the trained network was used. The average oxygen concentration and temperature errors, based on the ANN estimation, were 0.5 percent and 7 ^{o}C respectively, corresponding to actual tolerances. This approach has been found to allow a practical guide to understand the sources of spontaneous combustion in coal stockpiles (Zhang et al., 2009).

2.5.2 Experimental methods

Several works have attempted to measure the self-heating and liability of coal to undergo spontaneous combustion using various laboratory techniques and different countries around the globe are adopting different techniques for this purpose. The results produced from these techniques were used to categorise coal based on their liability indices. Gouws et al. (1991) developed an adiabatic calorimeter to determine the liability of coal to undergo spontaneous combustion. The calorimeter was produced to replace and simplify the test procedure of the ignition temperature apparatus. Results obtained from the adiabatic tests were compared with those generated from the XPT and DTA tests for some selected coal. This was done by creating a definition for a reliable liability index for coal (Gouws et al., 1991). Panigrahi et al. (1997) performed laboratory tests to examine the Russian U-Index using coal samples from Jharia Coalfields. The XPT of the samples were assessed and their intrinsic properties were analysed. The Russian U-Index displayed similar relationships with these coal properties in relation to spontaneous combustion as shown by the XPT (Panigrahi et al., 1997).

Uludag (2007) examined the liability indices of some coal samples from the South African Coalfields using the Wits-Ehac apparatus. The results from the coal properties and the DTA and XPT methods were analysed. The relationships obtained from the tests between DTA are compared to the actual amount of moisture. It was found that the Wits-Ehac Index increased as the coal moisture rose, while the volatile matter had a catalytic influence on self-heating when the moisture was reduced. Nevertheless, as carbon content rises, the value of the Wits-Ehac Index increases, while the likelihood of spontaneous combustion decreases (Uludag, 2007). Sahu et al. (2004) examined the spontaneous combustion liability of 30 Indian coal using the DSC. The study observed that an increase in temperature is an indication of self-heating potential. In another study, Sahu et al. (2005) used XPT, DTA and DSC to estimate the liability of some Indian coal samples. The study observed that the initial temperature results from the DSC experiment has been found to produce better results than the XPT. The effect of maceral and mean reflectance (Rmax) on the propensity of coal to spontaneous combustion was determined by Choudhury et al. (2007). The liability of the coal was determined using the TGA, and the quantity of vitrinite and liptinite in the coal indicated a high liability index. Tognotti et al. (1988) reported a laboratory tests for determining the ignition temperature of various cylinder-shaped coal beds. The models of thermal ignition were used to describe and extrapolate the test results. Measurement of the heat produced under isothermal conditions was recorded and compared to the results of the ignition test. The study showed that the coal undergoes self-heating at a temperature of between 120 and 220 ^{o}C.

Kucuk et al. (2003) examined the impacts of the gas absorption, moisture and particle size of lignite coal samples from a stockpile by using the XPT in Turkey. The study showed that with a decrease in particle size and air humidity, and an increase in the amount of moisture, the self-heating potential of lignite increased. Zhang et al. (2011) created a nested spontaneous combustion furnace with improved performance in thermal insulation. To solve the problems of heat dissipation and temperature control, the temperature control system with higher accuracy was selected. Taraba & Pavelek (2014) used pulse flow calorimetry (PFC) to examine the spontaneous combustion liability of different coal by using the data obtained from oxidation heat q30 (wKg^{-1}). The study categorised the liability of these coal samples according to the value of the oxidation heat q30.

2.5.3 Statistical methods

Several works have used coal characterisation tests such as proximate and elemental analysis, petrographic properties and spontaneous combustion tests to evaluate different liability indices using statistical analysis. Nimaje & Tripathy (2016) studied the influences of intrinsic properties and liability indices of 49 coal samples by using the XPT, DTA, FT, Olpinski Index and WOE. The study showed that the elements of ultimate analysis indicated a better relation with the Olpinski index than the other liability indices based on the statistical analysis (Nimaje & Tripathy, 2016). Another study by Nimaje et al. (2013) used various methods such as petrography, thermal and oxygen absorption, and XPT to evaluate the liability of coal to undergo spontaneous combustion. The study showed that the Mixture Surface Regression (MSR) method offers better residual values relative to the polynomial and simple multiple regression models using statistical analysis based on the results produced from the XPT and proximate tests. Kaymakci & Didari (2000) established relations between the liability indices and coal properties using linear and multiple regression analysis. The linear regression analysis indicated that the hydrogen, ash, carbon, volatile matter, exinite, and inertinite affects spontaneous combustion, while the multiple regression analysis showed that the sulphur, volatile matter, nitrogen, oxygen, carbon, hydrogen and inertinite influences the spontaneous combustion of coal (Kaymakci & Didari, 2000).

A similar study was conducted by Singh & Demirbilek (1987) by using statistical analysis to examine the relations between coal properties and liability indices of coal. A statistical evaluation comprising of multiple regression between the liability indices (dependent variable) and coal properties (13 independent variables) was applied to form models to predict the liability of coal to undergo spontaneous combustion. The effects of the selected coal properties on liability indices was established using the isolated factor techniques (Singh & Demirbilek, 1987). Onifade & Genc (2018) established relationships between selected coal properties and liability indices (Wits-CT Index and Wits-Ehac Index) of coal and coal-shale using linear regression analysis. Analyses of linear regression show that ash, total carbon, hydrogen, and nitrogen influences coal spontaneous combustion. Whereas the Wits-CT index confirms this result, the Wits-Ehac Index does not suggest any significant relationship with any of the independent variables. In addition, analyses of linear regression analyses show that ash, carbon, hydrogen, and nitrogen affects the spontaneous combustion of coal-shale, and is confirmed by the Wits-CT index (Onifade, 2018). The study showed that there are positive or negative relations between the coal properties and liability indices.

Research globally have examined the liability of coal to undergo spontaneous combustion by using several methods such as XPT, differential scanning calorimetry (DSC), R70, Wits-CT Index, WOE, adiabatic calorimeter, TGA Wits-Ehac Index, Olpinski Index, Russian U Index, MR Index, FCC Index, etc. The crux of their work is the use of a laboratory experiment to measure the liability of different coal samples to self-heat. A number of experimental methods have been reviewed and a decision was made to carry out experimental tests using the XPT and DTA. Some characteristics of coal thermograms thought to be indicative of spontaneous combustion liability such as XPT, Stage II Slope and low transition temperatures from stage II to stage III referred to as simple indices, are identified to be inconsistent in the assessment of the liability of coal to spontaneous combustion. The concept of composite indices, comprising of two or more simple indices were evaluated and compared with the results obtained from the simple indices in order to overcome these difficulties.

Considering the values obtained from each of the liability indices, coal has been mostly grouped based on their liability to undergo spontaneous combustion as high, moderate and low liability. However, out of these testing methods, no comparisons have been established from the results obtained from each technique to validate the liability of different coal to self-heat. In this work, as previously stated, an attempt was made to evaluate selected coal samples from the Witbank Coalfields and to conduct laboratory tests to examine their liability to undergo spontaneous combustion by using established simple and composite indices. Furthermore, selected properties (moisture, ash, volatile matter, fixed carbon, total carbon, hydrogen, nitrogen, oxygen and total sulphur contents) of all the coal samples which contribute to spontaneous combustion are determined. The results of the different spontaneous combustion tests are compared with each other and related to the selected coal properties. The results of the experimental tests are used to evaluate the characteristics known to be an indicator of the liability of coal to self-heat.

Despite the long history of spontaneous combustion occurring in coal and other related industries, studies have shown that the physical and chemical processes that are responsible for this issue are very complex. The challenges associated with better understanding of the causes of spontaneous combustion and evaluation of the spontaneous combustion liability of coal are complicated due to the fact that no simple test is considered to be the standard for determining coal that is prone to spontaneous combustion. Geologically, similar coal from the same seam at different bands varies significantly (Onifade et al., 2019).

Several works have used conventional characterisation of coal properties to estimate the liability of coal to spontaneous combustion (Arisoy & Beamish, 2015; Nimaje et al., 2013; Sahu et al., 2004). The experience of the coal industries from previous works has shown that the liability of coal varies in individual mines. The Frank-Kamenetskii theory of self-heating materials (mathematical model derived from a mass and energy balance) has been used by different researches to calculate coal spontaneous combustion liability. While some studies have used mathematical models and simulations to determine coal spontaneous combustion taking into account heat of oxidation, heat conduction, and convective heat transfer, and ignoring other mechanisms of heat exchange by using the equation of thermal equilibrium. It is possible to determine variables such as gas variability, temperature, oxygen consumption, heat intensity and assess the combustion zone using the empirical approach by simulating the spontaneous combustion of coal.

A number of computerised systems have been created for data acquisition and evaluation of the self-heating characteristics of coal. An analytical solution to the problem is difficult to find and the problem is therefore approached either experimentally or numerically on a regular basis. A number of preliminary mathematical models were developed to numerically examine the issue before using high-performance fluid-thermodynamics computational code. The use of mathematical models to determine the self-heating and spontaneous combustion liability provides valuable insights into this issue, but accuracy and applicability are challenged as these models are developed with many simplifications and constraints. The current improvement in the use of soft computing techniques and regression analysis to interpret, evaluate and model the liability of coal to undergo spontaneous combustion within reasonable engineering accuracy have been proved by previous works to be achievable (Said et al., 2020; Nimaje & Tripathy, 2016).

Research has been performed by various institutes, industries and academics to determine the cause of spontaneous combustion in coal mines using various intrinsic and extrinsic properties and various spontaneous combustion test methods. Spontaneous combustion tests have been used for many years to measure the liability of coal to undergo spontaneous combustion. Researchers, however, do not agree on the use of a particular method to predict the liability of coal. Several works have shown that some methods may be attempted to assess the liability of some coal fairly accurately. In view of this, there are a range of experimental methods available to evaluate the liability of specific coal for spontaneous combustion, but none is superior to the other. This study acknowledges that an investigation of different spontaneous combustion liability indices is of importance and in-depth analysis is necessary for the comparisons of the spontaneous combustion test methods. Studies have shown that the liability of coal to spontaneous combustion can be tested using different methods. Such methods have been shown to be accurate in their application, but the point that no precise testing technique has become a standard confirms that there is still uncertainty as to their validity. This necessitated this research to compare the results of a number of spontaneous combustion test approaches and their trend of linear relationship with the coal recording basis.

2.6 Summary

This chapter revised the mechanism of spontaneous combustion, factors influencing spontaneous combustion, and prediction methods and techniques for the analysis of spontaneous combustion. This section reviewed a number of competing factors and mechanisms in low-temperature oxidation of coal that determines whether spontaneous combustion can take place. The various experimental methods used in this research are described in the subsequent Chapter.

3 EXPERIMENTAL METHODS

3.1 Introduction

This chapter explains the experimental methods and detailed information on the coal sample testing procedures for proximate, ultimate, total sulphur and spontaneous combustion tests. The origin of the coal seams and the methods used for the collection of samples in the study area are discussed. The statistical tool used to evaluate the trend in linear relations between the coal properties and liability indices is described.

3.2 Sample location

Thirty representative *in-situ* coal samples produced from selected open cast mines in the Witbank Coalfields were used. Figure 3.1 shows the area (highlighted in red) in which the samples were collected and other South African Coalfields.

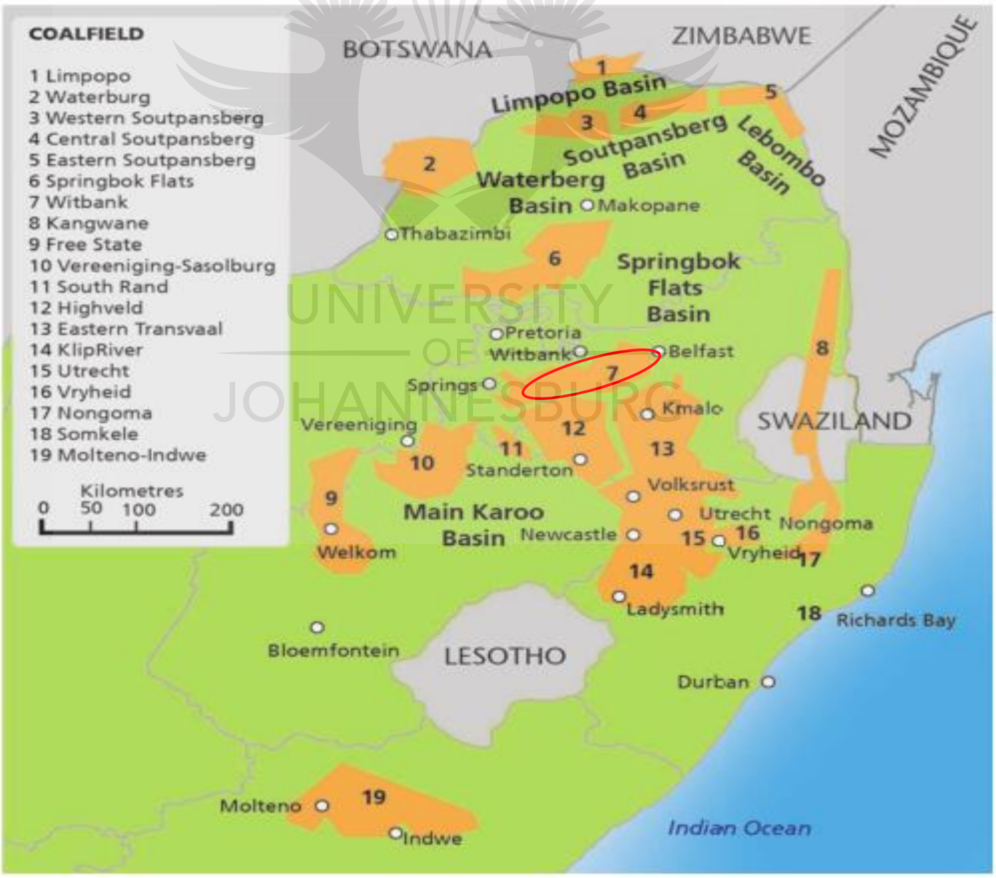


Figure 3.1 South African Coalfields indicating the study area (Dejager, 1983)

3.3 Description of the study area

The Witbank Coalfields have different types of coal that make it economically suitable for power generation, metallurgical, domestic and chemical use, etc. The Witbank coalfields comprises of No. 5 seams with a thick sedimentary order (shale, mudstone, sandstone, and siltstone) as illustrated in Figure 3.2. The seams are numbered from 1 to 5 from the bottom to the top. Pre-Karoo topography influences the distribution of the seams No. 1 and No. 2 whereas the seams No. 4 and No. 5 are eroded over a large area (Snyman, 1998). Figure 3.2 describes the typical stratigraphic column for the Vryheid Formation in the Witbank Coalfields.

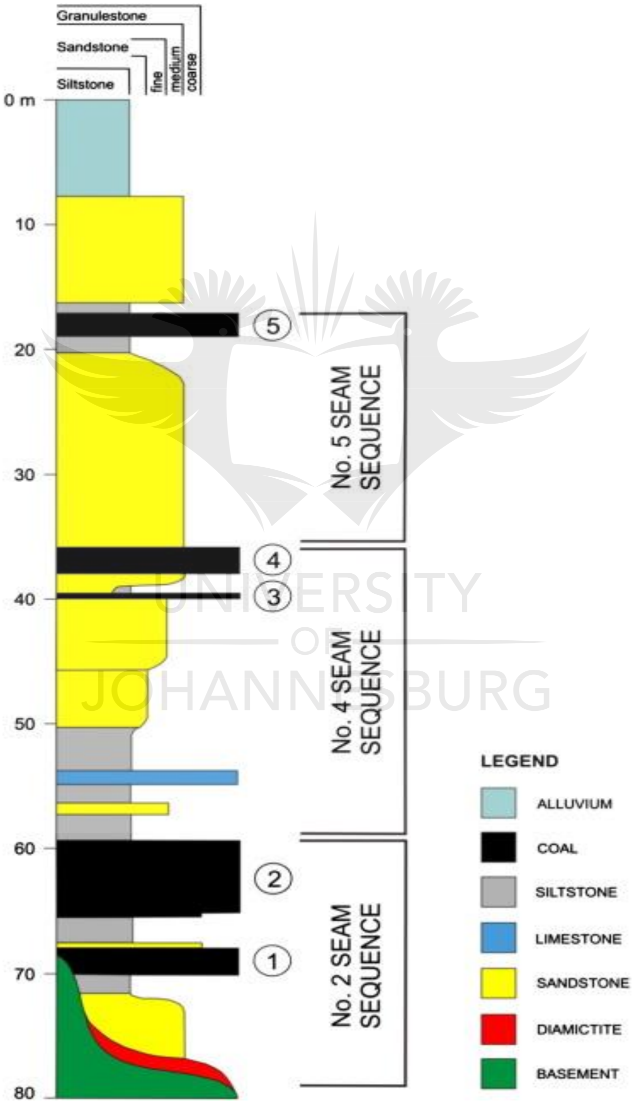


Figure 3.2 Stratigraphic column of the Vryheid Formation for Witbank Coalfields, South Africa (Hancox & Gotz, 2014).

(i) The No.1 coal seam: The coal seam No.1 is a high-grade seam coal that is appropriate for export after beneficiation (Snyman, 1998). It consists mainly of lustrous to dull coal with shale sandstone partings resulting in localised No. 1 lower seam around the Arnot area. The seam is low in phosphorus and is mined as a metallurgical feedstock separately in most cases (Pinheiro, 1999).

(ii) The No.2 coal seam: It is the basis of steam coal for high-yield exports (Pinheiro, 1999). It consists of different partings of sandstone and shale, with the thickest parting in the upper seam. This leads to the division of the seam into No. 2 and No. 2 upper seam (Snyman, 1998). It ranges in thickness and reaches a peak thickness of 8 m in the region of Ogies before thinning to around 3 m in the East of Arnot Colliery. It is mainly extracted for low-ash metallurgical coal production and steam coal exports.

(iii) The No. 3 seam is inconsistently developed and thin, in most places rarely reaching 0.5 m thickness. Because of its thickness, it is generally assessed as being uneconomic. It is locally of high quality which enables the seam to be extracted by opencast methods with a thickness of 0.8 m (Snyman, 1998).

(iv) The No. 4 seam: In the central Witbank region, the thickness of the No. 4 seam varies from 2.5 m to 6.5 m (Snyman, 1998). The seam comprises of various mudstones of sand and partings of siltstone. It separates the seam into seams No.4, often referred to as seams No.4 lower, No.4 higher, and No.4 A. Usually the partings are not too thick for selected mining (Pinheiro, 1999). The upper portion of the seam mostly consists of dull to dull lustrous coal with poor quality.

(v) The No. 5 seam rarely reaches 2 m. It consists of a mixture of mainly bright, banded coal with thin shale and mudstone partings. The seam is eroded in large areas. The seam is mostly used for local steel processing by the metallurgical industries (Snyman, 1998).

3.4 Sample collection and characterization

Thirty coal samples obtained from Witbank Coalfields were used in this study. The samples were stored in an airtight bag to prevent further oxidation. They were pulverized to -250 µm size fraction for the proximate and ultimate analysis and -212 µm for the spontaneous combustion tests. The proximate analyses for the samples are performed according to the American Society for Testing and Materials (ASTM, D5142). Each of the sample (1 g) was used to calculate the moisture, ash and volatile matter contents, while the fixed carbon is calculated as the 100% minus (volatile matter + ash + moisture). For the ultimate analysis, 0.25

g of each coal was used at a temperature up to 1,450 ^{o}C with an analysing time from 60 to 300 seconds according to the ASTM (ASTM, D5373-14:2015) for carbon (C), hydrogen (H) and nitrogen (N) with the use of the equipment (LECO CHN 628 with an add on 628 S module). A brief description of experimental procedure is described below:

Fixed carbon content

The fixed carbon is calculated by subtracting from 100% the percentage volatile matter, ash and moisture as seen in Equation 3.1.

$$\mathrm{FC\%} = 100 - (\mathrm{M\%} + \mathrm{VM\%} + \mathrm{A\%}) \qquad (3.1)$$

3.4.1 Ultimate analysis

The amounts of total carbon (C), hydrogen (H), nitrogen (N) and total sulphur (S) of the samples were conducted in compliance with ASTM, D5373-14:2015 using a LECO TruSpec CHN and LECO C & S analyser for the sulphur.

Determination of Carbon, Hydrogen, Nitrogen and Total sulphur

The C, H, N module comprised of an electric furnace that functions at a temperature of 950 ^{o}C where the sample is introduced and combusted. C and H were found by two respective infrared cells, and N was found by a thermal conductivity cell. The TruSpec LECO design had two modules, one for the analysis of sulphur and the other for carbon, hydrogen and nitrogen. The sulphur module has a furnace that can reach a temperature of 1350 ^{o}C. The sulphur oxidised to form SO_2, which flowed through the detection system that comprises of an infrared cell located in the C, H, N module when the sample was placed in a pure oxygen environment.

Determination of oxygen content (ASTM, D3176-15)

The amount of oxygen (O) is calculated by subtracting from 100% the percentage hydrogen, nitrogen, total carbon, total sulphur, moisture and ash contents as seen in Equation 3.2.

$$\mathrm{O\%} = 100 - (\mathrm{C\%} + \mathrm{H\%} + \mathrm{N\%} + S\% + \mathrm{M\%} + \mathrm{A\%}) \qquad (3.2)$$

where **O**% is the amount of oxygen (%)

The following relations (ASTM: D3180 – 07) were used to convert the as received results of the proximate and ultimate analyses to % db and % daf basis.

For proximate analysis:

Dry basis (db)

$$Ash = \frac{Ash\ \%\ on\ air\ dried\ basis}{100 - Moisture\ \%} \times 100 \tag{3.3}$$

$$VM = \frac{Volatile\ matter\ \%\ on\ air\ dried\ basis}{100 - Moisture\ \%} \times 100 \tag{3.4}$$

$$FC = 100 - (Ash + Volatile\ matter) on\ \%\ dry\ basis \tag{3.5}$$

Dry ash free basis (daf)

$$VM = \frac{Volatile\ matter\ \%\ on\ air\ dried\ basis}{100 - (Moisture + Ash)\ \%} \times 100 \tag{3.6}$$

$$FC = 100 - Volatile\ matter\ on\ \%\ daf\ basis \tag{3.7}$$

For ultimate analysis:

Dry basis

$$C = \frac{C\ \%\ on\ air\ dried\ basis}{100 - Moisture\ \%} \times 100 \tag{3.8}$$

$$H = \frac{H\ \%\ on\ air\ dried\ basis}{100 - Moisture\ \%} \times 100 \tag{3.9}$$

$$N = \frac{N\ \%\ on\ air\ dried\ basis}{100 - Moisture\ \%} \times 100 \tag{3.10}$$

$$S = \frac{S\ \%\ on\ air\ dried\ basis}{100 - Moisture\ \%} \times 100 \tag{3.11}$$

$$O = 100 - (C + H + N + S + ash)\ on\ \%\ dry\ basis \tag{3.12}$$

Dry ash free basis

$$C = \frac{C\ \%\ on\ air\ dried\ basis}{100 - (Moisture + ash)\ \%} \times 100 \tag{3.13}$$

$$H = \frac{H\ \%\ on\ air\ dried\ basis}{100 - (Moisture + ash)\ \%} \times 100 \tag{3.14}$$

$$N = \frac{N\ \%\ on\ air\ dried\ basis}{100 - (Moisture + ash)\ \%} \times 100 \tag{3.15}$$

$$S = \frac{S\,\%\,on\ air\ dried\ basis}{100-(Moisture+ash)\,\%} \times 100 \tag{3.16}$$

$$O = 100 - (C + H + N + S)\ on\ \%\ daf\ basis \tag{3.17}$$

3.5 Recording standards for coal analysis

Different analyses and experiments use various preparations of coals and generally report the results as some type of "basis". The understanding of the basis in which the results are reported for correct correlation to parameters or their use in equations is significant. Figure 3.3 illustrates the various recording standards used for different measurements of coal properties. The results of a coal analysis can be calculated on a number of basis differing from each other in the way by which the moisture and ash are treated. Except for data reported on a db, it is important that a suitable moisture content is given in the data report. This would avoid uncertainty and allow a means for conversion of data to other basis.

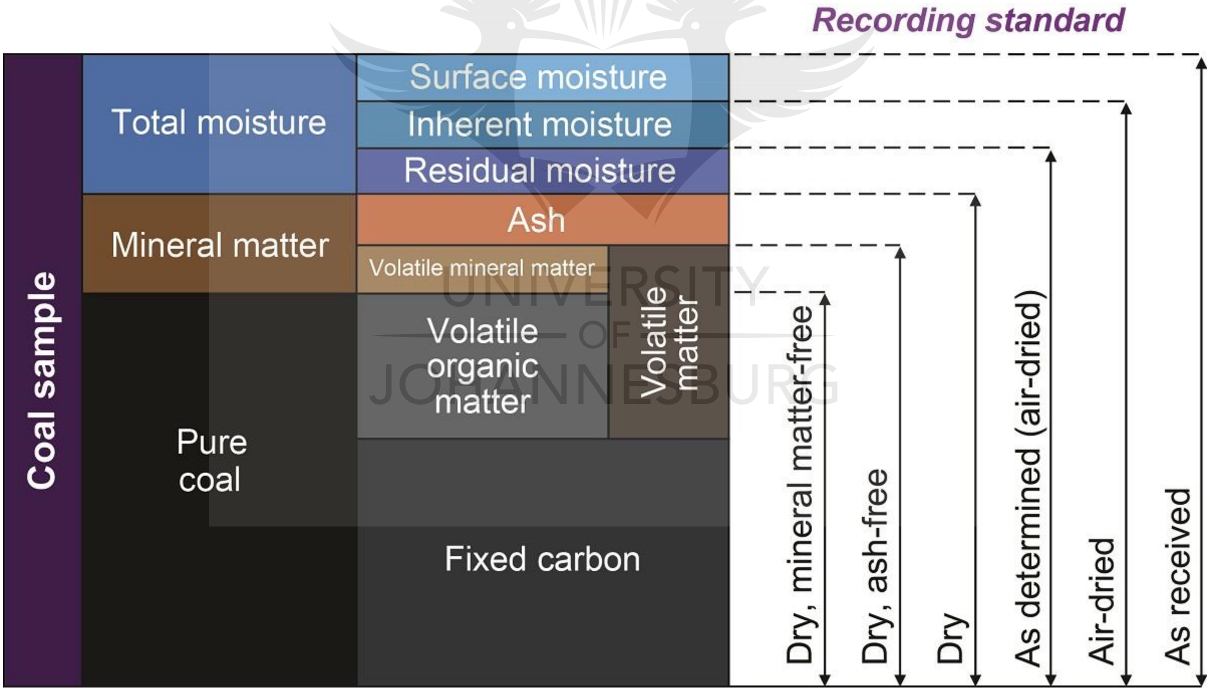


Figure 3.3 Different recording standards are used for different measurements of coal components (modified from Ward, 1984).

3.6 Spontaneous combustion tests

The spontaneous combustion tests used in this study measures the thermal decomposition per minute and heat flow to examine the spontaneous combustion liability of coal. The spontaneous combustion tests, namely; the simple and composite indices are used to determine the spontaneous combustion liability of the samples.

The Wits-Ehac test has been reported for many years to estimate the tendency of coal to undergo spontaneous combustion (Genc et al., 2018; Genc & Cook, 2015; Gouws & Eroglu, 1993; Wade et al., 1987). The main components (a sample cell comprising three same samples of coal and three inert reference materials is kept in an insulated bath consisting of heat transfer oil) of the experimental apparatus for the spontaneous combustion tests are displayed schematically in Figure 3.4. The oil is heated from room temperature at a continuous rate of 1 ^{o}C /min and the heat circulates through the bath. Oxygen in the form of air is supplied to the device containing the coal sample from a compressor. The air supply is controlled with a flowmeter.

Three stages are involved during the use of this apparatus, firstly at Stage 1, an inert material (calcined alumina) temperature is greater than the coal temperature, caused by the cooling impact of the evaporation of the amount of moisture of the coal in question. At Stage II, as the moisture of the coal evaporates, the heating rate of the coal sample increases than that of the inert material. This is due to the affinity of coal to self-heat, which tries to rise to the ambient temperature (the temperature of oil bath). A high heat is released at a region that the line marks the zero base line, defined as the XPT. Stage II is known to be an indicator of self-heating liability. At Stage III, the coal starts to burn. As suggested by Wade et al. (1987), coal with a value less than 3 are referred to as low risk, 3 to 5 are regarded to have medium risk, whereas coal with a liability value higher than 5 are referred to as high risk. The formula for the Wits-Ehac Index is expressed in Equation 3.18. The thermogram in Figure 3.5 shows the different stages and the XPT for a particular coal. Detailed information regarding the Wits-Ehac tests is reported by Onifade (2018) and Genc & Cook (2015).

The modified FCC Index that incorporates the XPT and Stage II Slope used to estimate the liability of coal is expressed in Equation 3.19 (Eroglu, 1992; Gouws & Wade, 1989). According to Feng et al. (1973), coal with a FCC value less than 5 are referred to be low risk, 5 to 10 are regarded to as medium risk, whereas coal with a liability value higher than 10 are referred to as high risk. Also, according to Mahadevan & Ramlu (1985), coal with a XPT greater than 160 ^{o}C have low liability, 140 to 160 ^{o}C have medium liability and coal with XPT between 120 to 140 ^{0}C are referred to as high risk.

Wits-Ehac Index = (Stage II slope/XPT) * 500 (3.18)

Modified FCC Index = (Stage II Slope/XPT) * 1000 (3.19)

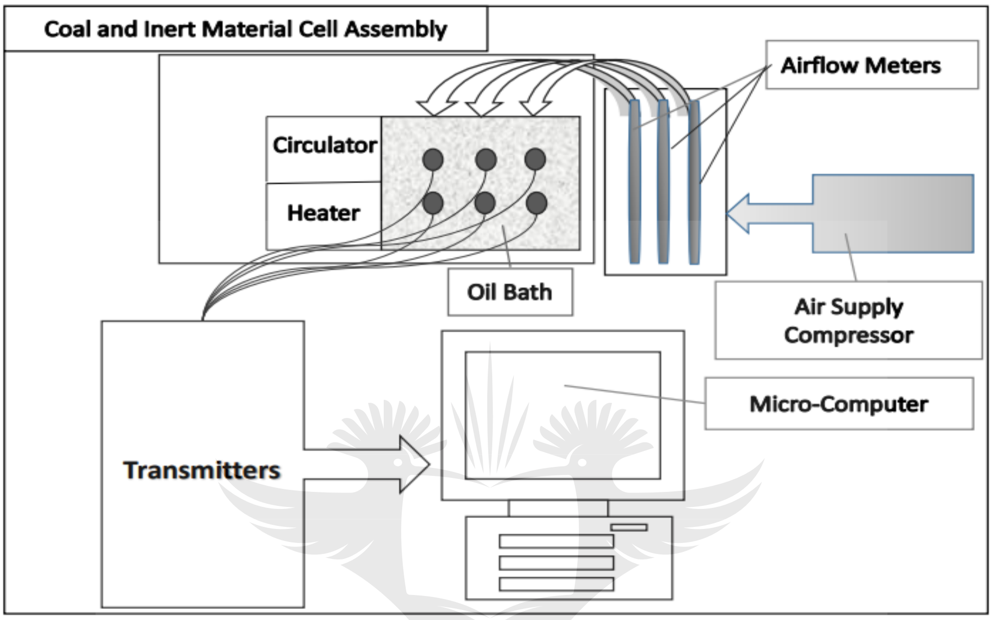


Figure 3.4 Diagram of the Wits-Ehac apparatus (Wade et al., 1987)

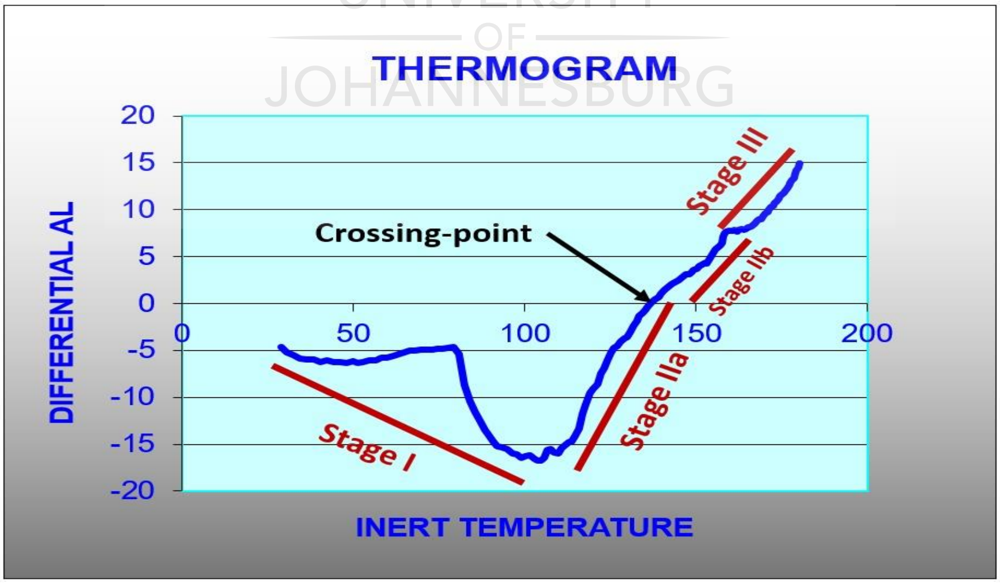


Figure 3.5 A differential thermogram of a coal sample (Wade et al., 1987)

3.7 Calculation of R-Squared in MS Excel

There are several approaches in MS Excel to estimate the R-squared value. The easiest way is to obtain two data sets and use the inbuilt R-squared formula in MS Excel as conducted in this study. Another option is to calculate the coefficient of correlation and then square it.

3.7.1 Regression analysis

The study of regression is a related technique for evaluating the relationships between a variable outcome and one or more risk factors/variables. The outcome variable is also referred to as the dependent variable, and the risk factors are termed the predictors or independent variables. For a regression analysis, the dependent variable is referred to as "Y" and the independent variables are referred to as "X".

Linear regression is a basic method of predictive analysis, which is widely used. The general theory of regression is to analyse two things: (a) if a set of predictor variables does a good job in predicting an outcome (dependent) variable and (b) which variables in specific are major predictors of the outcome variable, and by what means do they show the amount and sign of the better analyses and influence the outcome variable?

The regression analysis is widely used for (a) estimating the strength of predictors, (b) predicting an effect and (c) predicting trend. First, regression may be used to estimate the magnitude of the effect on a dependent variable that the independent variable(s) have. Secondly, it may be used to predict the influence of changes. That is, with a change in one or more independent variables, the regression analysis provides a better understanding of how much the dependent variable changes. Lastly, regression analysis evaluates trends and future values.

3.7.2 Interpretation of linear regression analysis

Linear regression analysis is carried out to estimate the dependent variable with respect to one or more independent variables. The basic equation for regression is:

$$Y = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + e \tag{3.20}$$

where **X** and **Y** are independent and dependent variables and **e** is the error of estimate.

β1 evaluates the influence of X_1 on Y. Similarly, β_2 evaluates the influence of X_2 on Y. The constant term (β_0) determines the value of Y if both X_1 and X_2 are zero. The error of estimate e comprises other parameters that influences Y other than X_1 and X_2.

3.7.3 R-squared values and correlation coefficients

The R-squared values indicates how much variance is described by the independent variable involved in the model of the dependent variable as shown in Equation 3.20. The overall strength of the independent variable and dependent variable can be estimated based on the R-squared value. The correlation coefficient is a statistical tool that evaluates the strength of the relations between the relative movements of two variables. The values vary from -1 and +1. A measured number greater than 1 or less than -1 indicates that there was an error in the correlation measurement. A correlation of -1 indicates a perfect negative correlation between the movements of the two variables, while a correlation of +1 specifies a perfect positive correlation between the movements of the two variables. A correlation of 0 means there is no relation between the movements of the two variables. In this study, the R-squared values and correlation coefficients between the liability indices and the coal properties of 30 coal samples are provided in Tables 5.2 to 5.4. The R-squared values are plotted using MS Excel and are illustrated in Figures 5.1 to 5.12, while the correlation coefficients are calculated from the MS Excel and confirmed with Equation 3.21.

$$r = \frac{n(\Sigma \mathrm{xy}) - (\Sigma \mathrm{x})(\Sigma \mathrm{y})}{\sqrt{\{n(x^2 - (\Sigma x)^2\}\{n\Sigma y^2 - (\Sigma y)^2\,\}}} \tag{3.21}$$

where, **Σ** is the sum, **X** is the independent variable, **Y** is the dependent variable, **XY** is the multiplication of X and Y, $\mathbf{X^2}$ and $\mathbf{Y^2}$ is the square of X and Y, $(\mathbf{\Sigma X})^2$ is the square of sum of X and $(\mathbf{\Sigma Y})^2$ is the square of sum of Y.

3.8 Data Analysis

This study used the coal samples preparation and reported the results as some type of basis. The understanding of the trend of linear relationships of these coal basis for the correct correlation on their liability indices are evaluated with the use of statistical tools. In order to establish the relationship of these coal basis (% ad, % db and % daf) on the liability indices, correlation analysis was conducted on nine independent and four dependent variables.

3.9 Summary

This chapter summarises the experimental approaches used for the analysis of the coal samples. The combination of these tests will enable a detailed understanding of coal recording basis and their relationships with liability indices. The results of the experimental tests are used to evaluate the characteristics known to be an indicator of the liability of coal to self-heat as explained in the subsequent chapter.

4 SPONTANEOUS COMBUSTION AND COAL PROPERTIES ANALYSIS

4.1 Introduction

This chapter presents and explains the results and interprets the findings achieved from the laboratory tests. Results have been discussed in detail and comparisons have been made from the tested samples.

4.2 Spontaneous combustion liability indices

Table 4.1 provides the spontaneous combustion test results obtained from 30 coal samples. The results are grouped into two; namely, the simple liability indices (XPT and Stage II Slope) and composite liability indices (FCC and Wits-Ehac). The XPT's of the coal samples ranged from 95.9 to 192.3 ^{o}C, the Stage II Slope varied from 1.06 to 1.66, the modified FCC Index ranged from 6.8 to 12.1 and the values of the Wits-Ehac Index varied from 3.41 to 6.07.

Table 4.1 Results of spontaneous combustion tests for the coal

	Simple indices		Composite indices	
Sample	**XPT**	**Stage II Slope**	**Wits-Ehac**	**FCC**
WC1	143.6	1.07	3.74	7.50
WC2	146.7	1.13	3.86	7.70
WC3	141.7	1.11	3.90	7.80
WC4	138.2	1.12	4.06	8.10
WC5	144.4	1.20	4.15	8.30
WC6	139.9	1.09	3.87	7.80
WC7	144.7	1.12	3.88	7.70
WC8	145.7	1.26	4.31	8.60
WC9	147.6	1.26	4.26	8.50
WC10	142.7	1.54	5.40	10.80
WC11	137.1	1.56	5.66	11.30
WC12	145.3	1.48	5.11	10.20
WC13	136.4	1.66	6.07	12.10
WC14	143.6	1.61	5.55	11.20
WC15	154.9	1.06	3.41	6.80
WC16	143.5	1.46	5.07	10.10
WC17	141.3	1.16	4.12	8.20
WC18	156.3	1.30	4.16	8.30
WC19	147.4	1.11	3.78	7.50
WC20	192.3	1.31	3.43	6.80
WC21	147.1	1.19	4.04	8.10
WC22	147.9	1.47	4.97	9.90
WC23	140.7	1.59	5.64	11.30
WC24	131.1	1.11	4.24	8.50
WC25	140.3	1.09	3.91	7.80
WC26	135.6	1.34	4.92	9.80
WC27	95.9	1.06	5.53	11.10
WC28	103.9	1.09	5.28	10.50
WC29	113.7	1.06	4.67	9.30
WC30	140.3	1.08	3.86	7.70

4.2.1 Simple liability indices

4.2.1.1 Stage II Slope

The Stage II Slope is the section of the differential curve between the maximum negative differential point and the transition-point between Stage II and Stage III (Banerjee, 1985). The Stage II Slope is an indicator of spontaneous combustion as documented by Eroglu (1992); Wade et al. (1987); and Banerjee (1985). According to Banerjee (1985), the steeper the Stage II Slope, the more liable the coal to undergo self-heat.

The Stage II Slope was lowest for samples WC15, WC27 and WC29 (1.06) and highest for sample WC13 (1.66). No study has developed a risk rating classification for Stage II Slope to evaluate the liability of coal. No study has developed a risk rating classification for Stage II Slope to examine the propensity of coal. However, Stage II Slope has always been incorporated with XPT to develop composite indices (MR, FCC and Wits-Ehac Indices) and to determine the liability of coal (Wade et al., 1987; Feng et al., 1973). The values for the Stage II Slope of the samples are listed in order of their increasing order as seen in Table 4.2.

Table 4.2 Increasing order of Stage II Slope for the coal samples

Risk Rating	**Samples**
Order of increasing	WC29 (1.06), WC27 (1.06), WC1 (1.07), WC30 (1.08), WC6 (1.09), WC25 (1.09), WC28 (1.09), WC3 (1.11), WC19 (1.11), WC24 (1.11), WC4 (1.12), WC7 (1.12), WC2 (1.113), WC17 (1.16), WC29 (1.19), WC5 (1.20), WC8 (1.26), WC9 (1.26), WC18 (1.30), WC20 (1.31), WC26 (1.34), WC16 (1.46), WC22 (1.47), WC12 (1.48), WC10 (1.54), WC11 (1.56), WC23 (1.59), WC14 (1.61) and WC13 (1.66)

4.2.1.2 XPT

The XPT is the temperature whereby the temperature curve of the coal and the curve of the bath temperature cross each other. The XPT is characteristic of a specific coal, as indicated on the DTA thermograms in Figure 3.4 and Figure 4.1. The XPT curves are created by plotting the temperatures of the coal and the inert material against each other. It is reported in many studies that a lower XPT signifies an increase in spontaneous combustion liability (Mahadevan

& Ramlu, 1985). It is not usually easy to assess the XPT from the XPT curves as a result of the tangential approach of the coal and the inert temperature especially for coal with low moisture content (Eroglu, 1992). The XPT was lowest for sample WC27 (95.9 °C) indicating that it has higher spontaneous combustion risk than the other samples. In an earlier study reported by Mahadevan & Ramlu (1985), it was indicated that the coal with XPT values from 120 to 140 °C or below 120 °C has a high spontaneous combustion risk. Hence, the samples with XPT's below 140 ^{0}C (WC4, WC6, WC11, WC13, WC24, WC26, WC27, WC28, WC29 and WC30) are highly prone to spontaneous combustion. Coal samples with XPT between 140 to160 °C (WC1, WC2, WC3, WC5, WC7, WC8, WC9, WC10, WC12, WC14, WC15, WC16, WC17, WC18, WC19, WC21, WC22, WC23 and WC25) are moderately prone to spontaneous combustion, while only coal sample WC20 has an XPT greater than 160 °C. Hence, it is considered to have a low liability to spontaneous combustion. The risk rating classifications of the coal samples based on the XPT results are shown in Table 4.3.

Table 4.3 Risk rating classification based on the XPT for the coal samples

Risk Rating	**Samples**
Low	WC20
Medium	WC1, WC2, WC3, WC5, WC7, WC8, WC9, WC10, WC12, WC14, WC15, WC16, WC17, WC18, WC19, WC21, WC22, WC23 and WC25
High	WC4, WC6, WC11, WC13, WC24, WC26, WC27, WC28, WC29 and WC30

4.2.1.3 *Comparisons of the simple liability indices*

In the assessment of the liability of a coal, the XPT and the stage II slopes of 30 samples can be compared. The liability of coal to self-heating rises with a lower XPT and a steeper Stage II Slope (Banerjee, 1985). Sample WC27 has the lowest XPT, it has the lowest Stage II Slope with coal samples WC15 and WC29 of 30 coal samples. Sample WC15 is the 28th lowest XPT, while sample WC29 is the 3rd lowest XPT of 30 coal samples. These factors independently, are not a reliable index of self-heating. This is similar to the study documented by Eroglu (1992). Some contradictory results are also found. Samples WC15, WC27 and WC29 have the lowest Stage II Slope, while for the XPT, sample WC27 has the 1st lowest XPT, sample WC29 is the 3rd lowest XPT and sample WC15 is the 28th lowest XPT of 30 samples.

4.2.2 Composite liability indices

Composite liability indices are established indices used to examine the propensity of coal to undergo spontaneous combustion. These indices involve two or more characteristics of a DTA coal curve. The major indices are the FCC index established by Feng et al. (1973), the MR index established by Mahadevan & Raju (1985) and the Wits-Ehac Index developed by Wade et al. (1987). In this work, these two indices are used to determine the liability indices of the samples.

4.2.2.1 FCC Index

The modified FCC index is a composite index that comprises of two characteristics of a DTA coal curve as shown in Equation 3.22. This index has been used by Eroglu (1992); Gouws & Wade (1989) and Feng et al. (1973) to estimate the liability of coal. The FCC was lowest for sample WC15 and WC20 (6.8) and highest for sample WC13 (12.1). No samples had FCC values below 5, hence, the FCC considered none of the samples to have low liability towards spontaneous combustion. The coal samples WC1, WC2, WC3, WC4, WC5, WC6, WC7, WC8, WC9, WC15, WC17, WC18, WC19, WC20, WC21, WC22, WC24, WC25, WC26, WC29 and WC30 are moderately prone to spontaneous combustion as they had FCC values from 5 to 10. The coal samples WC10, WC11, WC12, WC13, WC14, WC16, WC23, WC27 and WC28 are highly prone to spontaneous combustion as they had FCC values greater than 10. The risk rating is classified according to the study reported by Feng et al. (1973). The risk rating classifications of the coal samples based on the FCC results are shown in Table 4.4.

Table 4.4 Risk rating classification based on the FCC Index for the coal samples

Risk Rating	**Samples**
Low	-
Medium	WC1, WC2, WC3, WC4, WC5, WC6, WC7, WC8, WC9, WC15, WC17, WC18, WC19, WC20, WC21, WC22, WC24, WC25, WC26, WC29 and WC30
High	WC10, WC11, WC12, WC13, WC14, WC16, WC23, WC27 and WC28

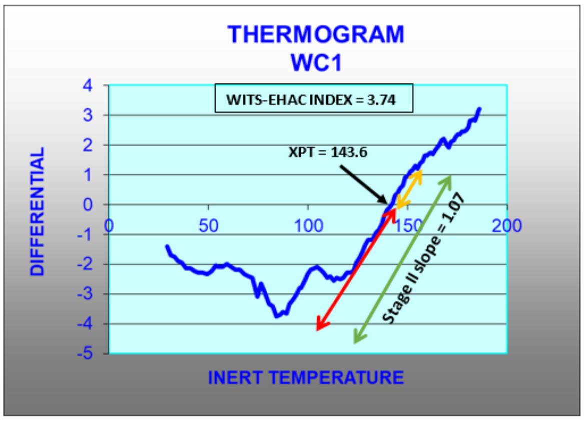
4.2.2.2 Wits-Ehac Index

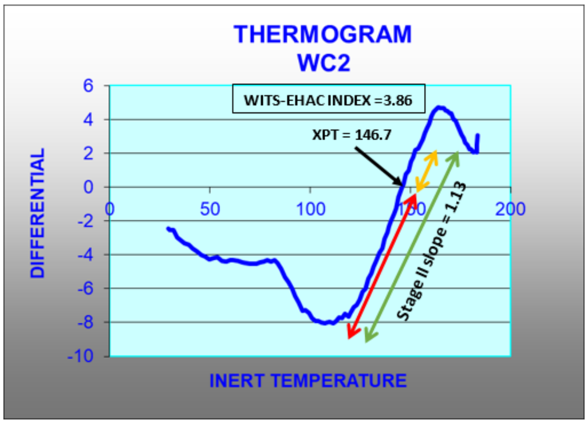
The Wits-Ehac Index has been in existence since 1987 by using the XPT and Stage II Slope to calculate the liability of coal. Coal with a higher Wits-Ehac Index value signifies a higher risk than coal with a lower value of Wits-Ehac Index (Wade et al., 1987). Therefore, coal with a higher value of Wits-Ehac Index will experience self-heating quicker than coal with a lower liability index value. As shown in Figure 3.4 and Figure 4.1, the common form of the coal thermogram with a well-defined, nearly linear, Stage II exothermicity was not always shown in the analysed samples. In some cases, a noticeable displacement appeared at the XPT, while the thermogram gradient decreased at higher temperatures. This characteristic can be described by the decrease in thermodynamic heat transfer rates.

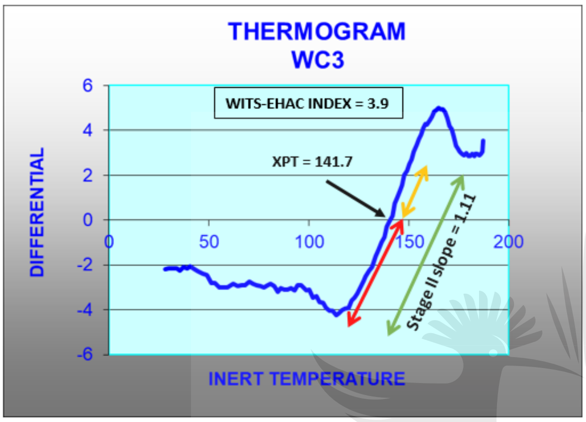
The value of the Wits-Ehac Index was lowest for sample WC15 (3.41) and highest for sample WC13 (6.07). No samples had a Wits-Ehac value below 3, hence, none of the samples has low spontaneous combustion risk. The coal samples WC1, WC2, WC3, WC4, WC5, WC6, WC7, WC8, WC9, WC15, WC17, WC18, WC19, WC20, WC21, WC22, WC24, WC25, WC26, WC29 and WC30 are moderately prone to spontaneous combustion as they had values between 3 and 5. The coal samples WC10, WC11, WC12, WC13, WC14, WC16, WC23, WC27 and WC28 are highly prone to spontaneous combustion as they had values greater than 5. The risk rating classification of the samples based on the Wits-Ehac results are shown in Table 4.5.

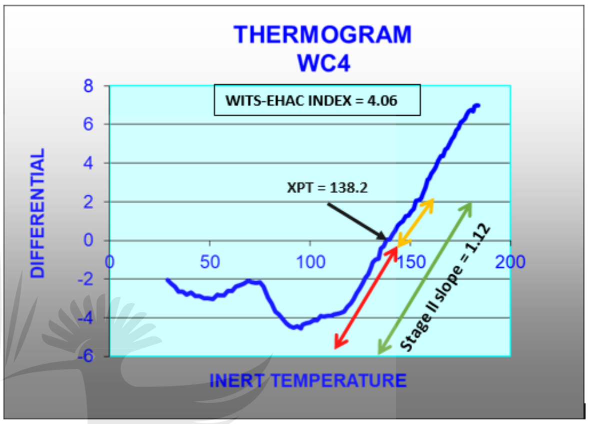
Table 4.5 Risk rating classification based on the Wit-Ehac Index for the samples analysed

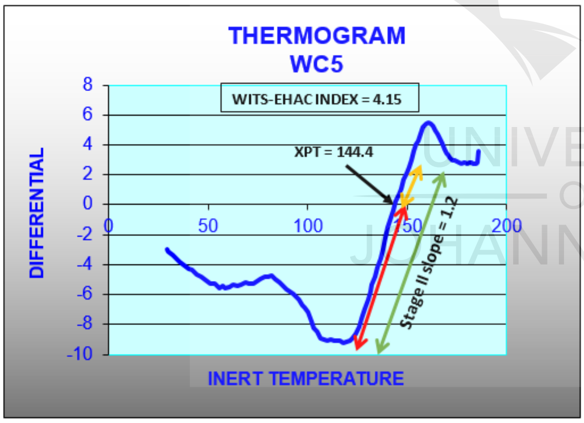
Risk Rating	**Samples**
Low	-
Medium	WC1, WC2, WC3, WC4, WC5, WC6, WC7, WC8, WC9, WC15, WC17, WC18, WC19, WC20, WC21, WC22, WC24, WC25, WC26, WC29 and WC30
High	WC10, WC11, WC12, WC13, WC14, WC16, WC23, WC27 and WC28

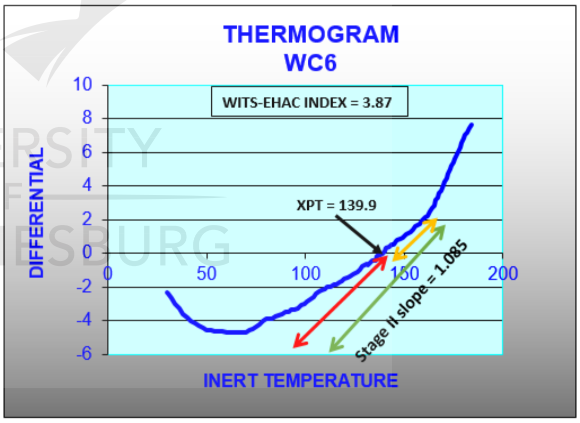

THERMOGRAM
WC1
WITS-EHAC INDEX = 3.74
XPT = 143.6
Stage II slope = 1.07
DIFFERENTIAL
INERT TEMPERATURE
4
3
2
1
0
-1
-2
-3
-4
-5
0
50
100
150
200

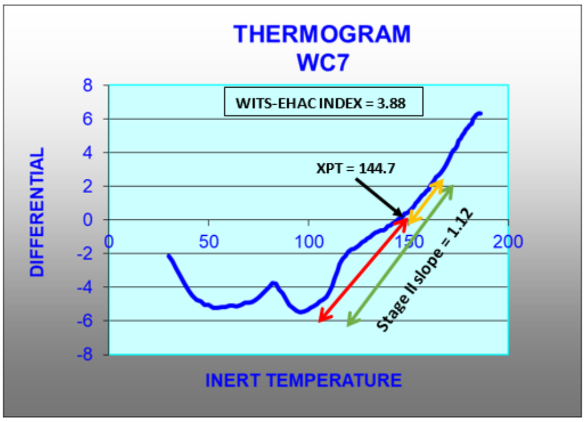

THERMOGRAM
WC2
WITS-EHAC INDEX =3.86
XPT = 146.7
Stage II slope = 1.13
DIFFERENTIAL
INERT TEMPERATURE
6
4
2
0
-2
-4
-6
-8
-10
0
50
100
150
200

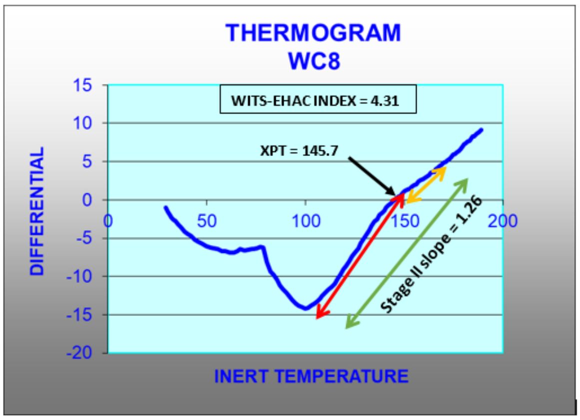

THERMOGRAM
WC3
WITS-EHAC INDEX = 3.9
XPT = 141.7
Stage II slope = 1.11
DIFFERENTIAL
INERT TEMPERATURE
6
4
2
0
-2
-4
-6
0
50
100
150
200


THERMOGRAM
WC4
WITS-EHAC INDEX = 4.06
XPT = 138.2
Stage II slope = 1.12
DIFFERENTIAL
INERT TEMPERATURE
8
6
4
2
0
-2
-4
-6
0
50
100
150
200


THERMOGRAM
WC5
WITS-EHAC INDEX = 4.15
XPT = 144.4
Stage II slope = 1.2
DIFFERENTIAL
INERT TEMPERATURE
8
6
4
2
0
-2
-4
-6
-8
-10
0
50
100
150
200


THERMOGRAM
WC6
WITS-EHAC INDEX = 3.87
XPT = 139.9
Stage II slope = 1.085
DIFFERENTIAL
INERT TEMPERATURE
10
8
6
4
2
0
-2
-4
-6
0
50
100
150
200


THERMOGRAM
WC7
WITS-EHAC INDEX = 3.88
XPT = 144.7
Stage II slope = 1.12
DIFFERENTIAL
INERT TEMPERATURE
8
6
4
2
0
-2
-4
-6
-8
0
50
100
150
200


THERMOGRAM
WC8
WITS-EHAC INDEX = 4.31
XPT = 145.7
Stage II slope = 1.26
DIFFERENTIAL
INERT TEMPERATURE
15
10
5
0
-5
-10
-15
-20
0
50
100
150
200

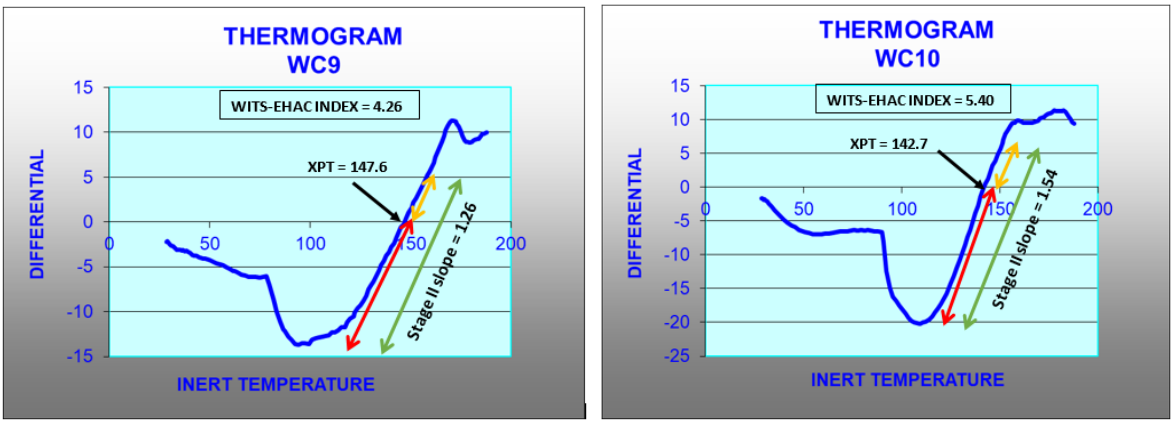


Figure 4.1 Differential analysis thermogram for some of the coal samples

Figure 4.1 illustrates the Stage II Slope and XPT curves which are characteristic of a particular coal, as indicated on the DTA thermograms, where the blue line is the XPT, the red line is the Stage IIA Slope, the yellow line is the Stage IIB Slope and the green line is the Stage II Slope.

4.2.2.3 Comparisons of the composite liability indices

The results for the risk rating indicated that all the samples tested have medium to high liability indices according to the FCC and the Wits-Ehac risk rating classification. Coal samples with a medium to high FCC indices have similar liability indices with the Wits-Ehac Index. However, based on the composite indices, each coal will experience self-heat faster than each other with respect to their liability values. The study confirms that no sample fell into the category of low risk as seen in Table 4.4 and Table 4.5. According to the liability index values, the results obtained from the laboratory test were found to be similar to the field observations. From the evaluation of the different spontaneous combustion tests, it was indicated that each coal varies in their liability indices.

4.3 Coal properties analysis and spontaneous combustion liability indices

The results for the coal properties on % ad, % db and % daf basis for the samples are provided in Table 4.6 to 4.8. The results show that the coal properties vary from one sample to the other irrespective of the coal recording basis used for the analysis. The likely parameters linked to the source of spontaneous combustion based on the selected coal properties examined are discussed below.

Table 4.6 Results from the proximate, elemental and total sulphur (ad, wt. %) for the coal

Sample ID	**M**	**VM**	**A**	**FC**	**S**	**C**	**H**	**N**	**O**
WC1	2.16	18.18	56.78	22.88	1.55	29.40	2.09	0.71	7.31
WC2	4.27	30.27	19.20	46.26	1.49	60.10	3.85	1.47	9.62
WC3	3.43	28.33	17.74	50.50	1.39	64.20	3.88	1.50	7.86
WC4	2.52	25.02	29.78	42.68	0.71	54.90	3.62	1.34	7.13
WC5	3.59	29.27	20.47	46.68	0.71	61.70	4.10	1.35	8.08
WC6	1.22	13.12	52.66	32.99	1.49	38.40	2.08	1.02	3.13
WC7	1.24	19.16	22.20	57.39	2.34	66.00	3.54	1.94	2.74
WC8	3.29	16.41	48.15	32.15	0.90	36.80	2.21	0.96	7.69
WC9	3.26	21.97	16.18	58.59	1.18	67.10	3.57	1.65	7.06
WC10	5.53	27.47	15.04	51.97	1.06	64.20	4.06	1.68	8.43
WC11	5.26	28.54	20.26	45.94	1.43	59.40	3.84	1.46	8.35
WC12	5.05	26.60	19.61	48.74	1.12	60.50	3.71	1.45	8.56
WC13	4.35	28.91	23.54	43.20	1.52	56.40	3.75	1.35	9.09
WC14	4.25	26.71	19.18	49.86	1.10	61.50	3.61	1.35	9.01
WC15	4.05	24.82	28.44	42.69	1.06	52.70	3.31	1.25	9.19
WC16	4.57	28.26	18.83	48.33	1.25	61.70	3.71	1.58	8.36
WC17	4.54	21.78	31.65	42.03	0.21	49.20	2.67	1.05	10.68
WC18	4.71	28.43	17.11	49.75	0.60	62.20	3.44	1.43	10.51
WC19	4.84	28.80	9.05	57.31	0.66	70.40	4.23	1.73	9.09
WC20	4.73	27.22	17.31	50.73	2.54	63.20	3.64	1.36	7.22
WC21	4.28	28.85	14.25	52.62	1.04	66.60	4.08	1.62	8.13
WC22	4.91	31.43	21.46	42.19	4.24	56.50	4.11	1.33	7.45
WC23	5.06	25.50	16.70	52.74	0.82	63.90	3.62	1.68	8.22
WC24	4.19	23.33	30.35	42.13	1.68	51.70	3.14	1.34	7.60
WC25	4.18	21.56	28.08	46.18	0.33	50.80	3.06	1.33	12.22
WC26	2.68	17.87	40.35	39.09	0.48	43.50	2.53	0.99	9.47
WC27	2.30	22.53	51.28	23.90	1.56	32.60	2.25	0.77	9.24
WC28	2.47	22.75	39.77	35.01	0.21	43.30	2.59	0.97	10.69
WC29	1.8	19.61	51.90	26.69	0.39	33.20	2.45	0.80	9.46
WC30	2.54	25.86	25.61	45.99	1.17	58.20	3.39	1.40	7.69

Where M, A, VM, FC, C, H, N, S and O are moisture, ash, volatile matter, fixed carbon, total carbon, hydrogen, nitrogen, total sulphur and oxygen contents.

For the ad, the results are calculated to the moisture condition of the coal after being allowed to air-dry at room temperature as provided in Table 4.6. The moisture content varied from 1.22 to 5.53%, sample WC6 had the lowest, while sample WC10 had the highest moisture content. The volatile matter ranged from 13.12 to 31.43%, sample WC6 had the lowest, while sample WC22 had the highest volatile matter content. Ash ranged from 9.05 to 56.78%, sample WC19 had the lowest, while sample WC1 had the highest ash content. The fixed carbon varied from 22.88 to 58.59%, sample WC1 had the lowest, while sample WC9 had the highest fixed carbon content. The total carbon varied from 29.4 to 70.40%, sample WC1 had the lowest, while sample WC19 had the highest carbon content. The hydrogen ranged from 2.08 to 4.23%, sample WC6 had the lowest, while sample WC19 had the highest hydrogen content. The nitrogen varied from 0.71 to 1.94%, sample WC1 had the lowest, while WC7 had the highest nitrogen content. The oxygen by difference ranged from 2.74 to 12.22%, sample WC7 had the lowest, while sample WC25 had the highest oxygen content. The sulphur content ranged from 0.21 to 4.24%, samples WC17 and WC28 had the lowest, while sample WC22 had the highest sulphur content.

Sample WC10 has the highest moisture content (5.53 %) corresponding to a high liability value. While sample WC6 has the lowest moisture content (1.22 %) corresponding to a moderate liability value as seen in Table 4.1 and Table 4.6. Moisture occurs, more or less, in all types of coal. The value of the moisture contents varied from one sample to the other which can result in variation in their liability values as shown in Table 4.1. With an increase in temperature, the moisture evaporates, takes in some heat and decreases the heat that accumulates within the coal. In addition, moisture creates a mist that encloses the surface of the coal. The moisture can impede the removal of heat and the interaction between coal and oxygen, if the moisture content is high (Kadioglu & Varamaz, 2003; Chen & Stott, 1993).

The volatile matter content for the coal is greater than 15%, except for sample WC6 (13.12%). Samples with an average to high volatile matter content have medium to high liability indices as shown in Table 4.1 and Table 4.6. This is similar with the studies documented by Panigrahi & Sahu, (2004), Banerjee (2000) and Eroglu (1992).

The variations in the ash content support the works reported on the characteristic of South African coal by Van Niekerk et al. (2008) and Falcon & Ham (1988). Samples (WC10, WC11, WC12, WC13, WC14, WC16, WC23, WC27 and WC28) with low ash content correspond to an average to high volatile matter and sulphur content greater than 1% correspond to high liability values. Differences in the quantities of ash may be due to distinctions in the frequency, quantity and course of the inflow of pre-existing minerals and rock into the region during peat formation (Gurdal et al., 2015). In addition, differences in their liability indices could result from the mineral absorbing heat in the samples (Beamish & Blazak, 2005).

Samples WC17 and WC28 had the lowest sulphur content (0.21%), while sample WC22 has the highest sulphur content (4.24%). Three of the samples contain more than 2% sulphur content, WC7 (2.34%), WC20 (2.54%) and WC22 (4.24%). High sulphur content can be correlated with the depositional environment and conditions of the peat, and alkaline depositional environments with concentrated sulfide mineralisation (Beamish & Blazak, 2005). Coal formed in fresh water and a calcium-rich environment is commonly sulphur-rich, which can lead to high quantity of sulphur in some coal. The amount of sulphur in a coal sample usually consists of organic and inorganic sulphur. Most of the inorganic sulphur is FeS_2. The interaction between the forms of sulphur and oxygen, produces H_2S, SO_2, and - SOH, that influences the oxidation of coal. The self-heating characteristics seems to be interrelated with the quantity of carbon, volatile matter, ash content, and sulphur in the coal. The oxygen by difference obtained from each coal does not clearly indicate any influences on the low-temperature oxidation. Therefore, the oxygen content does not seem to determine the coal's liability to self-heat (Onifade, 2018).

Table 4.7 Results from the proximate, elemental and total sulphur (db, wt. %) for the coal

Sample ID	**VM**	**A**	**FC**	**S**	**C**	**H**	**N**	**O**
WC1	18.58	58.03	23.39	1.58	30.05	2.14	0.73	7.47
WC2	31.62	20.06	48.32	1.56	62.78	4.02	1.54	10.04
WC3	29.34	18.37	52.29	1.44	66.48	4.02	1.55	8.14
WC4	25.67	30.55	43.78	0.73	56.32	3.71	1.37	7.86
WC5	30.36	21.23	48.41	0.74	64.00	4.25	1.40	8.38
WC6	13.28	53.31	33.41	1.51	38.87	2.11	1.03	3.17
WC7	19.40	22.48	58.12	2.37	66.83	3.58	1.96	2.78
WC8	16.97	49.79	33.24	0.93	38.05	2.29	0.99	7.95
WC9	22.71	16.73	60.56	1.20	69.36	3.69	1.71	7.31
WC10	29.08	15.92	55.00	1.12	67.96	4.30	1.78	8.92
WC11	30.12	21.38	48.50	1.51	62.70	4.05	1.54	8.82
WC12	28.01	20.65	51.34	1.18	63.72	3.91	1.53	9.01
WC13	30.22	24.61	45.17	1.59	58.96	3.92	1.41	9.51
WC14	27.90	20.03	52.07	1.15	64.23	3.77	1.41	9.41
WC15	25.87	29.64	44.49	1.10	54.92	3.45	1.30	9.59
WC16	29.61	19.73	50.66	1.31	64.65	3.89	1.66	8.76
WC17	22.82	33.16	44.02	0.22	51.54	2.80	1.10	11.18
WC18	29.84	17.96	52.20	0.63	65.27	3.61	1.50	11.03
WC19	30.26	9.51	60.23	0.69	73.98	4.45	1.82	9.55
WC20	28.57	18.17	53.26	2.67	66.34	3.82	1.43	7.57
WC21	30.14	14.89	54.97	1.09	69.58	4.26	1.69	8.49
WC22	33.05	22.57	44.38	4.46	59.42	4.32	1.40	7.83
WC23	26.86	17.59	55.55	0.86	67.31	3.81	1.77	8.66
WC24	24.35	31.68	43.97	1.75	53.96	3.28	1.40	7.93
WC25	22.50	29.30	48.20	0.34	53.02	3.19	1.39	12.76
WC26	18.36	41.46	40.18	0.49	44.70	2.60	1.02	9.73
WC27	23.06	52.48	24.46	1.60	33.37	2.30	0.79	9.46
WC28	23.33	40.78	35.89	0.22	44.40	2.66	0.99	10.95
WC29	19.97	52.85	27.18	0.40	33.81	2.49	0.81	9.64
WC30	26.53	26.28	47.19	1.20	59.72	3.48	1.44	7.88

The volatile matter ranged from 25.03 to 48.54%, sample WC7 had the lowest, while sample WC27 had the highest volatile matter content. The fixed carbon varied from 51.46 to 74.97%, sample WC27 had the lowest, while sample WC7 had the highest fixed carbon content. The carbon varied from 70.23 to 86.21%, sample WC27 had the lowest, while sample WC7 had the highest carbon content. The hydrogen ranged from 4.18 to 7.95%, sample WC17 had the lowest, while sample WC26 had the highest hydrogen content. The nitrogen varied from 1.65 to 2.53%, sample WC17 had the lowest, while WC7 had the highest nitrogen content. The oxygen by difference ranged from 3.58 to 19.9%, sample WC7 had the lowest, while sample WC27 had the highest oxygen content. The sulphur content ranged from 0.33 to 5.76%, samples WC17 and WC22 had the lowest, while sample WC22 had the highest sulphur content.

In the case of the db, the results are calculated to a theoretical base as if there were no moisture in the coal sample. This basis is usually used in testing laboratories due to issues related to determining moisture. From this basis of coal analysis, it was found that there was a slight increase in the percentage fixed carbon, volatile matter, ash, sulphur, nitrogen, carbon, hydrogen and oxygen contents when correlated with the ad basis as seen in Table 4.6 and Table 4.7. The reason has been that all the moisture content was excluded in the samples during analysis.

Table 4.8 Results from the proximate, elemental and total sulphur (daf, wt. %) for the coal

Sample ID	**VM**	**FC**	**S**	**C**	**H**	**N**	**O**
WC1	44.28	55.72	3.77	71.60	5.09	1.73	17.81
WC2	39.55	60.45	1.95	78.53	5.03	1.92	12.57
WC3	35.94	64.06	1.76	81.44	4.92	1.90	9.98
WC4	36.96	63.04	1.05	81.09	5.35	1.98	10.53
WC5	38.54	61.46	0.93	81.25	5.40	1.78	10.64
WC6	28.45	71.55	3.23	83.26	4.51	2.21	6.79
WC7	25.03	74.97	3.06	86.21	4.62	2.53	3.58
WC8	33.77	66.23	1.85	75.78	4.55	1.98	15.84
WC9	27.27	72.73	1.46	83.29	4.43	2.05	8.77
WC10	34.58	65.42	1.33	80.83	5.11	2.12	10.61
WC11	38.32	61.68	1.92	79.75	5.16	1.96	11.21
WC12	35.31	64.69	1.49	80.30	4.92	1.92	11.37
WC13	40.10	59.90	2.11	78.21	5.20	1.87	12.61
WC14	34.88	65.12	1.44	80.30	4.71	1.76	11.79
WC15	42.72	57.28	1.57	78.06	4.90	1.85	13.62
WC16	36.89	63.11	1.63	80.55	4.84	2.06	10.92
WC17	34.13	65.87	0.33	77.10	4.18	1.65	16.74
WC18	36.36	63.64	0.77	79.56	4.40	1.83	13.44
WC19	33.45	66.55	0.77	81.76	4.91	2.01	10.55
WC20	34.92	65.08	3.26	81.07	4.67	1.74	9.26
WC21	35.41	64.59	1.28	81.75	5.01	1.99	9.97
WC22	42.69	57.31	5.76	76.74	5.58	1.81	10.11
WC23	32.59	67.41	1.05	81.67	4.63	2.15	10.50
WC24	35.64	64.36	2.57	78.98	4.80	2.05	11.60
WC25	31.83	68.17	0.49	74.99	4.52	1.96	18.04
WC26	31.37	68.63	0.84	76.62	7.95	1.74	12.85
WC27	48.54	51.46	3.36	70.23	4.85	1.66	19.90
WC28	39.39	60.61	0.36	74.97	4.48	1.68	18.51
WC29	42.35	57.65	0.84	71.71	5.29	1.73	20.43
WC30	35.97	64.03	1.63	81.00	4.72	1.95	10.70

The volatile matter ranged from 13.28 to 33.05%, sample WC6 had the lowest, while sample WC22 had the highest volatile matter content. Ash ranged from 9.50 to 58.03%, sample WC19 had the lowest, while sample WC1 had the highest ash content. The fixed carbon varied from 23.39 to 60.56%, sample WC1 had the lowest, while sample WC9 had the highest fixed carbon content. The carbon varied from 30.05 to 73.98%, sample WC1 had the lowest, while sample WC19 had the highest carbon content. The hydrogen ranged from 2.11 to 4.45%, sample WC6 had the lowest, while sample WC19 had the highest hydrogen content. The nitrogen varied from 0.73 to 1.96%, sample WC1 had the lowest, while WC7 had the highest nitrogen content. The oxygen by difference ranged from 2.78 to 12.76%, sample WC7 had the lowest, while sample WC25 had the highest oxygen content. The sulphur content ranged from 0.22 to 4.46%, samples WC17 and WC28 had the lowest, while sample WC22 had the highest sulphur content.

In the case of daf, the data are calculated to a theoretical base as if there were no moisture or ash in the coal sample. A calculation is made as if the coal was only composed of volatile matter and fixed carbon as seen in Table 4.8. As can be seen in Table 4.8, the percentage of the fixed carbon, volatile matter, nitrogen, hydrogen, sulphur, carbon, and oxygen are higher as compared with the ad and db basis in Table 4.6 and Table 4.7. The reason has been that all the moisture and ash content were excluded in the samples during analysis.

4.4 Summary

In this section, a characterisation involving selected coal properties and the spontaneous combustion tests were presented and interpreted. The results of the different liability tests used in this study were compared and their relationship with coal recording basis were discussed. The liability of selected coal to spontaneous combustion was assessed using simple (XPT and Stage II Slope) and composite liability (FCC and Wits-Ehac) indices. The XPT's of the coal samples ranged from 95.9 to 192.3 ^{0}C, the Stage II Slopes had a range from 1.06 to 1.66, the modified FCC Index ranged from 6.8 to 12.1 and the Wits-Ehac Index varied from 3.41 to 6.07. The analysed coal samples are classified into three categories according to the fire risk rating of the XPT, the FCC Index and the Wits-Ehac Index. The risk rating for the Stage II Slope could not be evaluated because no classification has been established for the index. Some inconsistent results were obtained from both the XPT and Stage II Slope which necessitated the use of the composite indices (FCC and Wits-Ehac). The results obtained from the composite indices confirm a medium to high spontaneous risk for coal samples. In addition, coal samples with medium to high risk for FCC index have similar risk rating with the Wits-Ehac Index.

This study confirmed that the liability of coal to undergo spontaneous combustion and their intrinsic properties vary from one sample point to another. However, the trend of linear relationship between selected coal properties on different basis and their liability indices were not established. Thus, further analysis on these coal properties were carried out to create trend of linear relations with the liability indices using linear regression analysis as explained in the subsequent Chapter.

5 REGRESSION ANALYSIS BETWEEN COAL PROPERTIES AND LIABILITY INDICES

5.1 Introduction

In this section, trends of linear relationships between the liability indices and the coal properties were created by linear regression analysis. This will enable a detailed understanding of coal recording basis and their relationships with liability indices.

5.2 R-squared values (R) and correlation coefficients (r)

The R-squared values and correlation coefficients between the coal properties and liability indices were established by taking intrinsic properties; namely, fixed carbon, moisture, ash, volatile matter, nitrogen, carbon, oxygen, hydrogen and sulphur values as independent variables and the liability indices determined by XPT, Stage II Slope, Wits-Ehac Index and FCC Index as dependent variables. This study made it possible to make regressions of results of the liability indices against the results of the coal analyses. The interpretation of the relations between the coal basis and liability indices depends on the criterion laid down by Onifade (2018) as seen in Table 5.1. The set criterion uses the statistical tools to establish the strength of coal properties on the liability index.

Table 5.1 Criterion laid down to evaluate the strength of linear relationships between coal properties and liability of coal (Onifade, 2018)

Class	Conditions	Interpretations
1	Correlation coefficient/R-squared value from 0.95 to 1 or -0.95 to -1	A perfect positive or negative linear relationship
2	Correlation coefficient/ R-squared value from 0.51 to 0.94 or -0.51 to -0.94	A strong positive or negative linear relationship
3	Correlation coefficient/ R-squared value from 0.25 to 0.50 or -0.25 to -0.50	A moderate positive or negative linear relationship
4	Correlation coefficient/ R-squared value from 0.1 to 0.24 or -0.1 to -2.24	A weak positive or negative linear relationship
5	Correlation coefficient/ R-squared value lower than 0.1 but not zero	A very weak positive or negative linear relationship
6	Correlation coefficient of zero	No linear relationship at all

Table 5.2 R-squared values and correlation coefficients between liability indices and coal properties on ad basis

Independent Variables	**Dependent Variables**							
Coal properties	**XPT**		**Stage II Slope**		**Wits-Ehac Index**		**FCC**	
	R^2	**r**	**R^2**	**r**	**R^2**	**r**	**R^2**	**r**
Moisture	0.1839	0.4288	0.3933	0.6271	0.0727	0.2697	0.0711	0.2666
Volatile matter	0.0781	0.2795	0.1755	0.4189	0.0376	0.1939	0.036	0.1898
Ash content	0.2743	-0.5237	0.2000	-0.4472	0.0007	-0.0273	0.0005	-0.0227
Fixed carbon	0.3024	0.5499	0.1203	0.3468	0.0084	-0.0918	0.0092	-0.0958
Total carbon	0.2888	0.5374	0.1531	0.3913	0.0014	-0.0377	0.0017	-0.0415
Hydrogen	0.1989	0.4460	0.1891	0.4349	0.0042	0.0651	0.0037	0.0607
Nitrogen	0.2025	0.4500	0.1177	0.3431	0.0011	-0.0332	0.0014	-0.0371
Oxygen	0.0469	-0.2166	0.0047	0.0689	0.0530	0.2303	0.0509	0.2255
Total sulphur	0.0979	0.3130	0.0409	0.2023	0.0001	0.0094	0.0004	-0.0010

Table 5.3 R-squared values and correlation coefficients between liability indices and coal properties on db basis

Independent variables	**Dependent variables**							
Coal properties	**XPT**		**Stage II Slope**		**Wits-Ehac Index**		**FCC**	
	R^2	**r**	**R^2**	**r**	**R^2**	**r**	**R^2**	**r**
Volatile matter	0.0871	0.2957	0.1950	0.4416	0.0412	0.2030	0.0396	0.1989
Ash content	0.2728	-0.5223	0.1941	-0.4406	0.0005	-0.0222	0.0003	-0.0177
Fixed carbon	0.3112	0.5579	0.1399	0.3741	0.0051	-0.0717	0.0057	-0.0758
Total carbon	0.2954	0.5435	0.1732	0.4161	0.0003	-0.0186	0.0005	-0.0224
Hydrogen	0.2058	0.4537	0.2094	0.4577	0.0066	0.0811	0.0059	0.0768
Nitrogen	0.2131	0.4616	0.1386	0.3724	0.0002	-0.0138	0.0003	-0.0177
Oxygen	0.0362	-0.1904	0.0086	0.0929	0.0540	0.2323	0.0517	0.2275
Total Sulphur	0.1011	0.3180	0.0463	0.2152	0.0001	0.0008	0.0001	-0.0016

Table 5.4 R-squared values and correlation coefficients between liability indices and coal properties on daf basis

Independent Variables	**Dependent Variables**							
Coal properties	**XPT**		**Stage II Slope**		**Wits-Ehac Index**		**FCC**	
	R^2	**r**	**R^2**	**r**	**R^2**	**r**	**R^2**	**r**
Volatile matter	0.1282	-0.3580	0.0069	-0.0828	0.0479	0.2190	0.0482	0.2200
Fixed carbon	0.1282	0.3580	0.0069	0.0828	0.0479	-0.2190	0.0482	-0.2200
Total carbon	0.2785	0.5278	0.0519	0.2279	0.0388	-0.1970	0.0385	-0.1963
Hydrogen	0.0084	-0.0917	0.0240	0.1549	0.0359	0.1895	0.0344	0.1855
Nitrogen	0.0332	0.1821	0.0043	0.0656	0.0140	-0.1183	0.0138	-0.1177
Oxygen	0.2932	-0.5415	0.0683	-0.2614	0.0292	0.1710	0.0289	0.1700
Total sulphur	0.0212	0.1456	0.0022	0.0468	0.0010	0.0316	0.0008	-0.0281

5.2.1 XPT correlation analysis for coal basis

The set criterion provided in Table 5.1 based on correlation coefficients is used to establish the trend of linear relationship between the coal basis and the liability indices. The R-squared values and correlation coefficients for the liability indices on independent variables are shown in Tables 5.2 to Table 5.4.

As can be seen in Table 5.2 for the ad basis, the value of fixed carbon (0.5499) and carbon (0.5374) indicated a strong positive effect, moisture (0.4288), volatile matter (0.2795), hydrogen (0.4460), nitrogen (0.4500) and sulphur (0.3130) showed a moderate positive effect on the XPT, while ash (-0.5237) showed a strong negative effect and oxygen (-0.2166) showed a weak negative effect on the XPT.

From Table 5.3 for the db basis, the value of fixed carbon (0.5579) and carbon (0.5435) indicated a strong positive effect, hydrogen (0.4537), nitrogen (0.4616), volatile matter (0.2952) and sulphur (0.3180) showed a moderate positive effect on the XPT, while ash (-0.5223) showed a strong negative effect and oxygen (-0.1904) showed a week negative effect on the XPT.

As shown in Table 5.4 for the daf basis, the value of carbon (0.5278) indicated a strong positive effect, fixed carbon (0.3580) showed a moderate positive effect, nitrogen (0.1821) and sulphur (0.1456) showed a weak positive effect on the XPT, whilst oxygen (-0.5415) showed a strong negative effect, volatile matter (-0.3580) showed a moderate negative effect and hydrogen (-0.0917) showed a very weak negative effect on the XPT.

5.2.2 Stage II Slope correlation analysis for coal basis

As can be seen in Table 5.2 for the ad basis, the value of moisture (0.6271) showed a strong positive effect, volatile matter (0.4189), hydrogen (0.4349), nitrogen (0.3431), fixed carbon (0.3468) and carbon (0.3913) indicated a moderate positive effect, sulphur (0.2023) showed a weak positive effect and oxygen (0.0689) showed a very weak positive effect on the Stage II Slope. Only ash (-0.4472) revealed a moderate negative effect on the Stage II Slope.

As seen in Table 5.3 for the db basis, the value of volatile matter (0.4416), carbon (0.4161), hydrogen (0.4577), fixed carbon (0.3741) and nitrogen (0.3724) indicated a moderate positive effect, sulphur (0.2152) showed a weak positive effect and oxygen (0.0929) showed a very weak positive effect on the Stage II Slope. Only the ash (-0.4406) showed a moderate negative effect on the Stage II Slope.

From Table 5.4 for the daf basis, the value of carbon (0.2279) and hydrogen (0.1549) showed a weak positive effect, fixed carbon (0.0828), nitrogen (0.0656) and sulphur (0.0468) showed a very weak positive effect on the Stage II Slope. However, oxygen (-0.2614) showed a moderate negative effect and volatile matter (-0.0828) showed a very weak negative effect on the Stage II Slope.

5.2.3 Wits-Ehac Index correlation analysis for coal basis

As shown in Table 5.2 for the ad basis, the value of moisture (0.2697) showed a moderate positive effect, oxygen (0.2303) and volatile matter (0.1939) showed a weak positive effect and hydrogen (0.0651) showed a very weak positive effect on the Wits-Ehac Index. The fixed carbon (-0.0918), carbon (-0.0377), nitrogen (-0.0332), ash (-0.0273) and sulphur (-0.0094) showed a very weak negative effect on the Wits-Ehac Index.

From Table 5.3 for the db basis, the value of oxygen (0.2323) and volatile matter (0.2030) indicated a weak positive effect, hydrogen (0.0811) showed a very weak positive effect on the Wits-Ehac Index. The ash (-0.0222), fixed carbon (-0.0717), carbon (-0.0186), nitrogen (-0.0138) and sulphur (0.0009) showed a very weak negative effect on the Wits-Ehac Index.

As can be seen in Table 5.4 for the daf basis, the value of volatile matter (0.2190), hydrogen (0.1895) and oxygen (0.1710) indicated a weak positive effect on the Wits-Ehac Index. The fixed carbon (-0.2190), carbon (-0.1970) and nitrogen (0.1183) showed a weak negative effect and sulphur (-0.0316) showed a very weak negative effect on the Wits-Ehac Index.

5.2.4 FCC Index correlation analysis for coal basis

As shown in Table 5.2 for the ad basis, the value of moisture (0.2666) indicated a moderate positive effect, oxygen (0.2255) and volatile matter (0.1898) showed a weak positive effect and hydrogen (0.0607) showed a very weak positive effect on the FCC Index. Moreover, ash (-0.0227), fixed carbon (-0.0958), carbon (-0.0415), nitrogen (-0.0371) and sulphur (-0.0010) showed a very weak negative effect on the FCC Index.

From Table 5.3 for the db basis, the value of volatile matter (0.1989), oxygen (0.2275) indicated a weak positive effect and hydrogen (0.0768) showed a very weak positive effect on the FCC Index. In addition, ash (-0.0177), fixed carbon (-0.0758), carbon (-0.0224), nitrogen (-0.0177) and sulphur (-0.0016) showed a very weak negative effect on the FCC Index.

From Table 5.4 for the daf basis, the value of volatile matter (0.2200), hydrogen (0.1855) and oxygen (0.1700) indicated a weak positive effect on the FCC Index. Moreover, fixed carbon (-0.2200), carbon (-0.1963) and nitrogen (0.1177) showed a weak negative effect and sulphur (-0.0281) showed a very weak negative effect on the FCC Index.

The XPT shows a higher correlation coefficients with the coal properties on ad basis than the db and daf basis, while the db basis indicates a higher correlation coefficients with the coal properties and XPT than the daf.

The Stage II Slope shows a higher correlation coefficients with coal properties on the ad basis than the db and daf basis, while the db indicate a higher correlation coefficients with the coal properties and Stage II Slope than the daf.

The Wits-Ehac Index shows a higher correlation coefficients with coal properties on the ad basis than the db and daf basis, while the db indicate a higher correlation coefficients with the coal properties and the Wits-Ehac Index than the daf.

The FCC Index shows a higher correlation coefficients with coal properties on the ad basis than the db and daf basis, while the db indicate a higher correlation coefficients with the coal properties and FCC Index than the daf.

From the correlation analysis, the ad basis indicates a higher correlation coefficients than the db and daf basis for the XPT and FCC Index, while the daf shows a higher correlation coefficients than the ad and db basis for the Wits-Ehac Index. However, the trend of linear relationships of this coal properties differs from one liability index to the other. From all the liability indices examined with respect to coal properties (ad, db and daf basis), the XPT followed by the Stage II Slope show a higher correlation coefficients than the other liability indices.

The analysis of the dependent and independent variable sets for % ad, % db and % db basis (Moisture/ XPT, Stage II Slope, Wits-Ehac Index and FCC, volatile matter/ XPT, Stage II Slope, FCC and Wits-Ehac Index, ash/ XPT, Stage II Slope, FCC and Wits-Ehac Index, carbon/XPT, Stage II Slope, FCC and Wits-Ehac Index, sulphur/ XPT, Stage II Slope, FCC and Wits-Ehac Index, etc.) showed identical trends in some cases, i.e. the XPT and Stage II Slope trend increases with increasing moisture, volatile matter, fixed carbon, carbon, hydrogen, nitrogen and sulphur and decreases with increasing ash on ad and db basis. While XPT decreases with increasing oxygen on ad and db basis, Stage II Slope increases with increasing oxygen on ad basis and increases with increasing oxygen on db basis (Figures 5.1, 5.2, 5.5 and 5.6). The FCC and Wits-Ehac Index trend increases with increasing moisture, volatile matter, hydrogen and oxygen and decreases with increasing ash, fixed carbon, carbon and nitrogen on ad basis and db basis (Figures 5.3, 5.4, 5.7 and 5.8). As the Wits-Ehac Index increases with increasing sulphur on an ad basis, it decreases with increasing sulphur on db basis, while the FCC decreases with increasing sulphur on ad and db basis. For the daf basis, the XPT increases with increasing fixed carbon, carbon, nitrogen and sulphur and decreases with increasing volatile matter, hydrogen, oxygen, while the Stage II Slope trend increases with increasing fixed carbon, carbon, hydrogen, nitrogen and sulphur and decreases with increasing oxygen (Figure 5.9 and Figure 5.10). The Wits-Ehac Index and FCC trend increases with the increasing volatile matter, hydrogen and oxygen and decreases with increasing fixed carbon, carbon, nitrogen and sulphur on daf basis (Figure 5.11 and Figure 5.12).

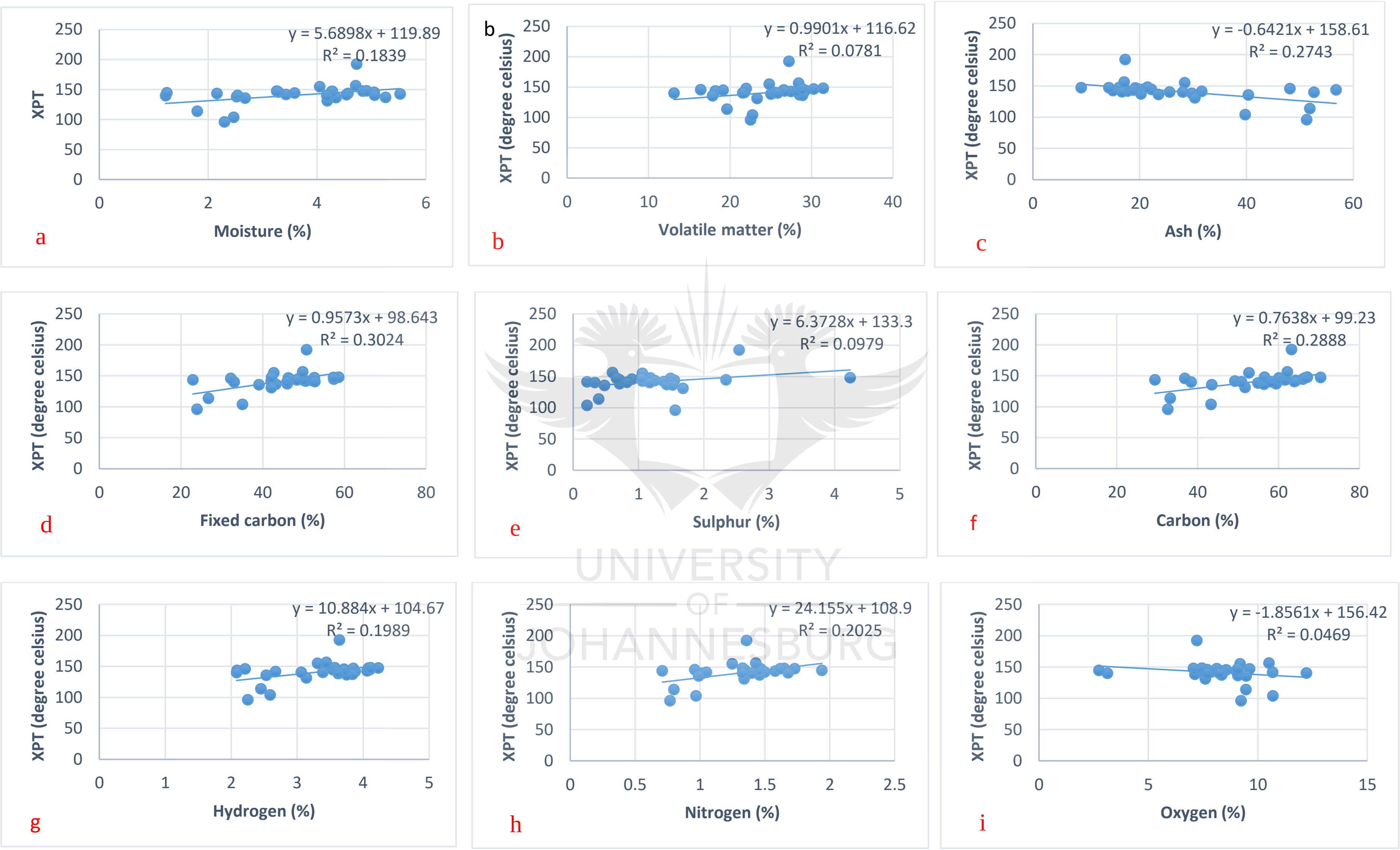


Figure 5.1 R-Squared values between XPT and coal properties (ad)

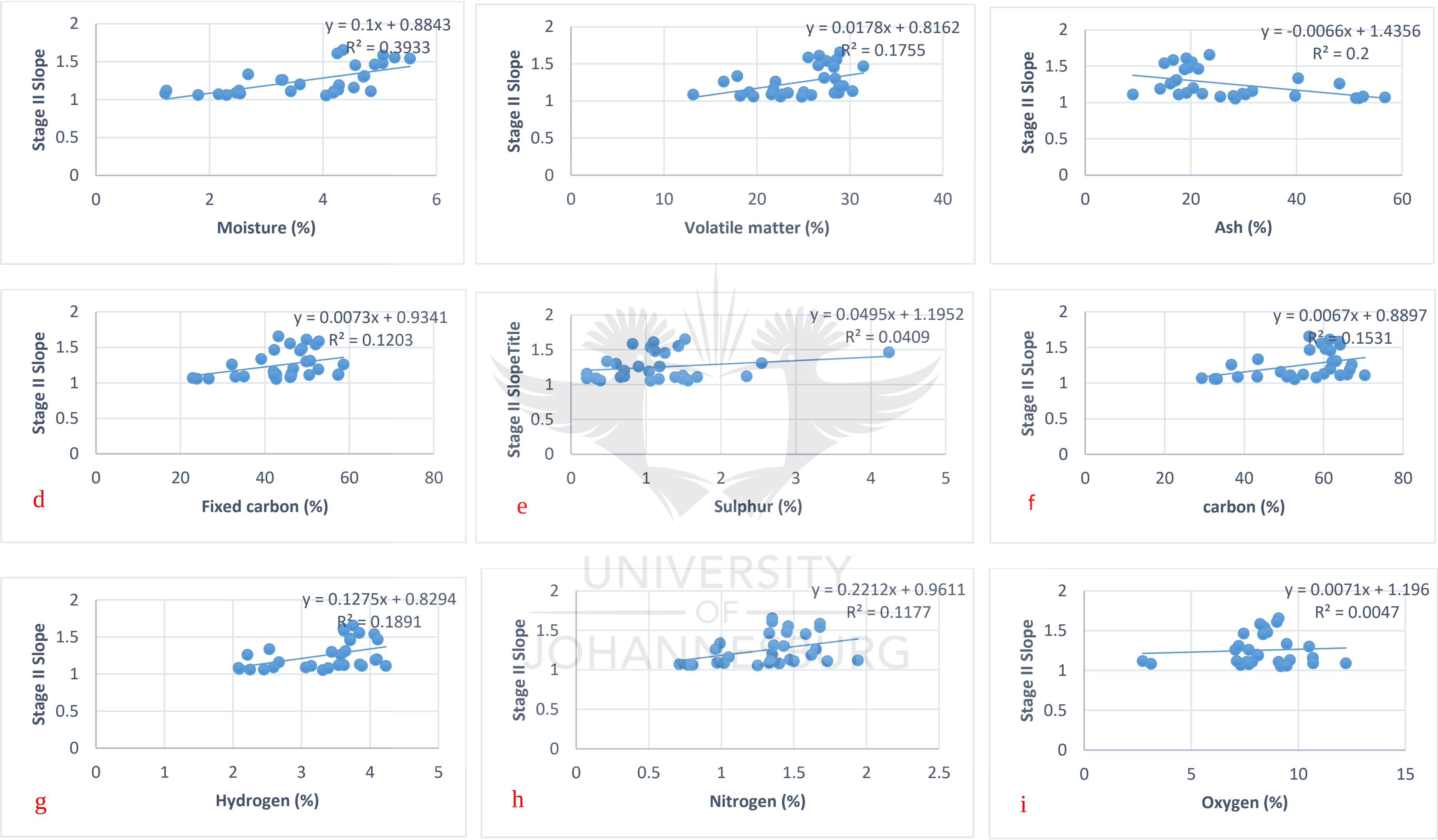


Figure 5.2 R-Squared values between Stage II Slope and coal properties (ad)

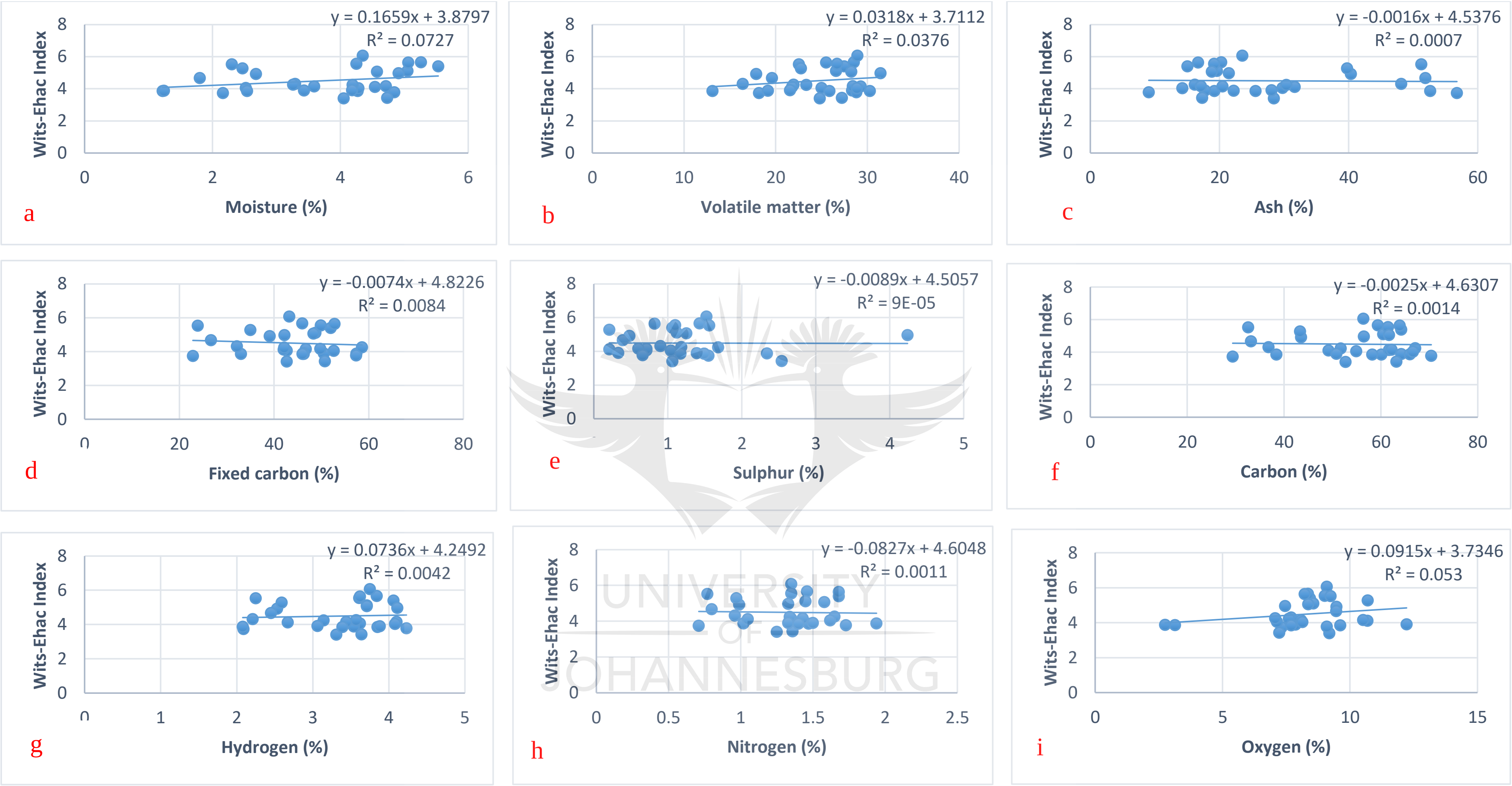


Figure 5.3 R-Squared values between Wits-Ehac Index and coal properties (ad)

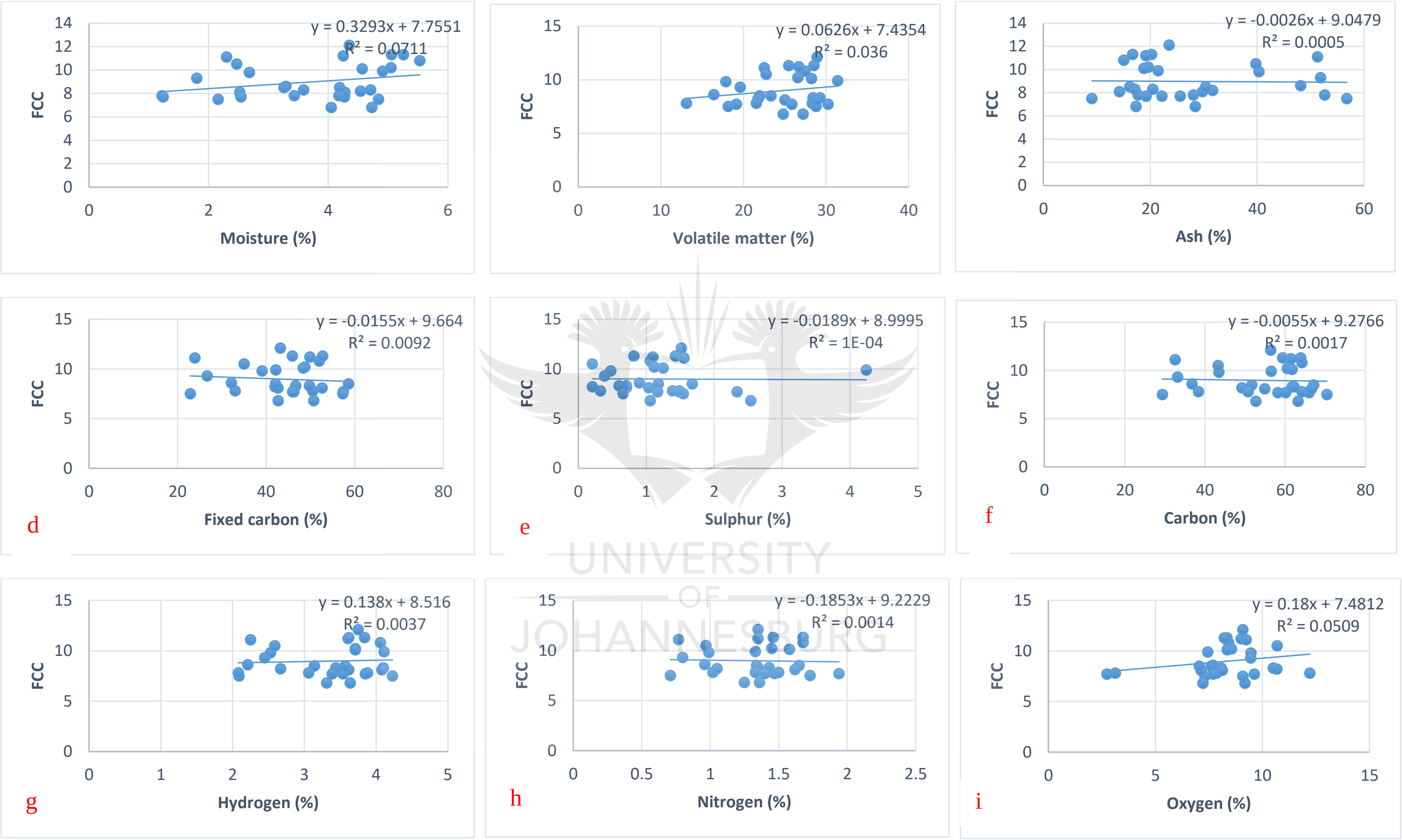


Figure 5.4 R-Squared values between FCC Index and coal properties (ad)

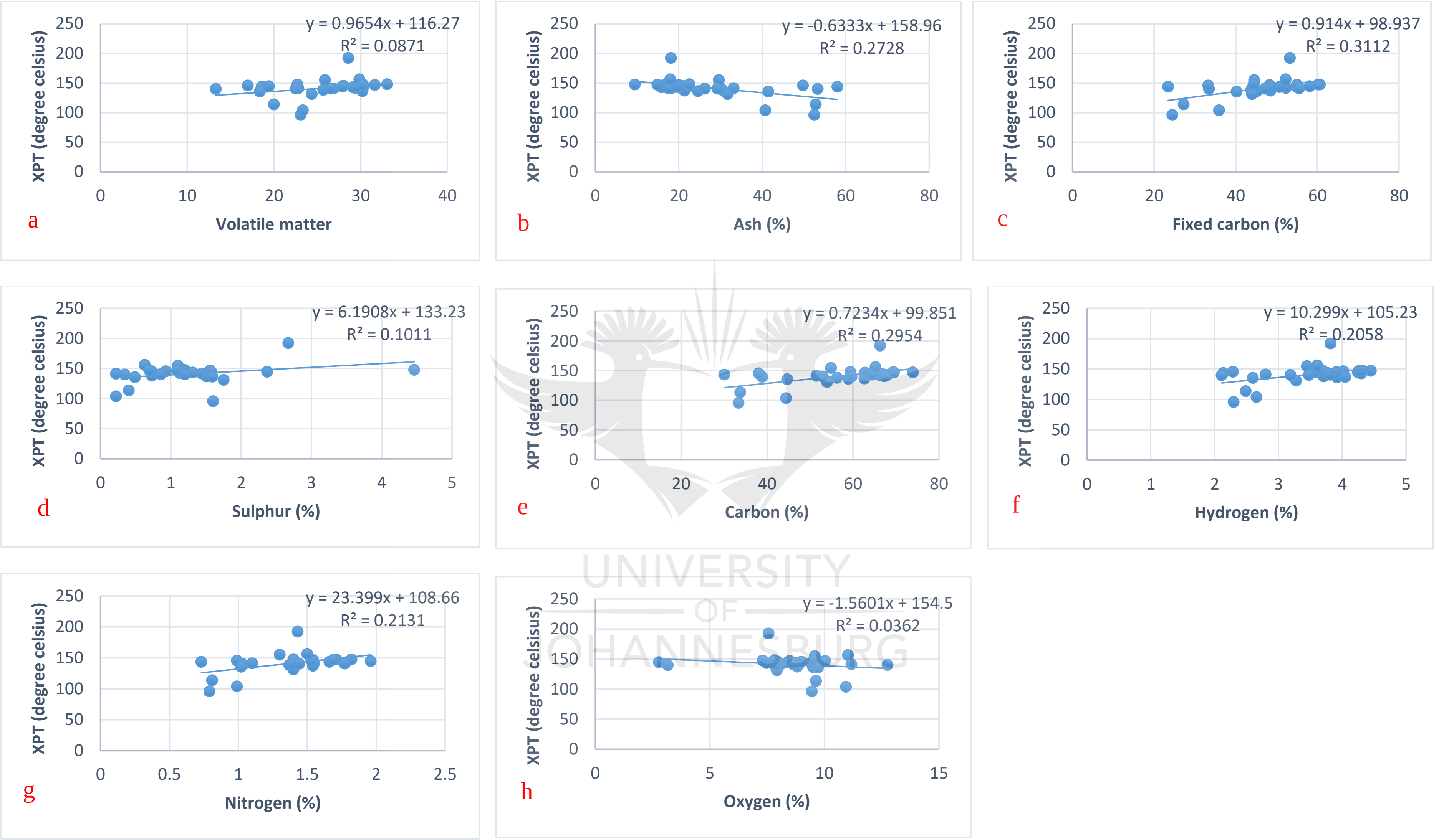


Figure 5.5 R-Squared values between XPT and coal properties (db)

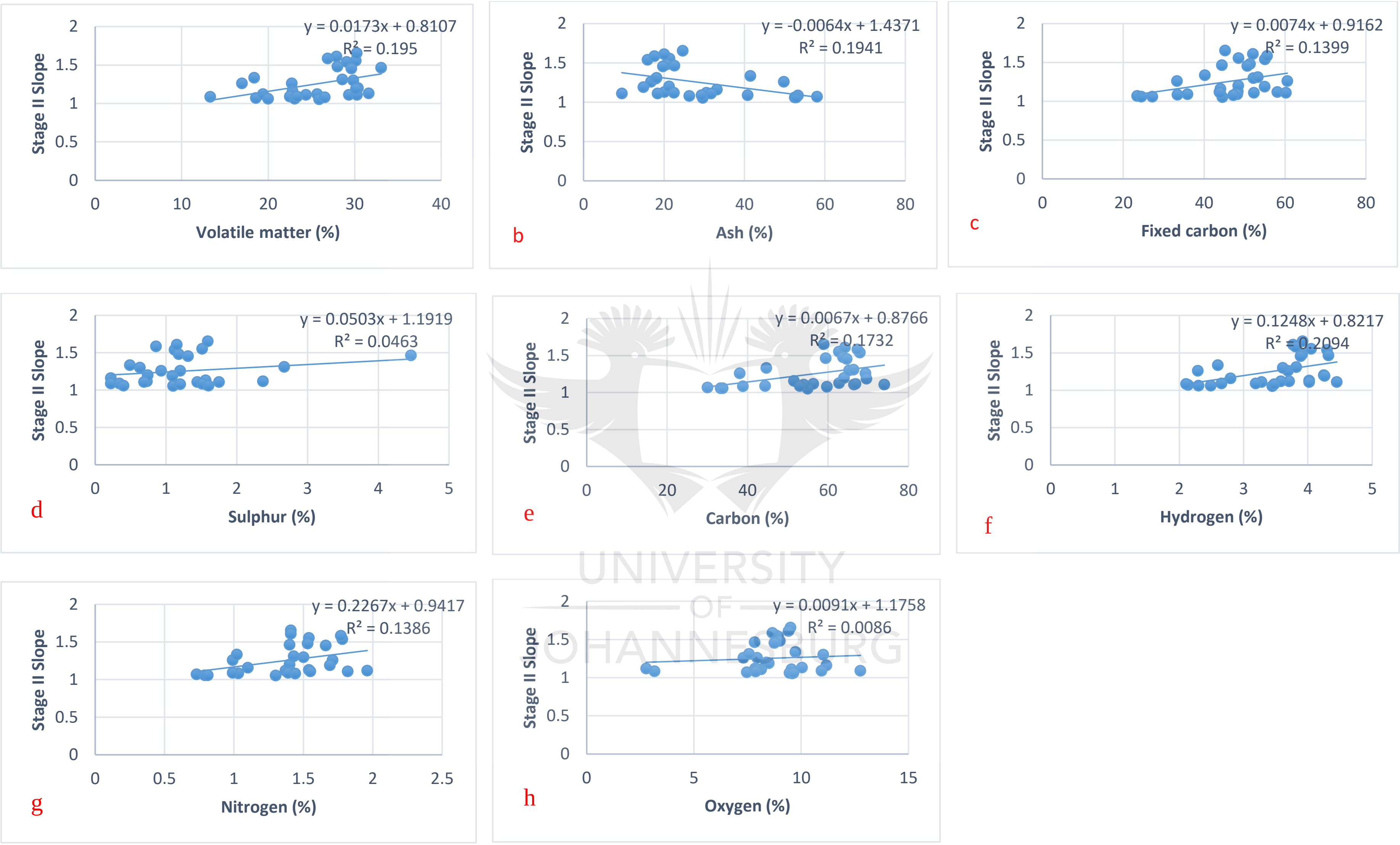


Figure 5.6 R-Squared values between Stage II Slope and coal properties (db)

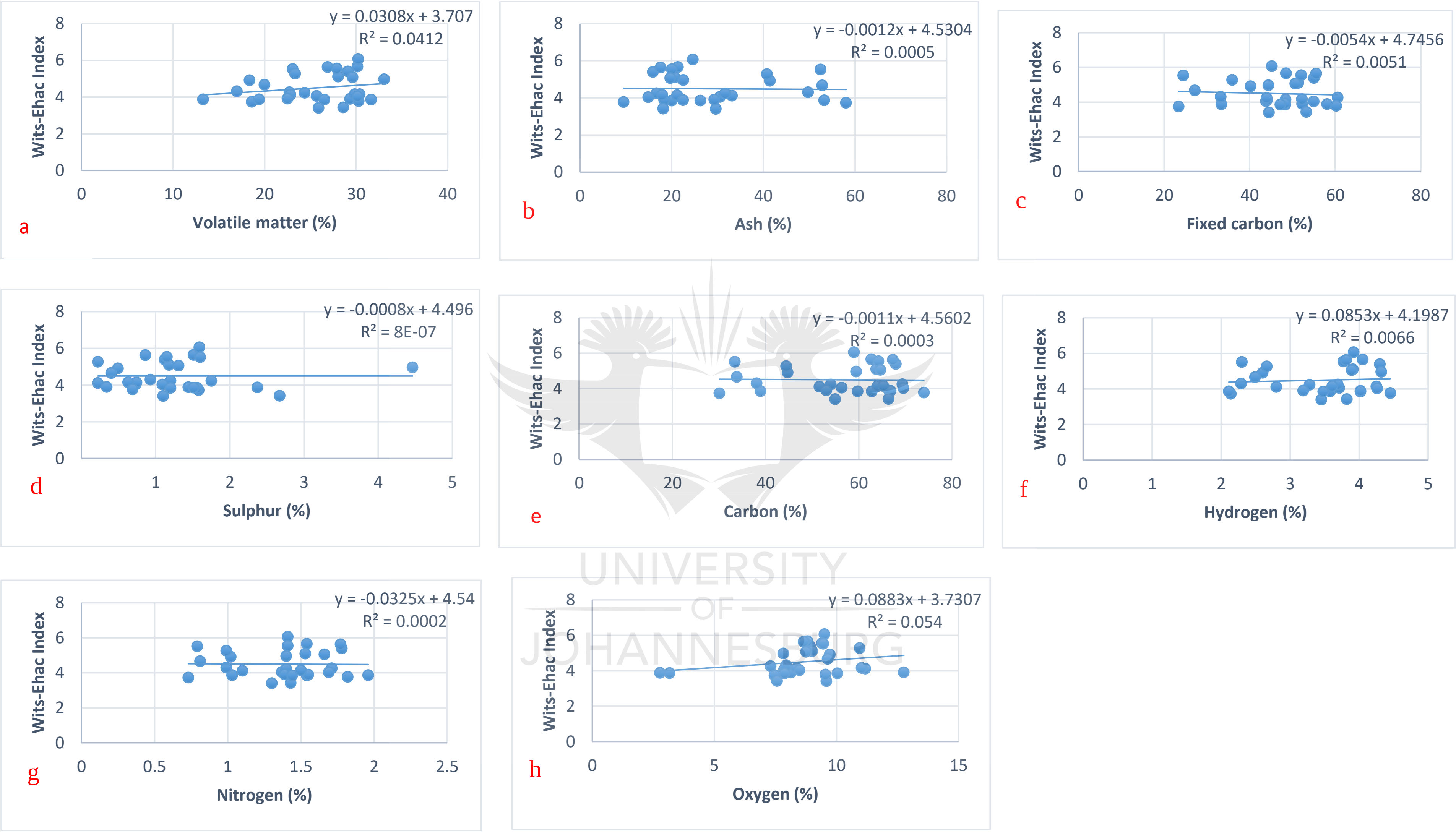


Figure 5.7 R-Squared values between Wits-Ehac Index and coal properties (db)

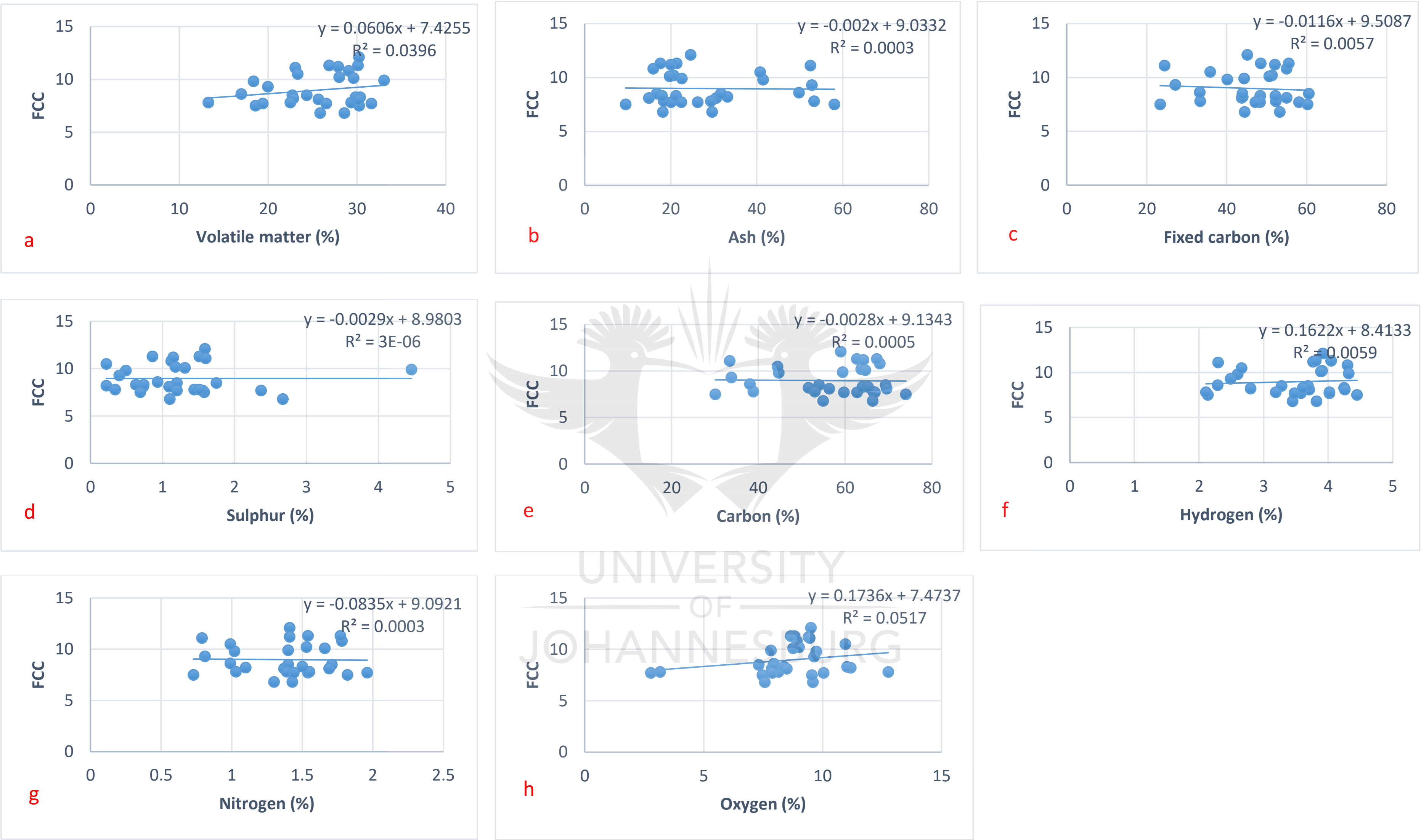


Figure 5.8 R-Squared values between FCC Index and coal properties (db)

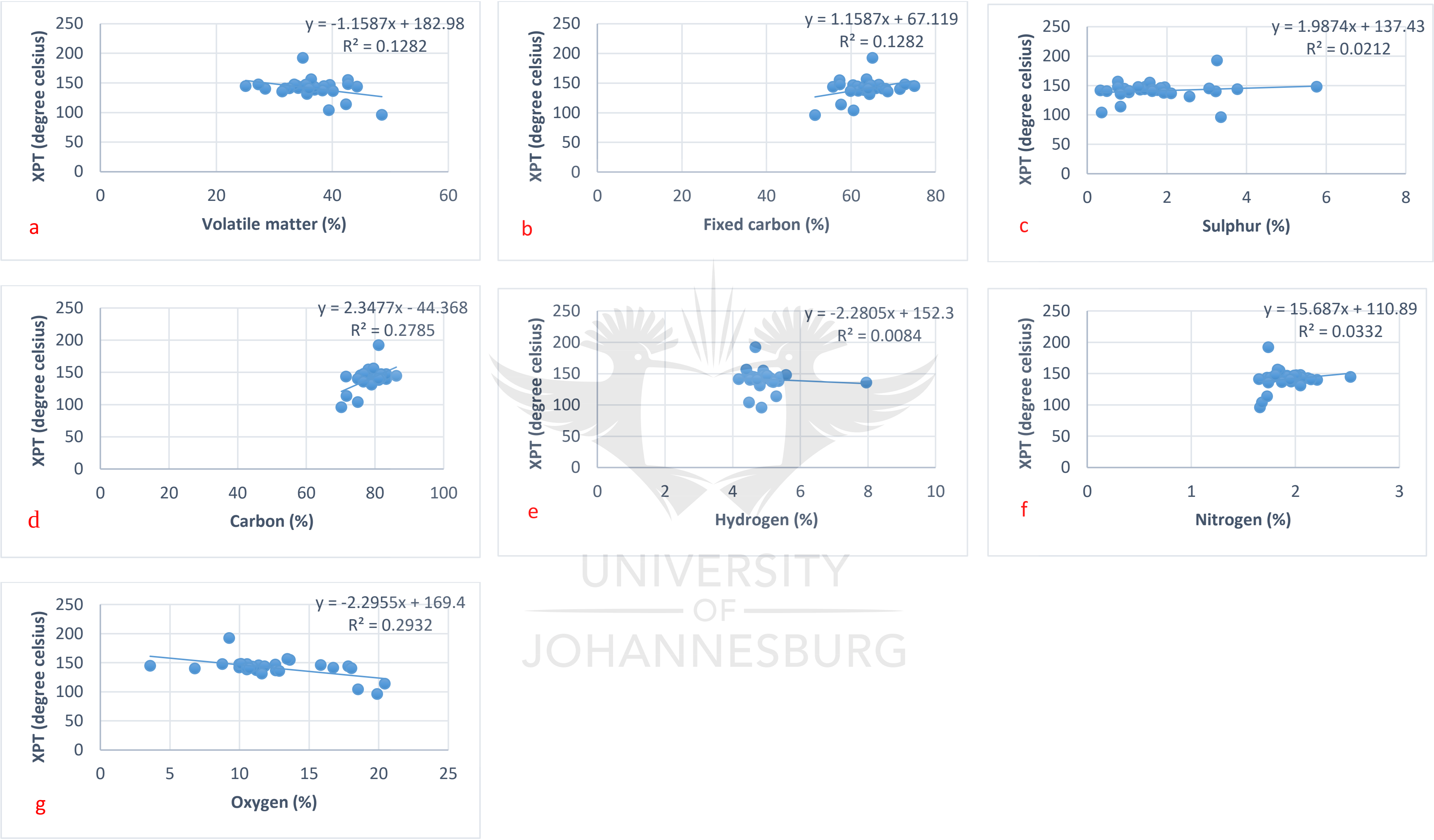


Figure 5.9 R-Squared values between XPT and coal properties (daf)

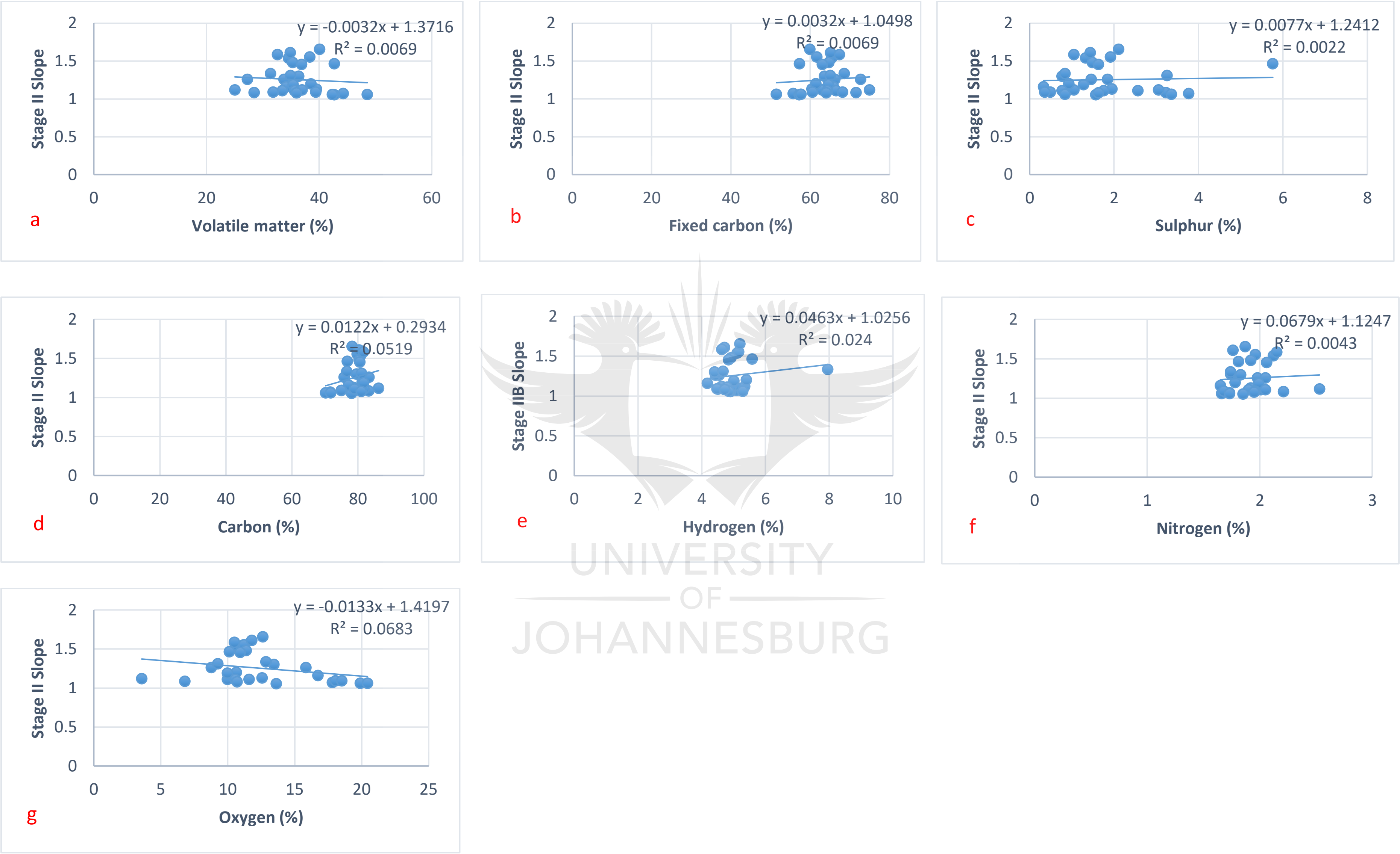


Figure 5.10 R-Squared values between Stage II Slope and coal properties (daf)

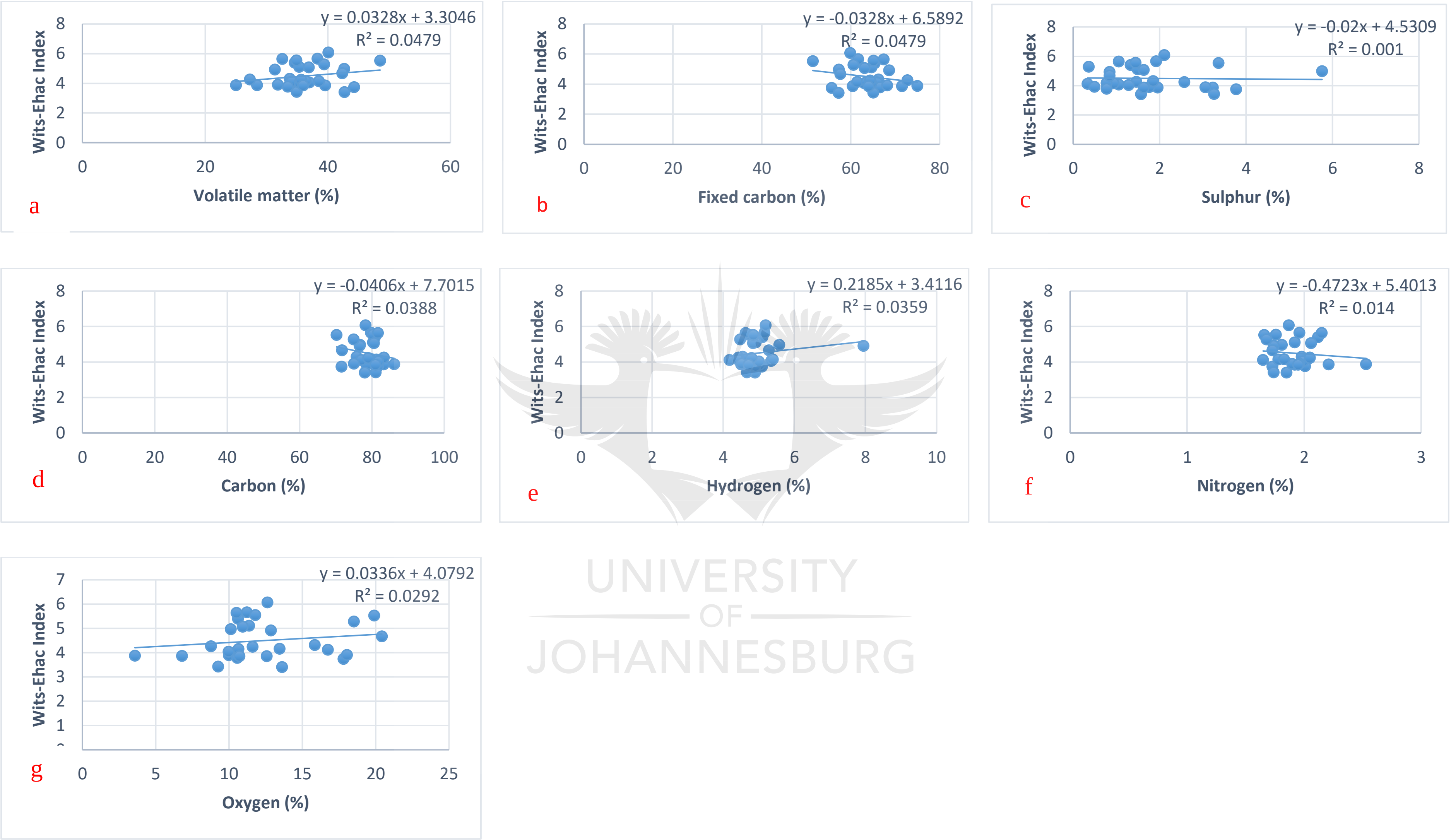


Figure 5.11 R-Squared values between Wits-Ehac Index and coal properties (daf)

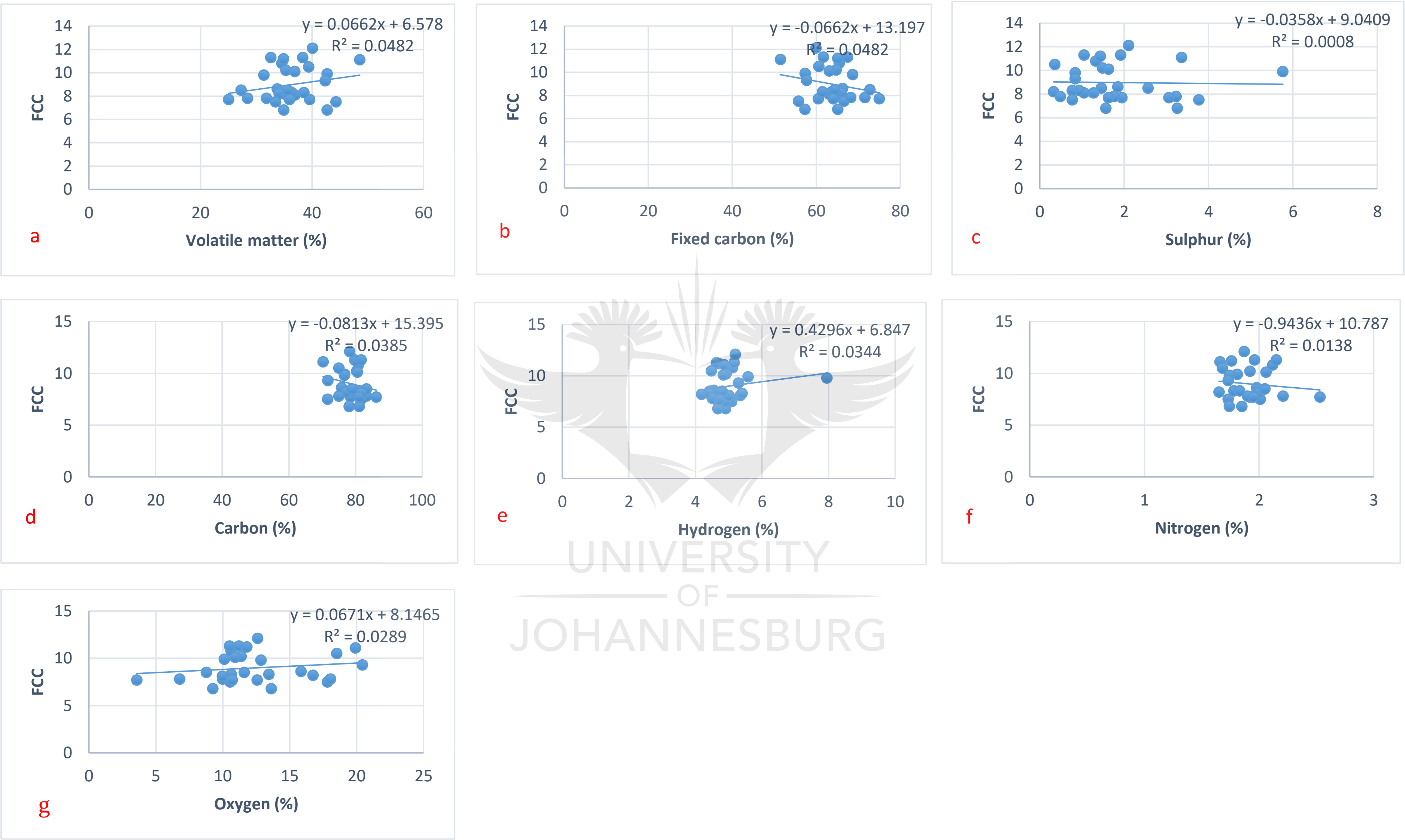


Figure 5.12 R-Squared values between FCC Index and coal properties (daf)

5.3 Summary

This study evaluated and established the trend of linear relationships between the coal properties and different liability indices of coal. The correlation analysis indicated variations between the trend of linear relationships between the dependence of the liability indices and the coal properties on different coal recording basis. From the correlation analysis, the ad basis indicates a higher correlation coefficients than the db and daf basis for the XPT and FCC Index, while the daf shows a higher correlation coefficients than the ad and db basis for the Wits-Ehac Index. Hence, the variations in trend of these coal properties differs from one liability index to the next.

6 CONCLUSIONS AND RECOMMENDATIONS

6.1 Introduction

Spontaneous combustion of coal is a challenge that influence the environment, valuable and non-renewable natural resources, the economy, life, property, and human health. The self-heating characteristics of coal is a sign of their liability to undergo spontaneous combustion. Most of the coal fires in coalfields usually results from spontaneous combustion despite the different control measures in place. The liability of coal to undergo self-heating differs in various coal seams. Researchers internationally have examined the liability of coal to undergo spontaneous combustion by using several approaches. Coal reacts differently to self-heat when subjected to similar atmospheric effects due to variations in the coal organic and inorganic constituents. Some characteristics of coal thermograms thought to be indicative of spontaneous combustion liability such as XPT, Stage II Slope and low transition temperatures from Stage II to Stage III referred to as simple indices, are identified to be inconsistent in the assessment of the liability of coal to undergo spontaneous combustion. The theory of composite indices, comprising of two or more simple indices were evaluated and compared with the results obtained from the simple indices to control these difficulties.

Considering the values obtained from each of the liability indices, coal has been grouped based on their liability to undergo spontaneous combustion as high, moderate and low liability in most cases. However, out of these testing methods, no comparisons have been established from the results obtained from each approach to validate the liability of different coal to self-heat. This research aimed to study different spontaneous combustion tests methods and coal recording basis and examine the trend of linear relationships between them using regression analysis. The key goals and methodologies needed have all been successfully completed in order to provide the answers to the research question in section 1.6. A general overview of the specific findings answering the questions of the study is given as follows:

6.2 Key Findings of the Research

- The liability of selected coal to spontaneous combustion was assessed using simple and composite liability indices and classified into three categories (low/medium/high) according to the fire risk rating of the XPT, the FCC Index and the Wits-Ehac Index.

The risk rating for the Stage II Slope could not be evaluated because no classification has been established for the index;

- Some inconsistent results were obtained from both the XPT and Stage II Slope which necessitated the use of the composite indices (FCC and Wits-Ehac). The results obtained from the composite indices confirm a medium to high spontaneous risk for coal samples. In addition, coal samples with medium to high risk for the FCC index have similar risk ratings with the Wits-Ehac Index;
- The results generated from the spontaneous combustion tests were found to be similar with the field observations according to the FCC and the Wits-Ehac Indices;
- The coal properties were experimentally determined by the ASTM Standards and were found to vary from one sample to another. Additionally, it was observed that samples with moderate to high moisture, volatile matter, carbon and low ash content correspond to a high liability index;
- From the correlation analysis, the ad basis indicates a higher correlation coefficients than the db and daf basis for the XPT and FCC Index, while the daf shows a higher correlation coefficients than the ad and db basis for the Wits-Ehac Index. Hence, the trend of linear relationships of these coal properties differs from one liability index to the next; and
- From all the liability indices examined with respect to coal properties (ad, db and daf basis), the XPT show a higher correlation coefficients followed by the Stage II Slope than the Wits-Ehac Index and FCC Index.

6.3 Research Limitation

- This work has had a range of limitations. A number of studies on spontaneous combustion has been conducted, but there is no single research that satisfactorily covers all the various aspects of this phenomenon. The key limitations for this study are related with the number, type and conditions of the samples known to be susceptible to spontaneous combustion; and
- The scope of this research has been restricted by the limited time and access to the facilities for the various laboratory tests.

6.4 Recommendations for Future Research Work

There are ideas for further work in this section. The recommendations are as follows based on the findings, discussions and results obtained so far:

- It would be most useful to the industry/academics if further study can investigate and compare the results of other spontaneous combustion tests methods such as wet oxidation potential, thermogravimetry analysis, differential scanning calorimetry, Olpinski tests, R70, flammability temperature, etc. in order to establish a standard liability index for the prediction of spontaneous combustion of coal; and
- The influence of the gas content produced during self-heating should be examined with respect to spontaneous combustion.

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8 APPENDICES

8.1 Summary of paper abstracts on the research

The publications listed below have originated from this thesis so far:

8.1.1 Paper 1

Parts of Chapter 4 were compiled and published as the paper: **Abisola Risiwat Gbadamosi**, Moshood Onifade, Bekir Genc & Steven Rupprecht. Spontaneous combustion liability indices of coal. Journal of Combustion Science and Technology (Accepted). The following is an extract of the paper abstract:

"Abstract

The self-heating of coal due to oxidation potentially leading to its ignition is called "spontaneous combustion". The liability of coal to undergo spontaneous combustion for selected coal obtained from the Witbank Coalfields was examined using different spontaneous combustion tests. The simple indices [crossing point temperature (XPT) and Stage II Slope] obtained from differential thermal analysis and composite indices [FCC (Feng, Chakravorty, Cochrane) and Wits-Ehac Indices] were used to examine the liabilities of 30 coal samples. Two characteristics produced from a differential thermogram (XPT and Stage II Slope) are evaluated as indicative of the liability of coal to self-heat. It was found that these simple indices provide inconsistent predictions of spontaneous combustion liability, while the composite liability indices derived from one or more of simple indices provide more reliable results than the simple indices. The study compared various spontaneous combustion tests to assess the liability of coal to undergo spontaneous combustion. The coal properties of the samples have also been found to have impacts on liability indices and vary from one sample point to another.

Keywords**: Coal; coalfield; crossing point temperature; liability indices; intrinsic properties and Wits-Ehac Index**".

8.1.2 Paper 2

Parts of Chapter 4 and 5 were compiled and published as the paper: **Abisola Risiwat Gbadamosi**, Moshood Onifade, Bekir Genc & Steven Rupprecht. Analysis of coal recording standard/basis on spontaneous combustion liability indices. International Journal of Mining Science and Technology (In Press). The following is an extract of the paper abstract:

"Abstract

The characterization of the physical and chemical properties of coal on a standard provides an understanding of its characteristics towards spontaneous combustion. The trend of linear relationships between coal recording standards [% air dried (ad), % dry (db) and % dry ash free (daf) basis] of 30 selected coal samples from the Witbank coalfields and spontaneous combustion liability indices was evaluated. The spontaneous combustion liability indices of these samples were evaluated by crossing point temperature (XPT), Stage II Slope, FCC (Feng, Chakravorty, Cochrane) and the Wits-Ehac tests, while the coal properties were determined from the proximate and ultimate analyses. The results obtained from these coal properties were related to different liability indices to develop trends of linear relationships using regression analysis. The ad basis indicates a higher correlation coefficients than the db and daf basis for the XPT and FCC Index, while the daf shows a higher correlation coefficients than the ad and db basis for the Wits-Ehac Index. It was found that the trend of linear relationships of these coal properties differs from one liability index to the other. The XPT show a better trend followed by the Stage II Slope on the coal properties among the spontaneous combustion liability indices evaluated.

Keywords: *Coal basis; correlation analysis; liability indices; spontaneous combustion"*

Publications (Published Conference Proceedings)

8.1.3 Paper 3

Parts of chapter 4 were compiled and published as the paper: **Abisola Risiwat Gbadamosi**, Moshood Onifade, Bekir Genc & Steven Rupprecht. Evaluation of spontaneous combustion risk in South African coalfields; In Proceedings of the 2nd Biennial CEMEREM Conference on "Sustainable Development in the Extractives Industry in Africa"- 17th to 18th September 2019, Voi-Mombasa, Kenya. The following is an extract of the paper abstract:

"Abstract

Spontaneous combustion is a challenge in the coal mining industry since millennia, both from safety and economic perspectives. The self-heating of coal and carbonaceous shale is the main *cause of fire in underground collieries, open cast mines, spoil heaps and stockpiles, and this concern coal producer and users in many countries. The School of Mining Engineering, University of the Witwatersrand developed a spontaneous combustion liability test apparatus (Wits-Ehac test) in the late 1980s to measure the tendency of coal to self-heat. The apparatus incorporated the crossing-point temperature (XPT) and the differential thermal analysis (DTA) to establish an index called the Wits-Ehac Index. Four hundred and ninety-two (492) representative coal samples from various coal seams and production coalfields in South Africa over 10 successive years (2009 and 2018) were tested and a database to classify the spontaneous combustion liability of coal was developed. The classification of risk rating toward spontaneous combustion for different coal was established and found to vary from high, medium and low using the Wits-Ehac apparatus. This risk evaluation database reviewed, assessed and compared the spontaneous combustion liability indices for different coal samples obtained from the South African coalfields for a decade.*

Keywords*: Coalfields, crossing point temperature, risk rating, self-heating, Wits-Ehac Index".*


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