SPONTANEOUS COMBUSTION LIABILITY OF COALS AND COAL-SHALES IN THE SOUTH AFRICAN COALFIELDS

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A thesis submitted to the Faculty of Engineering and the Built Environment, University of the Witwatersrand, Johannesburg, in fulfilment of the requirements for the degree of Doctor of Philosophy.

Johannesburg, 2018

DECLARATION

I declare that this thesis is my own, unaided work. Where use has been made of the work of others, it has been duly acknowledged. It is being submitted for the Degree of Doctor of Philosophy in the University of the Witwatersrand, Johannesburg. It has not been submitted before in any form for any degree or examination in any other University.

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This _____ day of _____ 2018

ABSTRACT

Spontaneous combustion is one of the major challenges in the South African coal mining sector. The event involves a range of complex physical and chemical processes, caused by the chemical reaction between coal, coal-shale and oxygen. Coal and coal-shale undergo low-temperature oxidation when exposed to oxygen in the air. The frequent occurrence of self-heating of coal-shale was reported in some South African coal mines to be the likely cause of spontaneous combustion but not the coal alone. Coal-shale found between layers (above and below) of coal seams vary considerably in intrinsic properties and proneness to spontaneous combustion. This thesis evaluates the main factors which most strongly affect the spontaneous combustion liability of coal and coal-shale.

In South Africa, the widely accepted spontaneous combustion liability index, the Wits-Ehac Index was used to measure the spontaneous combustion liability index of fourteen (14) bituminous coal samples and 14 coal-shale samples respectively. The results of the Wits-Ehac Index show that most of the samples are liable to spontaneous combustion. However, the Wits-Ehac Index was unable to obtain a liability index during the testing of some coal-shale samples. This necessitated the development of a new device.

A new apparatus which accurately measures the spontaneous combustion liability by using the reactivity of oxygen within coal and coal-shale was developed. A series of spontaneous combustion tests were conducted with this apparatus and a new liability index referred to as the Wits-CT Index was created. The Wits-CT Index uses the total carbon content and the temperature variations obtained from the samples during their reaction with oxygen to predict their spontaneous combustion liability. It was found that coal and coal-shale with a high Wits-CT Index are more liable to spontaneous combustion. The results from the two liability indices were compared with respect to what is happening in the mines and proved that samples with higher spontaneous combustion liability indices are more prone to spontaneous combustion than those with lower liability indices.

The relationships between the spontaneous combustion liability (obtained from the Wits-Ehac Index and Wits-CT Index) and the geochemical data ((proximate and ultimate analysis, total sulphur and sulphur forms, petrographic composition, X-ray fluorescence (XRF) and X-ray diffractometer analysis (XRD)) of the samples were evaluated. The experimental results show that intrinsic properties of these materials complement the spontaneous combustion liability

test results. Comparative analyses of the intrinsic properties and spontaneous combustion liability indicated similarities between the mechanism of coal oxidation and that of the oxidative processes undergone by coal-shale. It was found that the coal samples have higher intrinsic properties and spontaneous combustion liability than the coal-shale samples. This study indicates that the South African coal and coal-shale are enriched with more inertinite macerals than the vitrinite and liptinite macerals. The distribution of the macerals has been shown to have reasonable influences on spontaneous combustion liability. The contents of the main ash oxides are strongly related to the mineral constituents of the samples as indicated by the XRF. The quantity and mineral constituents in coal and coal-shale were evaluated by chemical procedures and optimised by the XRD analysis. The XRD analysis confirmed the presence of mineral constituents in the samples as identified by the XRF.

The influence of the intrinsic properties of coal-shale in relation to coal properties affecting spontaneous combustion liability was established using a statistical method (regression analysis). The results provide quantitative descriptions and show the relationships between the dependence of the spontaneous combustion liability index (the Wits-Ehac Index and the Wits-CT Index) and independent variables (intrinsic properties). The linear regression analysis shows that the spontaneous combustion liability index indicates linear relationships with some of the intrinsic properties based on the set criterion and thus, identifies the major intrinsic factors affecting spontaneous combustion liability. It was found that a definite positive or negative correlation coefficient exists between the intrinsic factors and spontaneous combustion liability. A set of models to predict the spontaneous combustion liability was derived by using multiple regression analysis between the dependent variables and independent variables. The best significant correlation along with the most appropriate model as indicated by the R-squared values, the coefficient of correlations and standard error was used to predict the most reliable spontaneous combustion liability index. The results obtained from these models have been used as a reliable tool to support previous works on the role of intrinsic properties on spontaneous combustion liability.

The application of chemical inhibitors on coal and coal-shale under laboratory studies were found to create an oxidative barrier on the surface of these materials to prevent self-heating and spontaneous combustion. The study indicated that by altering the self-heating characteristics (i.e. chemical and physical characteristics of a coal and coal-shale surface) through the use of chemical inhibitors, spontaneous combustion liability can be minimized.

LIST OF PUBLICATIONS

The publications listed below have emanated from this research work so far:

Patent obtained

1. **Moshood Onifade** & Bekir Genc. A system and method for determining spontaneous combustion propensity of carbonaceous materials. Provisional Patent, Application No. **2018/00851, Our Ref. P79581ZP00.**

Publications (ISI/IBSS Journals)

1. **Moshood Onifade** & Bekir Genc., 2018. A review of Spontaneous combustion studies - South African context by International Journal of Mining, Reclamation and Environment, https://doi.org/10.1080/17480930.2018.1466402.
2. **Moshood Onifade** & Bekir Genc., 2018. A new Apparatus to Establish the Spontaneous Combustion Propensity of Coal and coal-shale by International Journal of Mining Science and Technology, Volume 28(4), pp. 649-655.
3. **Moshood Onifade** & Bekir Genc., 2018. Spontaneous Combustion of Coal and coal-shale by International Journal of Mining Science and Technology, DOI: https:// doi.org/10.1016/j.ijmst.2018.05.013.
4. **Moshood Onifade** & Bekir Genc., 2018. Modelling spontaneous combustion liability of carbonaceous materials by International Journal of Coal Science and Technology, Volume 5(2), pp. 191-212.
5. **Moshood Onifade** & Bekir Genc., 2018. Comparative analysis of coal and coal-shale intrinsic factors affecting spontaneous combustion by International Journal of Coal Science and Technology, Volume 5(3), pp. 282-294.
6. **Moshood Onifade** & Bekir Genc. Prediction of the spontaneous combustion liability of coal and coal-shale using statistical analysis by the Southern African Institute of Mining and Metallurgy, Volume 118, pp. 799-808.
7. **Moshood Onifade**, Bekir Genc, & Nicola Wagner. Influence of organic and inorganic properties of coal-shale on spontaneous combustion liability by International Journal of Mining Science and Technology (Under Review).

Publications (Published Conference Proceedings)

1 **Moshood Onifade** & Bekir Genc. Establishing relationship between spontaneous combustion liability indices. Proceedings of the 21st International Coal Congress of Turkey "ICCET" April 11-13, 2018, Zonguldak, Turkey; 2018, pp. 1-11.

2 Bekir Genc, **Moshood Onifade**, & Alan Cook. Spontaneous combustion risk on South African Coalfields: Part 2. Proceedings of the 21st International Coal Congress of Turkey "ICCET" April 11-13, 2018, Zonguldak, Turkey; 2018, pp. 13-25.

3 **Moshood Onifade** & Bekir Genc. Ash, volatile matter and carbon content influence on spontaneous combustion of coal-shale, 18th International Symposium on Environmental Issues and Waste Management in Energy and Mineral Production, 19-23 November, 2018, Santiago, Chile (Accepted).

The paper abstracts are included in Appendix 9.1.

ACKNOWLEDGEMENTS

Immeasurable appreciation and sincere gratitude to the following individuals for their guidance and support, and the following companies for their contribution in making this study successful.

- My Supervisor, Prof B. Genc (School of Mining Engineering, University of the Witwatersrand) for his invaluable academic guidance, trust and patience towards the successful completion of this research;
- Prof N. Wagner (University of Johannesburg, South Africa) for her contribution and advice during the stage of conceptualizing this research;
- Prof C. Musingwini, Head, School of Mining Engineering, University of the Witwatersrand, for providing School's support needed to successfully execute the research programme;
- Prof Emeritus H. R. Phillips for his priceless academic guidance during the research.
- Mr A. Carpede for his technical support during construction of a new equipment for making the experimental testing possible;
- Glencore and Anglo American Coal Mines for assistance during samples collection.
- My wife (A. Onifade), family members, and friends for selflessly giving their love and moral support while I undertook this research work;
- Anzolette, Zeenat, Lindy, Sonja, Andrew, Joseph, Paulos and Jacob, School of Mining Engineering, University of the Witwatersrand, for their moral support throughout the stay at the School; and
- Coaltech South Africa (Project Number: 3.3.5) and the Julian Baring Scholarship Fund (JBSF) for sponsoring this Ph.D. research.
- Gratitude expressed towards F. Genc for editing this thesis.

Lastly, the opinions expressed in this thesis are those of the author and may not necessarily represent the policies of the companies and institutions mentioned in this thesis.

DEDICATION

To my late mother (Muslimat Onifade), father (Abdul-Razak Onifade) and brother (Muritada Onifade) with whom I would have liked to share exciting moments such as this one. May Allah bless us all!

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LIST OF IMPORTANT SYMBOLS USED IN THE THESIS

% = Percentage

> = Greater than

μm = Micrometer

Btu/lb = British thermal unit per pound

ε = Porosity of the medium

g = Gram

J/kg = Joule/kilogram

Kg = Kilogram

kJ/mol= Kilo joule per mole

kJ= Kilo joule

ℓ = Density (kg/m3)

mm = Millimetre

ms^{-1} = Meter per second

^{o}F = Degree Fahrenheit

$^{o}C/h$ = Degree per hour

$^{o}C/min$ = Degree per minute

T = Tonne

LIST OF IMPORTANT SUBSCRIPTS USED IN THE THESIS

%Cad = the air-dried percentage of carbon content of the sample

Al_2O_3 = Aluminium oxide

$BaSO_4$ = Barium Sulphate

$CaCl_2$ = Calcium chloride

$CaCO_3$ = Calcium carbonate

CH_4 = Methane

CO_2 = Carbon dioxide

Cr_2O_3 = Chromium (III) oxide

Da = Darcy

Fe_2O_3 = Iron (III) oxide

HNO_3 = Hydrogen nitrate

K_2O = Potassium oxide

$KMnO_4$ = Potassium permanganate

$Li_2B_4O_7$ = Lithium tetraborate

$Mg(AC)_2$ = Magnesium acetate

$MgCl_2$ = Magnesium chloride

$MgCO_3$ = Magnesium carbonate

Na_2O = Sodium oxide

$Na_2SiO_3.5H_2O$ = Sodium metasilicate pentahydrate

Na_3PO_4 = Disodium phosphate

$NaNO_3$ = Sodium nitrate

NH_4Cl = Ammonium chloride

$NH_4H_2PO_4$ = Ammonium dihydrogen phosphate

NH_4HCO_3 = Ammonium bicarbonate

O_2 = Oxygen gas

P_2O_5 = Phosphorus pentoxide

R_{max} = Mean reflectance

SiO_2 = Silicon oxide

SO_2 = Sulphur dioxide

TiO_2 = Titanium dioxide

T_M = the difference between the sum of maximum temperatures of each thermocouple inside equipment and room temperature (22^oC)

T_R = the difference between the maximum temperature and initial temperature during oxidation reaction in degree Celsius

W_1 = Initial weight

W_2 = Final weight

W_3 = weight of ash and crucible

LIST OF IMPORTANT ACRONYMS USED IN THE THESIS

A = Rate coefficient

A% = Percentage of ash content

ANFIS = Adaptive neural fussy inference system

ANN = Artificial neural network

ANOVA = Analysis of variance

ASTM = American society for testing and materials

c = Intercept (where the line crosses the y axis).

C% = Percentage of carbon content

CaO = Calcium oxide

CaO = Calcium oxide

Cc = Calcite content (%)

cc = Cubic centimetre

CCSEM = Computer controlled scanning electron microscopy

CMC = Critical micelle concentration

CO = Carbon oxide

Cp = Specific heat capacity of coal at constant pressure (J/Kg/K)

CPE = Cost-performance evaluation

D = Dolomite content (%),

DSC = Differential scanning calorimetry

DTA = Differential thermal analysis

E = Activation energy (kJ/mol)

e = Exponential (natural log)

FAHP = Fuzzy analysis hierarchy process

FC% = Percentage of fixed carbon content

Fe = Iron

FTIR = Fourier transform infrared

G = Gypsum content (%)

GCV = Gross calorific value

H = Haematite content (%)

H% = Percentage of hydrogen content

HCl = Hydrogen chloride

HGI = Hardgrove grindability index

I = Illite content (%)

I = Slope at stage I

I = Total inertinite content (vol.%, mmf)

ICCP = International committee for coal and organic petrology

ICP-OES = Inductively Coupled Plasma Optical Spectrometry

ID = Identification number

II = Slope at stage II

IIa = Slope at stage IIa

IIb = Slope at stage IIb

III = Slope at stage III

IRH = Initial rate of heating

ISO = International standard organisation

K = Kaolinite content (%)

KCl = Potassium chloride

KOH = Potassium hydroxide

L = Total liptinite content (vol.%, mmf)

LOI = Loss on ignition

m = Meter

M% = Percentage of moisture (%)

MAIT = Minimum auto-ignition temperature

Mc = Microline content (%)

MgO = Magnesium oxide

MM = Mineral matter content (vol.%)

mmf = Mineral matter free basis

Mn = Magnetite content (%)

MnO = Manganese (II) oxide

MS Excel = Microsoft Excel

MSR = Mixture surface regression

Mu = Muscovite content (%)

MVA = Multivariable regression analysis

MVRA = Multivariate regression Analysis

N% = Percentage of nitrogen content

NaAc = Sodium acetate

NaCl = Sodium chloride

NaOH = Sodium hydroxide

NiO = Nickel (II) oxide

O% = Percentage of oxygen content

OC = Organic carbon (%)

OS% = Percentage of organic sulphur content (%)

P% = Percentage of pyrite content (%)

Pc = Plagioclase content (%)

PCA = Principal component analysis

PE = Performance evaluation (%)

PEG = Polyethylene glycol

PFC = Plus flow calorimetry

Pt100 = Platinium100 thermocouple

PVA = Polyvinyl alcohol

Q = Heat of reaction (J/Kg)

Q = Quartz content (%)

R = Correlation coefficient

R = Molar gas constant (J/mol.K)

RSV = R-squared values

S% = Percentage of total sulphur

Sa = Siderite content (%)

SANS = South African National Standard

SDL = Sodium dodecyl sulphate

SEE = Standard error of estimate

SEM = Scanning electron microscope

SS% = Sulphide sulphur content

T = Temperature (K)

t = Time (min)

TGA = Thermogravimetric analysis

TP = Flammability temperature

TTR = Total temperature rise

TWE = Wits-Ehac Index for treated sample

USBM = United State bureau of mines

UWE = Wits-Ehac Index for untreated sample

V = Total vitrinite content (vol.%, mmf)

VM% = Percentage volatile matter content

Vol.% = Percentage volume

w = Density (Kg/m^3)

WC = Wits-CT Index

WE = Wits-Ehac Index

WLC = Western lignite corporation

WOP = Wet oxidation potential

X = Coordinate of any point on the line X

XPT = Crossing point temperature

XRD = X-ray diffraction

XRF = X-ray fluorescence

y = Coordinates of any point on the line Y

GLOSSARY OF IMPORTANT TERMS USED IN THIS THESIS

Coal: A solid, brittle, stratified, combustible rock-like material which is composed of a number of microscopic organic (macerals) and inorganic constituents (minerals) which occur together in varying proportions. It comprises of less than 50% ash content.

Coalfield: An area containing significant coal deposits.

Coal-shale: A solid, brittle, stratified, combustible rock-like material which is composed of a number of microscopic organic (macerals) and inorganic constituents (minerals) which occur together in varying proportions. It comprises of 50 to 90% ash content.

Correlation coefficient: A number between +1 and −1 calculated so as to represent the linear interdependence of two variables or sets of data.

Crossing point temperature (XPT): The temperature at that point where the coal temperature begins to exceed the surrounding temperature.

Differential thermal analysis (DTA): A technique for identifying and quantitatively analysing the chemical composition of substances by observing the thermal behaviour of a sample as it is heated.

Inhibitor: a substance which slows down or prevents a particular chemical reaction or other process.

R-squared value: A statistical measure of how close the data are to the fitted regression line.

Self-heating: A process that results in an increase in temperature of a thermally-isolated mass of coal or other carbonaceous materials.

Spontaneous combustion: A type of combustion which occurs by self-heating (an increase in temperature due to exothermic internal reactions) followed by thermal run away (self-heating that rapidly accelerates at a high temperature) leading to auto-ignition.

Stage II is one of the best indicators of spontaneous combustion liability. It continues from the XPT to the point referred to as the kick–point.

Standard error of estimate: A measure of the accuracy of predictions made with a regression line.

1 INTRODUCTION

1.1 Chapter overview

Coal oxidises naturally over time and results in an exothermic reaction that generates heat. The heat produced determines the energy transfer between the coal and its surroundings due to the temperature changes. Spontaneous combustion has been a problem since coal was being mined. The event occurs when coal absorbs oxygen in the air. The coal oxidizes and ends into a flaming fire caused by the accumulated heat. The heat increases the temperature of the coal and promotes the oxidation process. Due to the complex nature of coal and its surrounding factors involved in the generation of heat, no positive decision has been reached with reference to the source of initial heating. The initial heat can be related to the oxidation of either coal and its surrounding material (coal-shale) or other external factors.

According to Alpern & Lemos de Sousa (2002), coal-shale consists of 50% to 90% ash. Coal and coal-shale which consists of varying proportions of organic matter (macerals) and inorganic materials (mainly crystalline) may undergo spontaneous combustion (Mastalerz *et al.,* 2010 and Restuccia *et al.,* (2017)). These materials have pore spaces together with the occurrence of a carbon-rich element (Alpern & Lemos de Sousa (2002), Dullien (1979) and Restuccia *et al.,* (2017)). This enables the rock to be porous to air, and with the increased surface area, the organic particles have reactive oxidation sites (Dullien, 1979). Self-heating of coal-shale has been reported in South African coal mines to be capable of starting spontaneous combustion but not the coal alone. The reasons for coal-shale to undergo spontaneous combustion has been reported to be caused by the amount of pyrite, organic composition, reactive nature and rank of the associated coal (Restuccia *et al.,* (2017) and Rumball *et al.,* (1986)). Incidents of coal-shale self-heating have been observed in areas such as overburden shale, selected bands of coal seams, spoil heaps and highwalls. Therefore, self-heating of coal and coal-shale is an important phenomenon that requires an in-depth investigation. Studies of self-heating in a selected band of coal seams (above and below), coal-shale, and highwalls are limited and no sufficient information is available to investigate and predict the spontaneous combustion liability of coal-shale in relation to coal. However, these materials consist of varying microscopic organic and inorganic constituents which accelerate spontaneous combustion.

This study examines selected intrinsic factors (moisture, ash, volatile matter, carbon, hydrogen, nitrogen, total sulphur, forms of sulphur and maceral composition) affecting spontaneous combustion liability of coal and coal-shale and provides statistical interpretation of these materials analysis data. Furthermore, the application of selected chemical inhibitors to minimize spontaneous combustion liability was investigated. All these, will be useful for establishing significant relationships between coal and coal-shale in terms of spontaneous combustion liability.

1.2 Background of the study

Spontaneous combustion of coal takes place in nearly every coalfield worldwide, though the frequency and extent of the incidents vary from country to country. One of the principal dangers faced in the coal mining industry over the years is spontaneous combustion. For many years, it was the key cause of underground fires in South African coal mines and a frequent concern in open cast mines, generally in the run-of-mines coal and, in waste dumps, stockpiles and spoil heaps. The experience of spontaneous combustion has approached into a phase that makes practical illustrations feasible as a result of the re-established awareness of self-heating of coal and coal-shale in coal mines (Kim & Chaiken (1990), Restuccia *et al.,* (2017) and Rumball *et al.,* (1986)). Past researcher's efforts to measure the liability of coal stockpiles were primarily based on laboratory tests of one or more of the coal characteristics associated with the self-heating behaviour. A limited number of medium-large scale tests on coal have been carried out in the past. However, such tests have not been conducted frequently due to the high cost and long period of time required and the difficulties in the interpretation of the results.

Frequent occurrence of spontaneous combustion is one of the major challenges faced by the South African coal mining sector (Eroglu (1992), Genc *et al.,* (2018) and Gouws & Wade (1989b)). Coal oxidation occurs in an environment where there is sufficient oxygen. The oxidation rate of coal cannot be easily detected due to the reaction being very slow. In open cast mining, the ingress of oxygen in the air into discontinuities and exposure of the highwall for long periods are among the factors affecting spontaneous combustion. In underground coal mines, the combined effects of the quantity of the airflow and self-heating areas can cause spontaneous combustion. It is known that coal, roof shales, spoil heaps and other carbonaceous materials can self-heat and liberate heat naturally when exposed to oxygen in the air (Kim & Chaiken (1990) and Restuccia *et al.,* (2017)). The heat generated becomes higher than the heat dissipated to the surroundings.

Coal seam, spoil heaps and waste dumps comprise of weathered coal, clay, pyritic shale, coal-shale and other strata associated with them. The chemical reactions between oxygen and external active structures of the mentioned materials which releases heat are the required constituents of self-heating (Kim & Chaiken (1990), Mastalerz *et al.,* (2010) and Rumball *et al.,* (1986)). The spontaneous combustion of coal with a potential transition into endogenous fires constitute a direct risk to the safety of the working conditions and unfavourably influence the mine environments. There are two points of concern when investigating the self-heating rate of carbonaceous materials due to the presence of oxygen. Firstly, the accumulation of overburden materials (such as shale and sandstone etc.) on the coal surface when exposing a new coal seam. This is common in coal mining operations both in the past and present. The second is geology of the material, particularly during the formation of the deposits.

Experimental investigations on spontaneous combustion of bulk coal samples have been carried out under a medium-large scale test (Akgun & Arisoy (1994a), Beamish *et al.*, (2001), Chen (1991), Cliff et *al.,* (1998), Monazam *et al.,* (1998), Smith *et al.,* (1991) and Stott (1980)) with a heating system used to initiate the self-heating process. Studies on spontaneous combustion of coal stockpiles under the influence of atmospheric conditions have been reported (Fierro *et al.,* (1999), Fierro *et al.,* (2001), Ozdeniz & Sensogut (2006), Ozdeniz & Yilmaz (2009) and Ozdeniz *et al.,* (2015)). However, limited studies reported experimental methodologies to imitate the effects of atmospheric conditions on coal spontaneous combustion in the laboratory without an applied heating system. An acceptable standard widely used in South Africa to predict the spontaneous combustion liability of coal, known as the Wits-Ehac Index has been in existence and used for more than 30 years (Genc *et al.,* (2018), Genc & Cook (2015), Gouws & Wade (1989a) and Gouws & Wade (1989b). The test involved the use of relatively small amounts of pulverized, dry samples to determine the spontaneous combustion liability of coal. The test measures the temperature in degrees and the influence of limiting factors can be evaluated only under the available experimental conditions, unlike the medium-large scale test that considered the self-heating of a substantial coal mass under atmospheric conditions. Spontaneous combustion is dependent on the composition of the material, air temperature, sample temperature and mine environment. Researchers have indicated that the ignition temperature tests are at high temperatures where an external source of heat has been applied at the surface of the coal particles. The effects of oxygen, moisture and temperature variations within a coal mass under airflow conditions, i.e. the most likely scenario at a mine, cannot be integrated into the small-scale test.

The spontaneous combustion liability of some coal-shale could not be determined by the Wits-Ehac Index because of their low reactivity under the available experimental conditions. This necessitated the need to develop a device that can measure the spontaneous combustion liability of coal, coal-shale and other carbonaceous materials under the influence of oxygen which can produce similar results compared to the existing Wits-Ehac Index. The use of realistic medium experimental scale tests that emulate the influence of oxygen in air on various coal and coal-shale types to predict spontaneous combustion liability is important. The principal argument against laboratory testing in predicting the liability of coal toward spontaneous combustion is that the process is unlike the natural conditions under which spontaneous combustion takes place. On the basis of these criteria, a medium scale laboratory apparatus is developed to generate realistic data and predict the spontaneous combustion liability of coal and coal-shale. The apparatus considers a number of parameters such as airflow rate and temperature variations due to the influence of the oxygen. The need for a reliable method to capture the large volume of data used to predict the occurrence of spontaneous combustion motivates this new methodology.

This thesis investigated the intrinsic properties (proximate, ultimate, total sulphur and forms of sulphur, petrographic composition, XRF and XRD) and spontaneous combustion tests carried out on fourteen (14) coal and 14 coal-shale samples in South African coal mines. Spontaneous combustion tests were conducted using the Wits-Ehac test and also with the new apparatus (Wits-CT). The analyses from this thesis have established significant relationships between coal and coal-shale in terms of spontaneous combustion liability.

1.3 Description of the Study Area

The study areas are located in the Witbank Coalfields, South Africa. The Witbank Coalfields have been the heart of coal mining in South Africa from the time mining began in the 1890s. The majority of collieries in the Witbank region have in the past adopted bord and pillar mining methods, usually with low coal recovery ratios, leaving behind a considerable quantity of coal in pillars as well as roof and floor coal. The flow of oxygen in the air into mine openings caused low-temperature oxidation and chemisorption of coal resulting in spontaneous combustion. The influences of spontaneous combustion at the Middleburg colliery, Witbank coalfield was investigated by Bell *et al.* (2001). The occurrence of spontaneous combustion was first observed when the mining operations were stopped in 1947. The self-heating of coal was first found in the Sasolburg coalfield in 1985 in unexcavated coal in the New Vaal colliery when

old workings in the middle seam were exposed to oxygen in the air. The need to drain water from the old workings took place due to large quantities of water beyond the normal limit in the old workings which resulted in serious spontaneous combustion in the existing seams (top, middle and bottom). The mine faced serious challenges in minimizing spontaneous combustion due to the problem of finding the old underground workings and locating the bords while drilling. The mine also experienced spontaneous combustion in spoil heaps, highwall, waste dumps, blast holes and stockpiles.

The Boschmanskrans pit in the Middleburg colliery frequently encountered the event of spontaneous combustion in the highwall, spoil heaps, stockpiles and the No. 2 seam due to the earlier underground workings. There was a problem of insufficient sand in the mine for the cladding of heating in the highwall. The high liability to spontaneous combustion of the No 3 seam and its high sulphur content at the Arthur Taylor colliery was the reason the coal seam could not be regularly excavated for commercial purposes. The mine experienced a spontaneous combustion problem in the spoil heaps and the underground old workings in the Northern part of the lease region and the problem was visible in the interburden. Kleinkopje Colliery has experienced serious spontaneous combustion in spoil heaps, waste dumps and in the highwalls once it is exposed to air and when oxygen in the air penetrates into the underground old workings from connection points between two ramps. The frequent incident of spontaneous combustion at Landau colliery was mitigated in 2003 when the parting between two seams in the mine was at its thickest. This is because the coal mining operations were required to be carried out in two phases. The threat of coal self-ignition in the mine was critical and long term and reliable control techniques were introduced to minimize the problem.

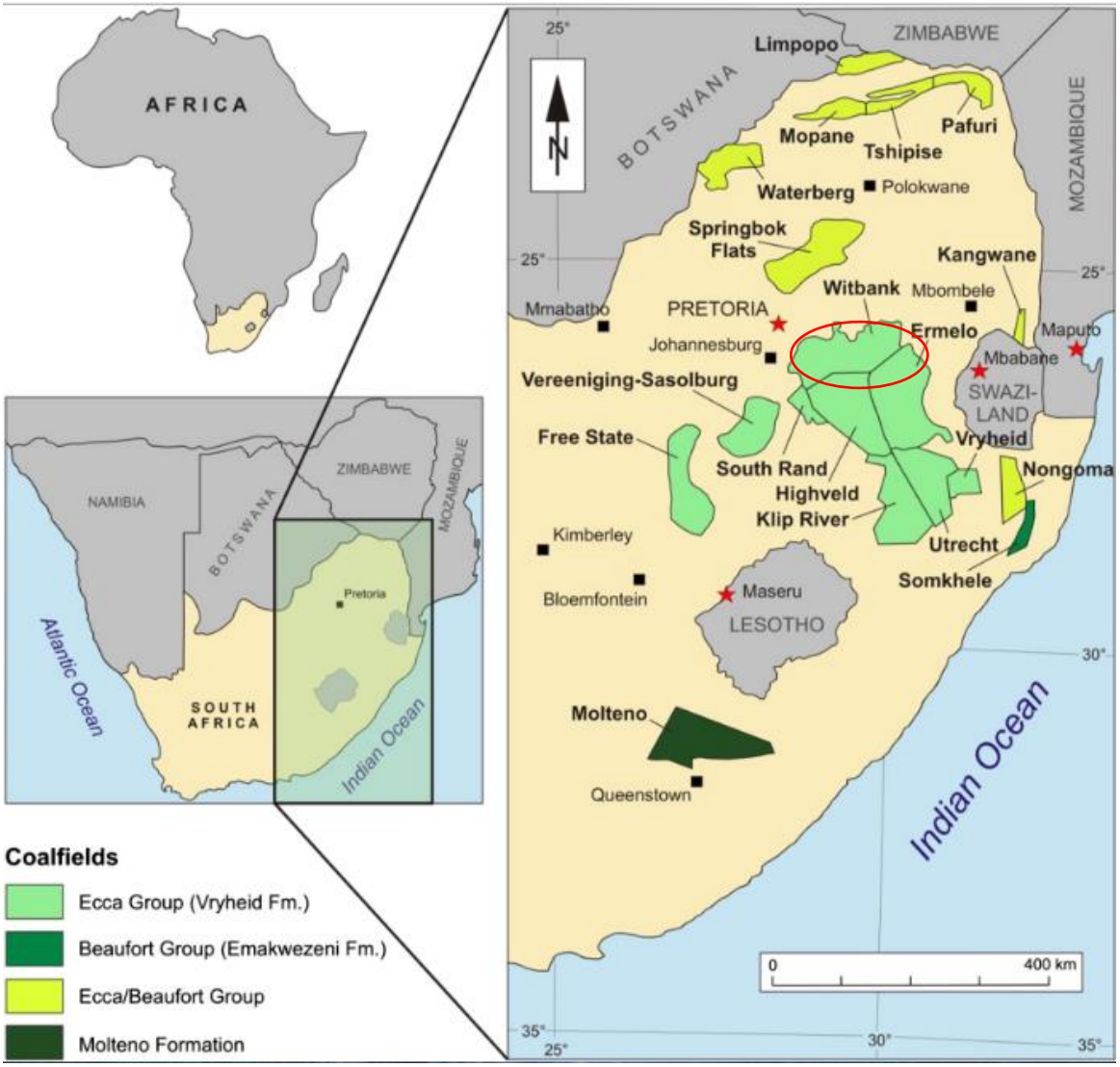

Figure 1.1 Coalfields of South Africa. The Mopane, Tshipise and Pafuri Coalfields collectively form the Soutpansberg Coalfield. Fm. = Formation (Snyman, 1998)

1.4 Spontaneous combustion problem in Glencore and Anglo Mines

Recent studies shows that spontaneous combustion incidents are not common in South African underground coal mines (Genc & Cook (2015) and Stenzel (2002)). However, open-cast mines in the Witbank area experiences spontaneous combustion in areas earlier mined by the bord and pillar methods, spoil heaps, stockpiles, waste dumps and overburden shales. The occurrence of spontaneous combustions within these mines is rarely reported due to the control practice used in mitigating the incident. Research on the particular issue on spontaneous combustion and ignition in South African open cast mines are uncommon (Adamski (2003), Eroglu *et al.*, (1991), Genc & Cook (2015), Gouws & Wade (1989a), Gouws & Wade (1989b) and Wade (1989)). Stenzel (2002) undertook an evaluation of past events of spontaneous

combustion at the New Vaal colliery which is relevant to this study. A study by Phillips *et al.* (2011) in South Africa was restricted to the study of the problem of surface mining of previously mined seams where a number of coal operators and mines implemented some control measures to manage such incidents.

Recent studies and frequent incidents of coal and coal-shale self-heating have been reported by the Glencore Mines (Tweefontein Mine, Goedgevonden Colliery and iMpunzi Mine) and the Khwezela-Bokgoni Pit (Anglo Mine) to be capable of starting spontaneous combustion. Problems observed and reported as shown in Figure 1.2 in the mines include self-heating leading to spontaneous combustion in;

- Overburden shales, spoil heaps, highwalls, run-off-mines, selected bands of coal seams and waste dumps;
- Blast holes during drilling operations thereby creating a risk of premature detonation;
- Buffer zones and coal stockpiles undergoes spontaneous combustion leading to high rehabilitation costs, damage to equipment and injuries to workers.

Figure 1.2 (a) Self-heating of run-of-mine and (b) spoil heaps at Tweenfontein Mine (c) Signs of self-heating in coal seams at Khwezela Mine (Bokgoni Pit) and (d) self-heating of in-seam shale at Goedgevonden Colliery, Witbank, South Africa

There were conflicting views whether coal-shale is capable of starting spontaneous combustion or not after a visit to these mines. Evidence showed by Glencore Mines indicates that spontaneous combustion of coal-shale has significant influence in coal mining operations. Carbonaceous shale, shale roof and coal roof provide permanent pathways for self-ignition to

increase (Jones & Cameron (1987) and Kim & Chaiken (1990)). The properties of coal, coal-shale and other related carbonaceous materials can be a factor to evaluate the cause of spontaneous combustion between coal seams and abandoned mines. Hence, self-heating of coal-shale can be reasoned to cause spontaneous combustion in the coal mines.

1.5 Problem statement

One of the challenges in the coal industry is the spontaneous combustion of coal, as this becomes a frequent problem in the course of mining, stockpiling, and transportation. The characteristics of coal and coal-shale intrinsic properties between bands of coal seams affecting spontaneous combustion is yet to be studied. The accessibility of sufficient air in spoil heaps, highwall, coal-shale and mined out areas where the coal has been left, especially if it is loose coal (coal fragments larger in size than coal dust), may be the cause of spontaneous combustion in coal mines. Spontaneous combustion occurs between selected bands of coal seams, coal-shale, highwall, coal stockpiles, distribution processes, washed and sized coal etc. The application of laboratory scale tests on coal properties cannot ascertain the complex scaling results on spontaneous combustion liability of coal under the effects of atmospheric conditions. Exposure of open-pit walls for long periods could result in instability in walls, cracks and end up as spontaneous combustion. The rate of airflow and quantity of coal that accumulate in underground mines can combine to give an optimum condition for spontaneous combustion to occur.

Coal stockpiles are equally a significant area of concern and prevention of spontaneous combustion has been a problem both in bigger and smaller stockpiles. It is not realistic to exclude air completely from a stockpile or dump. Wind direction is a contributory factor for the ingress of air into old workings to cause a pressure differential between the top and the bottom of the highwall. The quantity of airflow (m^3/s) is a difficult issue because air provides oxygen and removes the heat generated (Baneerjee, (1985), Brooks *et al.,* (1988) and Fierro *et al.,* (1999)). There is a significant amount of air which provides adequate oxygen for coal oxidation but is not adequate to reduce the heat produced from accumulation (Fierro *et al.,* (1999), Kim, (1977), Krishnaswamy *et al.,* (1996), Smith *et al.,* (1991)). The variation in atmospheric conditions (ambient temperature and pressure changes) during drilling of blast holes and the ingress of oxygen in the air within the coal seam discontinuities (joints, fractures, fissures and cracks) can promote spontaneous combustion. Several cavities in the mine

workings exposed by strip mining, blast holes and core drilling allow oxygen in the air to intrude into the openings causing spontaneous combustion.

A number of critical experiences has been reported as a consequence of coal-shale self-heating potentially leading to spontaneous combustion in the South African coal mines. Kim & Chaiken (1990), Restuccia *et al.* (2017) and Rumball *et al.* (1986) indicated that carbonaceous shale is a phenomenon known to be liable to spontaneous combustion but the inherent characteristics of this material with related coal properties is yet to be investigated. Genc & Cook (2015) indicated the tendency for risk of spontaneous combustion despite the low frequency of underground incidents and suggested further research. Considering these concerns raised and the well-known fact that spontaneous combustion is a long-standing problem, a critical study of the main factors causing spontaneous combustion of coal and coal-shale is required. The reason is to establish relationships between coal and coal-shale intrinsic properties causing spontaneous combustion and prove the concept that inhibitors can minimize the occurrence.

1.6 Relevance of the research and research question

The negative environmental influence caused by spontaneous combustion of coal and coal-shale have resulted into environmental pollution and greater loss of precious resources. The increase in coal fines, reduction in production and increase in the cost of rehabilitation have caused great loss to the mines and damages to the surrounding areas. This is due to frequent growth in the event of spontaneous combustion in the affected coal mines at Witbank, South Africa. The increase in the cost of minimizing this incident is caused by the continuous occurrence of spontaneous combustion in different distribution processes and areas of the mine. The best way to manage this occurrence is to critically investigate the contributory factors influencing the phenomenon.

The experimental investigation of this study creates a practical workflow that gives a clear picture and better understanding of the main factors that are responsible for the spontaneous combustion liability of coal and coal-shale. This is done for the purpose of establishing techniques that can be applied to minimize the incident. This study establishes an extensive database to compare selected intrinsic properties of coal and coal-shale with respect to spontaneous combustion liability on a relative rating scale and develops a classification scheme for coal and coal-shale liable to spontaneous combustion.

The reaction of oxygen with coal, coal-shale and other carbonaceous materials is the prerequisite and basis for self-heating and spontaneous combustion. The liability of coal toward spontaneous combustion in the laboratory for their applications has been carried out with various techniques in different parts of the world, yet the incident continues. The techniques of monitoring, preventing and controlling of spontaneous combustion in a particular location is a difficult task because of the nature of coal and its surrounding bedrock. This requires a better technical and research-based study to investigate the occurrence. This is achieved by a clear perception of the combined processes that aid spontaneous combustion in the coal mines, taking into consideration the comprehensive studies of the characteristics of both coal and coal-shale. This study enables mine operators to obtain reliable data from experimental tests to evaluate the factors affecting spontaneous combustion and assist mine management to employ productive counter-actions.

Many nations evaluate the source of spontaneous combustion and control as a task in the responsibility of specific mines and engineers. The understanding of the mechanism of self-heating and spontaneous combustion varies in different mines on the basis of the type and amount of organic and inorganic matter, rank, reactive nature and the enviroment. The most suitable approach to prevent this event is to have a better knowledge of the coal and its overlying materials. As the incident of spontaneous combustion is a concern in the South African coal mining industry, the need to carefully manage the event is highly crucial.

From the previous sections, it can be noted that this thesis aims to provide answers to the question, *"What is the cause of spontaneous combustion in the South African Coalfields?* The question raises the following sub questions;

- *Are the causes coal and coal-shale or external related factors? and;*
- *Which appropriate model can be used to predict spontaneous combustion liability and how can the occurrence be minimized?*

Risk evaluation as an attempt to prevent spontaneous combustion problems in Coalfields provides logical and scientific guidelines to recognize hazards in an implicit environment of coal mines, storage and transportation. However, adapting controlled and reliable techniques through a critical investigation of coal and coal-shale intrinsic properties influence on spontaneous combustion liability can make prediction viable for spontaneous combustion in coal mines. The provision of an appropriate response to the questions will create a platform

for coal operators in all sections of their mines to work towards a collective goal to manage the occurrence to an extent that mining operations can proceed safely without a threat to people and the surrounding environment. The South African coal mining sector will benefit as this research provides appropriate models to predict the occurrence of spontaneous combustion and recommend suitable chemical inhibitors to minimize the event. It is important to investigate the new methodology that will be established to predict and manage the event.

1.7 Objectives of the research

Following the research question in Section 1.5, the main objective of the research was to study spontaneous combustion liability of coal and coal-shale incidents in the South African Coalfields. Coal and coal-shale tend to self-heat under favourable atmospheric conditions. To test the spontaneous combustion liability of coal, the Wits-Ehac Index has been used since the late 1980's. However, in some cases, the Wits-Ehac Index failed to produce results when testing some coal-shale samples. To overcome this problem, a new apparatus was introduced and developed to test coal and coal-shale under chemical reactions with oxygen. The apparatus determined the temperature variations due to chemical reactions between oxygen and coal and coal-shale in a closed system. The equipment keeps record and monitors the profile of temperature variations, changes in weight and airflow rate during the testing period. The new apparatus indicated reactivity of oxygen absorbed by a mass of coal and coal-shale in relation to the time rate of reaction. The equipment considers a number of parameters such as airflow rate, change in sample weight and temperature variations due to the influence of oxygen. The method is reliable, accurate and gives the necessary information on the effects of oxygen on spontaneous combustion liability of coal and coal-shale.

The equipment emulates the influence of oxygen adsorption on coal and coal-shale to create a new liability index referred to as the Wits-CT Index. The Wits-CT Index uses the total carbon content of the sample and the temperature variations obtained from the samples during the reaction with oxygen to predict spontaneous combustion liability. The results obtained from the two liability indices were compared and are in-line with what is revealed in the mines. This provides a better understanding of the fundamental mechanisms of spontaneous combustion achieved by research-based studies.

The second objective investigated selected properties of coal and coal-shale samples using laboratory tests and establish relationships with the spontaneous combustion liability index.

The characterization of the samples toward spontaneous combustion involved different techniques to study their physical, chemical, mineralogical and petrographic properties. The occurrence of various intrinsic and extrinsic parameters influencing the start and development of self-heating is the motivation to understand the mechanism of spontaneous combustion. For this reason, a number of different standardized analytical tests were carried out to determine the intrinsic factors (geochemical tests- proximate and ultimate analysis, sulphur forms, petrographic analysis, XRF and XRD). There is limited information to predict the spontaneous combustion liability of coal-shale. However, coal-shale may contain varying proportions of microscopic organic and inorganic constituents affecting spontaneous combustion liability. The tests established the characteristics relationship and enabled proper classification of coal and coal-shale known to be liable to spontaneous combustion. To make this study more logical, relationships surrounding the experimental studies to predict the cause of spontaneous combustion were evaluated with particular emphasis in the Witbank Coalfields, South Africa.

Furthermore, the third objective is to use the developed models that measure the spontaneous combustion liability of coal and coal-shale using statistical analysis. This provides a statistical interpretation of coal and coal-shale analysis data and attempts to study selected intrinsic factors influencing the liability of these materials toward spontaneous combustion. The combined effects of the major intrinsic properties of these materials on spontaneous combustion liability for predictive purposes using multiple regression analysis were evaluated. The models considered and identified the effects of selected factors affecting spontaneous combustion liability. This will be useful to establish significant relationships between coal and coal-shale spontaneous combustion liability. Furthermore, the fourth objective is to investigate four (4) selected inhibitors that can be used to minimize the occurrence of spontaneous combustion.

In order to achieve these objectives, the following were set out:

- To predict the spontaneous combustion liability of coal and coal-shale using the Wits-Ehac Index and the new liability index (Wits-CT Index);
- To investigate selected properties of coal and coal-shale using laboratory tests and to establish relationships with the spontaneous combustion test results;
- To develop models using statistical software to evaluate the incidence of spontaneous combustion in South African coal mines; and
- Investigate selected chemical inhibitors to minimize spontaneous combustion.

1.8 Methodology

To evaluate the spontaneous combustion liability of South African coal mines, detailed experimental tests were conducted to describe the distinctive nature of coal and coal-shale and indicate the mechanism that controls their liability towards spontaneous combustion. The following tasks were carried out to achieve the objectives stated in Section 1.7:

- Site investigation to identify affected areas of spontaneous combustion;
- Literature studies on local and international findings of the industries, researchers and scientists in-line with the present research work;
- Sample collection from affected coal mines using the ply sampling technique;
- Determining spontaneous combustion liability index of coal and coal-shale;
- The development of a new method (Wits-CT apparatus) to determine spontaneous combustion liability of coal and coal-shale since the Wits-Ehac Index could not produce results for some coal-shale;
- The comparison and validation of results of the Wits-CT Index and the Wits-Ehac Index to the incident of spontaneous combustion in the mines;
- Coal and coal-shale geochemical testing (proximate and ultimate analysis, petrographic analysis, XRF and XRD analysis) and relationships with the spontaneous combustion liability index;
- To develop models with related computer programmes (statistical software) to evaluate spontaneous combustion incidence in coal mines; and
- To investigate selected chemical inhibitors to minimize and control spontaneous combustion occurrence.

1.9 Scope of research

Providing suitable solutions to the research question introduces simple research opportunities, which is the primary aim for this thesis. The main part of the study was focussed on problems experienced in the areas of the selected band of coal seams, coal-shale/overburden shale and highwalls. However, other areas such as waste dumps, run-off mine, spoil heaps, underground old workings areas, washed and sized coal have been reviewed but not incorporated as part of the investigations in the study. Fourteen (14) bituminous coal and 14 coal-shale collected from areas prone to self-heating in the Witbank Coalfields have been studied. The relationship between the geochemical data of coal and coal-shale and spontaneous combustion liability

were evaluated and discussed. Statistical analysis was applied to establish relationships based on parameters that showed significant correlations between spontaneous combustion liability and coal and coal-shale properties. The best reliable results along with the most suitable model was selected and recommended to predict spontaneous combustion liability of coal and coal-shale. Furthermore, the investigation of selected chemical inhibitors to minimize and control the event of spontaneous combustion was carried out to select the most suitable chemical inhibitors.

1.10 Structure of thesis

The thesis consists of seven chapters:

Chapter 1: Introduction

This section introduces the research topic and includes the background, study areas, problem statement, the objectives, the relevance of the research and research questions, the scope of the work, structure of thesis and the information each chapter presents in the thesis concisely.

Chapter 2: Literature Review

This section provides a summary of the research topic with a comprehensive review of literature that centres on the national and international studies on similar work being conducted on spontaneous combustion of coal and coal fires and numerical modelling for the evaluation of spontaneous combustion risk in coal mines.

Chapter 3: Experimental methods

This section describes the various experimental techniques carried out on different coal and coal-shale and provide detailed information on the testing procedures. The experimental studies involved geochemical tests (proximate analysis, ultimate analysis, total sulphur and forms of sulphur, petrographic analysis, XRF and XRD analysis) and spontaneous combustion tests. The liability of the samples towards spontaneous combustion was determined using the Wits-Ehac tests and the newly developed apparatus (the Wits-CT tests).

Chapter 4: Spontaneous combustion and geochemical test results and discussions

This section provides and discusses the major results and interpreted the research findings obtained from the experimental tests performed. The results of the newly developed apparatus,

the Wits-Ehac Index and the geochemical tests of the samples were summarised and presented. Results were discussed in detail and comparisons were drawn from the samples tested.

Chapter 5: Model development and discussion

This section details the results of selected intrinsic properties of coal and coal-shale and established relationships with the spontaneous combustion liability index using statistical analysis (regression method). The statistical analysis was conducted to identify the significantly correlated parameters. The best significant parameters along with the most suitable model were selected, validated and recommended for the evaluation and prediction of spontaneous combustion incidents in South African Coalfields.

Chapter 6: Investigation and selection of chemical inhibitors

This section investigated four inhibitors and selected the most suitable inhibitor that can minimize the spontaneous combustion liability of coal and coal-shale.

Chapter 7: Conclusions and Recommendations

This section provides a general review of the study based on the observations drawn from the experimental and model investigations. The research findings and their likely suggestions were discussed. Recommendations were made around the findings to provide important meaning and implications.

1.11 Summary

This chapter attempts to introduce the reader to some background information, the main research question, motivation and structure of the thesis. Furthermore, it described why it was necessary to highlight the influence of coal and coal-shale spontaneous combustion incidents to the South African coal mining sector and other beneficiaries through this research. The next Chapter itemises the findings of literature survey on topics related to this research.

2 LITERATURE REVIEW

2.1 Introduction

This section provides comprehensive studies that centres on the international position on research being conducted by academics, different research institutes and industries on spontaneous combustion and coal mine fires. The principle and possible factors causing the event as well as different methods used to evaluate the risk were reviewed.

2.2 The basics of spontaneous combustion

The occurrence of spontaneous combustion is not limited to coal but the phenomenon is known to take place in a number of coal-shale and other carbonaceous materials (Kim & Chaiken (1990), Restuccia *et al.,* (2017) and Rumball *et al.,* (1986)). The principles that control spontaneous combustion are similar in all cases, irrespective of the material that is involved. The rate of oxygen adsorption in coal is a function of coal rank and the heat generated by coal is the cause of coal oxidation and it depends on a number of factors (Brooks *et al.,* 1988). Coal exposed to oxygen in the air for long periods of time (old coal) is less reactive than freshly exposed coal (Brooks *et al.,* (1988) and Phillips *et al.,* (2011)). A study reported by Brooks *et al.* (1988) indicated that fine coal particles require limited amounts of oxygen to infiltrate due to its low permeability, while coarse coal is very reactive due to its high permeability. Oxygen can penetrate into coal seams from the top and other portions that are in contact with the atmospheric condition. The rate of reactivity with time within a coal seam depends on the coal age and oxygen concentration (Kaitano *et al.,* 2007). As time increases, more heat will be produced at an increased depth, hence making it difficult to distribute the heat. Therefore, the heat generated to cause spontaneous combustion cannot be eliminated.

Self-heating of coal occurs when sufficient oxygen in the air is available to support the low-temperature reaction of coal with oxygen. The heat produced by coal oxidation is not sufficiently dissipated by conduction or convection, thereby causing a net temperature increase in the coal mass. The interaction of coal with oxygen at low temperatures is exothermic as a whole, although some reaction sequences could be endothermic (Shi *et al.,* (2005) and Wang *et al.,* (2003)). Low-temperature oxidation is the cause of heat that leads to spontaneous combustion in a coal mass. Exothermic processes such as microbial metabolism, the interaction of coal with water and oxidation of pyrite can promote self-heating in a coal mass if the heat

produced by coal oxidation is not sufficiently dissipated to the surroundings via conduction, convection and radiation.

The conditions that necessitate the occurrence of spontaneous combustion in Coalfields exclusive of the effect of external factors are;

(a) Coal must react with oxygen;
(b) Reaction must be exothermic followed by generation of heat; and
(c) The rate of heat generated must exceed the rate of heat dissipated.

Coal oxidation begins with exothermic chemical reactions as shown in Equation (2.1) (Schmal (1989), Singh & Singh (2005), and Stracher & Taylor (2004)).

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

In general, oxidation of coal can be expressed as a process in the following logical sequences which are;

(1) Physical adsorption;
(2) Chemical adsorption to form a coal-oxygen complex; and
(3) Chemical reaction.

The chemical reaction breaks down the less stable coal-oxygen complex to form gaseous products such as CO, CO_2 and H_2O (Mohalik *et al.,* 2009). The process is simplified in Equation (2.2) (Singha & Singh, 2005).

$$\text{Coal} + O_2 \rightarrow CO_2 + \Delta H\uparrow \quad (2.2)$$

The reaction above can be completed in a number of steps and pathways. The complete coal oxidation can be represented by Equation (2.3) (Schmal (1989) and Singha & Singh (2005)).

$$C_{110}\,H_{74}\,O_{11} + 113O_2 \rightarrow 100CO_2 + 37H_2O + 4.2 \times 10^8 J/KmolO_2 \quad (2.3)$$

The first stage of the reaction is the chemical absorption of oxygen on the coal surface as shown in Equation (2.4) (Schmal (1989) and Singha & Singh (2005)).

$$C_{110}H_{74}O_{11} + 17.5O_2 \rightarrow C_{110}H_{74}O_{46} + 2.5 \times 10^8 J/KmolO_2 \quad (2.4)$$

The exothermic reactions occur in the presence of catalytic substances such as pyrite, as represented in Equation (2.5) and (2.6) (Schmal (1989) and Singha & Singh (2005)).

$$2FeS_2 + 7O_2 + 2H_2O \rightarrow 2H_2SO_4 + 2FeSO_4 + 0.37 \times 10^8 J/KmolO_2 \quad (2.5)$$

$$4FeS_2 + 15O_2 + 2H_2O \rightarrow 2H_2SO_4 + 2Fe_2(SO_4)_3 + 0.56 \times 10^8 J/KmolO_2 \quad (2.6)$$

The reaction of coal oxidation in air can be better simplified as proposed by (Kim & Chaiken (1993) and Singh & Singh (2005)) in Equation (2.7).

$$CH_{1.18}N_{0.15}O_{0.35}S_{0.005} + 1.12O_2 + 4.15N_2 \rightarrow CO_2 + 0.58H_2O + 0.005SO_2 + 4.15N_2 + Heat \quad (2.7)$$

The organic constituent of coal consists of hydrogen, oxygen, nitrogen and sulphur in addition to carbon that can lead to the introduction of several simultaneous reactions as indicated in Equation 2.7.

The three principal variables that take place in the low temperature of coal reported by Glasser & Bradshaw, 1990 are;

- The chemical reaction between coal and the gaseous reactant result in spontaneous combustion;
- The transfer of the gaseous reactant into the bed; and
- The rate of heat dissipated from the bed.

The inhomogeneous nature of waste piles and coal seams considerably add to the understanding of the three variables. Spontaneous combustion of coal increases during mining when coal seams are more or less exposed to the irresistible effect of atmospheric conditions. The basis for investigating the influence of the above variables is significant to better understand the cause of spontaneous combustion.

2.3 Areas of coal self-heating

Self-heating is a major challenge in the coal chain (Arisoy & Akgun (2000), Banerjee (1985), Beamish *et al.,* (2001), Brooks *et al.,* (1988), Cliff *et al.,* (2014), Fierro *et al.,* (1999), Genc & Cook (2015) and Phillips *et al.,* (2011)). The danger of self-heating of coal stockpiles mostly occur in the long-term storage of thermal power stations, in open cast mines, underground old workings, highwall, spoil piles and transportation in cargo ships or trains covering long distances (Carras & Young (1994), Moghtaderi *et al.,* (2000) and Zhu *et al.,* (2013)). Due to the high cost of running industrial scale tests and the period required to run one experiment, limited industrial scale tests have been conducted on the characteristic of large coal mass. Stockpiles can experience subcritical self-heating which minimizes both the calorific value and

amounts of coal stocked. The difference in the ambient temperature due to sun radiation and variation in stockpile particle sizes may affect spontaneous combustion (Nelson & Chen, 2007).

Coal barge/silo/bunker causing subsequent unplanned closure is another area which is known to undergo self-heating. Coal stored in the silo over a long time may oxidise and begin to heat. The time it takes coal to reach the burning point is dependent on the coal reactivity. When coal is in a silo, it is kept over a period of time during a programmed maintenance shutdown, it may combust spontaneously and by the time the fire is observed, it may be very difficult to get down into the silo to access the root of the fire (Hoover, 2005). Areas in which spontaneous combustion have occurred are shown in Figure 2.1.

Figure 2.1 (a) Coal waste heap during spontaneous combustion (b) Symptom of self-heating in large fracture commonly observed in Wuda coal mining area (Singh & Singh, 2005), (c) Spontaneous combustion of open bords and (d) highwall at New Vaal, South Africa (Stenzel, 2002)

2.4 Factors influencing coal spontaneous combustion

A number of factors considerably accelerate low-temperature oxidation of coal. Such factors include the oxygen concentration and temperature (Van der Plaats *et al.*, 1984), inherent moisture content (Bhattacharyya, 1971a), the presence of mineral particles (Sujanti & Zhang, 2001) and the surface area (Singh & Demirbilek, 1987). The reaction can also be influenced

by the amount of each maceral composition (Falcon (1987), Mastalerz *et al.,* (2010), Misra & Singh (1994), Scott (2002) and Scott & Glasspool (2007)), coal rank (vitrinite reflectance) (Misra & Singh, 1994) and volatile matter and chemical composition of coal (Marinov, 1977). Factors such as geological, environmental, mining, physical and chemical conditions affect the process of self-heating and spontaneous combustion in coal (Eroglu (1992) and Phillips *et al.,* (2011)). Geological factors include seam thickness, seam gradient, organic matters and geological discontinuities (fissures, joints, cracks and faults). In open cast mines, the quantity of coal left on the coaling bench, micro and macro cracks in benches and outcrops are the main factors affecting spontaneous combustion. In underground mines, factors such as pillar and roof conditions, rate of advance, ventilation system and airflow, panel dimension, mining method, multi-seam working, ratio of coal extraction, etc. are the mining factors affecting spontaneous combustion of coal (Phillips *et al.,* 2011). Environment factors affecting spontaneous combustion include air pressure, relative humidity, wind speed and direction, oxygen concentration, moisture and sun radiation (Moghtaderi *et al.,* (2000), Ozdeniz *et al.,* (2015), Ozdeniz & Yilmaz (2009), Sensogut & Ozdeniz (2005) and Sensogut *et al.,* (2008)). External factors like discarding of hot ash, frictions of the conveyor belt, electrical short circuits and ignition may also affect spontaneous combustion. Some of the physical and chemical parameters affecting the spontaneous combustion liability of coal include moisture, volatile matter, ash, carbon, oxygen, organic sulphur, weathering, bacteria, temperature, ventilation, conductivity and ozone (Falcon (2004), Gurdal & Bozcu (2011), Kaymakci & Didari (2002), Phillips *et al.,* (2011) and Pone *et al.,* (2007)).

Particle size and porosity affect the spontaneous combustion liability of coal (Itay *et al.,* (1989) and Mastalerz *et al.,* (2010)). Coal with varying large particle sizes experience lower spontaneous combustion liability than coal with a slight range of particle distribution (Hansel *et al.,* 2004). This is because large particles possess the influence of reduced specific surface area and bulk density. Kucuk *et al.* (2003) and Ren *et al.* (1999) indicated that the spontaneous combustion liability of coal increases with a decrease in particle sizes. Wang *et al.* (1999) indicated that the rate of coal oxidation with high porosity is kinetically controlled. In most cases, coal does not self-heat under insufficient oxygen. Granular coal is reactive and undergoes spontaneous combustion rapidly but a combination of different coal particle sizes is highly liable to spontaneous combustion (Kucuk *et al.,* 2003). The extent of self-heating could be based on a complex relationship between various intrinsic and extrinsic factors of coal (Uludag, 2007)). Nugroho *et al.* (2000a) confirmed that particle size is a cause of coal

spontaneous combustion and the self-heating characteristics of high-rank coal is dependent on particle sizes. Nugroho *et al.* (2000b) found that coal mass of different particle sizes is more liable than those with a smaller particle size. Bhat & Agarwal (1996) indicated that the coal particle size may influence heat loss through convection and the heat produced by the mass transfer coefficient, which controls the degree of moisture condensation. Kim (1977) confirmed that the particle size has an inverse relation to the spontaneous combustion liability of coal.

The influences of moisture content and oxygen in the air on the spontaneous combustion liability of coal have been studied (Akgun & Arisoy (1994b), Arisoy & Akgun (2000), Bhattacharyya (1971b), Bhat & Agarwal (1996), Didari (1988), Panigrahi *et al.,* (2000) and Ren *et al.,* (1999)). Some previous studies found that a low moisture content may support spontaneous combustion liability, while a high moisture content impedes spontaneous combustion (Beamish & Hamilton (2005) and McPherson (1993)). Ren *et al.* (1999) indicated that the moisture content of coal has a high influence at the initial stages of self-heating. The unusual wetting and drying of coal on stockpile surfaces accelerate spontaneous combustion due to absorption or adsorption (Clemens & Matheson, 1996). Akgun & Arisoy (1994a) investigated the effects of air humidity and inherent moisture content of coal affecting spontaneous combustion. It was observed that when dry air is transported over fairly wet coal, moisture is removed from the coal, thus causing a decrease in temperature. Although, when misty air enters dry coal, temperature increases due to moisture adsorbed from the air (Akgun & Arisoy, 1994a). The wetting and drying of coal particles promote heat exchange within a coal mass. The drying of coal involves temperature changes which influence the heat balance in the oxidation of a coal mass and causes self-cooling. The wetting of coal is an exothermic reaction which gives off heat to aid self-heating (Lyman & Volkner, 2001). During the wetting and drying of coal, heat is transferred between coal and oxygen in the air. The influence of moisture alters the rate of oxidation. An increase or a decrease in heat transfer affects the oxidation rate and moisture content (Lyman & Volkner, 2001).

Panigrahi & Sahu (2004) indicated that as ash content increases, the spontaneous combustion liability of coal decreases. Beamish & Blazak (2005) indicated a negative relationship between R_{70} (an Australian test to predict the spontaneous combustion liability of coal) and ash contents for low to high rank coal. Sia & Abdullah (2012), Siavalas *et al.* (2009) and Zivotic *et al.* (2013) reported in their studies that a high ash content could be due to the peat depositional environment where the condition of flooding of the paleomire occurs intermittently during

deposition. Blazak *et al.* (2001) reported that as ash content increases, spontaneous combustion liability decreases due to the reduction in the amount of inert and organic material acting as a heat sink. Studies reported by Falcon & Ham (1988), Hagelskamp & Snyman (1988), Snyman & Botha (1993) and Van Niekerk *et al.* (2008) indicated that the air-dried ash content of South African coal varies between 21.3wt.% and 38.8wt.%. Choudhury *et al.* (2007) found that some Indian coal have ash content greater than 45wt.%.

Studies reported shows that the carbon content of coal varies between 40wt.% and 52wt.% (Roberts, 2008)**,** 56.7wt.% and 69.6wt.% (Czaplicki & Smolka, 1998), 49.0wt.% and 58.23wt.% (Department of Minerals and Energy South Africa, 2004). Hydrogen content in coal ranges between 3.07wt.% and 3.20wt.% (Roberts, 2008), 2.60wt.% and 3.13wt.% (Department of Minerals and Energy South Africa, 2004) and nitrogen content varies between 0.76wt.% and 1.13wt.% (Czaplicki & Smolka, 1998) and 0.56wt.% and 1.44wt.% (Department of Minerals and Energy South Africa, 2004).

Jing *et al.* (2015) indicated that the self-heating potentially causing spontaneous combustion of coal reduces as oxygen concentration increases. This was confirmed in an experiment conducted under different oxygen concentrations of 5, 10, 15, 17, 21 and 30%. The degree of oxygen within a coal contributes significantly to the likelihood of spontaneous combustion. The low-temperature oxidation of coal could be caused by oxygen adsorption from the air or can be from natural oxygen within a coal mass. The rate of airflow is considered to be the main significant factor influencing spontaneous combustion liability of coal (Kim (1977) and Mastalerz *et al.,* (2010)). This factor is very complex because it provides both the oxygen needed for the oxidation process and disperse the heat generated. The airflow conditions determine the rate of heat transfer within a coal mass and are less important at low flow rate than at high flow rate (Adamski, 2003). The convective heat losses on the coal surface are measured by the surface temperature and temperature distribution within the coal mass. This heat transfer processes are dependent on the temperature distribution and geometry of the coal mass (Cliff *et al.,* 2014).

Walters (1996) investigated that airflow supplies sufficient oxygen for low-temperature oxidation to occur and disperse the heat produced by oxidation. An extremely high airflow offers indefinite oxygen and disperses heat effectively, while a low flow rate limits the required volume of oxygen for oxidation and allows self-heating within coal seams. Schmal *et al.* (1985) indicated that spontaneous combustion occurs in stockpiles when convection provides

sufficient oxygen, while heat is eliminated by conduction and not by convection. The movement of wind on coal surfaces has two impacts that increases thermal conductivity. In the first case, the increase in airflow assists with the provision of oxygen to the zone of reaction, thereby causing a greater rate of combustion. On the other hand, the heat transfer at the low-temperature oxidation zone makes the system less liable to spontaneous combustion (Brooks & Glasser, 1986).

The mineral matter within a coal have been evaluated by a number of researchers (Given & Miller (1985), Mastalerz *et al.,* (2010), Siddigui *et al.,* (2009), Stach *et al.,* (1975), Suarez-Ruiz & Crelling (2008), Ward & French (2005) and Winburn *et al.,* (2000)) based on the nature of coal. Among the number of minerals that commonly exist in coal includes pyrite, muscovite, quartz, kaolinite, illite, oxides (hematite and magnetite) and carbonates (calcite, dolomite and siderite). The presence of different organic and inorganic matter within a coal to prevent the start of spontaneous combustion had been shown in several studies (Beamish & Arisoy (2008a), Beamish *et al.,* (2005), Humphreys *et al.,* (1981) and Smith *et al.,* (1988)). Beamish & Arisoy (2008a) indicated that the distribution of mineral matter in coal impedes low-temperature oxidation due to their physical and chemical influences. The mineral matter in coal consists of a number of mineral constituents which differs in mineral composition and quality from one coal seam to the other. The mineral matter may be in the form of mineral phases and mineral species of different particle sizes. The species are chemically complex by nature of the mineral composition. The quantity of trace elements in coal has a great influence on the environmental, economic and spontaneous combustion liability (Finkelman & Gross (1999) and Swaine & Goodarzi (1995)). The trace elements in coal may exist as organic and inorganic matter (Cairncross *et al.,* (1990), Radenovic (2006), Swaine (1990) and Wagner & Hlatshwayo (2005)). The characteristics of trace elements during the utilization and combustion processes of coal can be controlled by the concentration of trace elements and their frequence of occurrence (Finkelman (1994), Finkelman (1995), Swaine (2000) and Xu *et al.,* (2003)). The occurrence of trace elements in mineral matter within coal influences the spontaneous combustion liability (Finkelman (2004), Mardon & Hower (2004) and Swaine (2000)).

The presence and distribution of sulphur content in coal have been reported by various researchers (Casangrade (1987), Chandra *et al.,* (1980), Mastalerz *et al.,* (2010) and Teichmuller & Teichmuller (1982)). Studies have shown that the total sulphur in coal varies between 0.93 and 3.35% (William, 1994), 0.4 and 1.29% (Wagner & Hlatshwayo, 2005), 0.43

and 0.63% (Czaplicki & Smolka, 1998), 1.75 % (Gryglewicz *et al.,* 2002), 1.47 and 1.56 % (Roberts, 2008), 19.58% (Marinov *et al.,* 2005), 0.59 and 9.45% (Hsieh & Wert, 1985), 0.74 and 1.23% (Department of Minerals and Energy South Africa, 2004), and 5.4 to 15.1% (Olivella *et al.,* 2002) for South African coal and some coal around the globe. The sulphur content in deep coal seams has been considered to be higher than shallow coal beds (He *et al.,* 1999). Excess amount of sulphur in coal promotes the event of self-heating leading to spontaneous combustion and influences the mine environment. The presence of high sulphur contents in coal has been reported in countries with massive coal resources (Chandra *et al.,* 1980). Chandra *et al.* (1980) indicated a high sulphur content in the upper seam of Assam coal more than the underlying seams. The occurrence of sulphur in coal can be divided into organic and inorganic sulphur. The organic sulphur together with the macromolecular structure combined in the form of covalent bonds with a complex structure and are hard to distinguish. The inorganic sulphur in coal occurs in the form of pyrite and sulphate. Studies reported on forms of sulphur shows that the pyrite content in coal varies between 0.11 and 1.15% (William, 1994), 0.30% (Marinov *et al.,* 2005), 3.9% (Hsieh & Wert, 1985) and 0.3 and 3.3% (Olivella *et al.,* 2002) for South African coal and some coal around the globe. It was reported that sulphate content in coal varies between 0.7 and 2.17 % (William, 1994), 0.03% (Gryglewicz *et al.,* 2002), 0.15% (Boudou *et al.,* 1987), 0.59% (Marinov *et al.,* 2005) and 0.3 and 7.6% (Olivella *et al.,* 2002). Previous studies indicated that the organic sulphur content in coal varies between 0.12% (William, 1994), 1.18% (Gryglewicz *et al.,* 2002), 1.7% (Boudou *et al.,* 1987), and 0.26 - 4.30% (Hsieh & Wert, 1985). Gupta *et al.* (1977) indicated moisture and sulphur content as factors that initiate the spontaneous combustion liability of coal stockpiles.

The amount of pyritic and sulphate sulphur varies in coal. Pyrite as the major constituent of inorganic sulphur has significant influences on the spontaneous combustion liability of coal (Mastalerz *et al.,* (2010), Rumball *et al.,* (1986) and Stach *et al.,* 1975)). Pyrite reacts with oxygen in the presence of water to form hydro-peroxide (H_2O_2) and therefore, initiates oxygen (Huffman *et al.,* (1985), Huggins *et al.,* (1983) and Nordstorm (1982)). Pyrite with a concentration above 2% has been reported to promote the spontaneous combustion liability of coal (Bhattacharyya, 1971a). Beamish & Beamish (2012) indicated that the type of pyrite within coal determines whether rapid self-heating would occur but not the volume of pyrite. Beamish *et al.* (2012) confirmed that coal consisting of high pyritic sulphur do not reach thermal runaway fast enough in a dry state compared to in a moist state. The reaction for pyrite oxidation is expressed in Equation 2.8 by Lain (2009).

$$Fe_2S + 7O_2 + 16H_2O = 2H_2SO_4 + 2FeSO_4.7H_2O + 1321kJ \quad (2.8)$$

The reaction above is exothermic and occurs at low temperature. The heat generated from the reaction is double that of coal with the same oxygen (Garcia (1999) and Martinez *et al.,* (2009)). Hence, the presence of pyrite is a contributory factor affecting spontaneous combustion liability of coal. Chandra *et al*. (1980) found that the distribution of sulphate sulphur in Assam coal remain the same from the lower to the upper seam. Dutta *et al.* (1983) indicated an increase in the percentage of total sulphur with a decrease in the sulphate sulphur content of coal. Sulphate sulphur in coal is known to be of a smaller amount (Iyenger *et al.,* 1959) compared with the other forms of sulphur (Goswami, 1982).

Chandra *et al.* (1991) indicated that the crossing point temperatures (XPT) and volatile matter decrease with increasing coal rank. Pattanaik *et al.* (2011) showed a continuous increase in spontaneous combustion liability with a consistent decrease in the degree of coalification. A study indicated an increase in volatile matter content up to a value of 35% with a decrease in XPT (Raju, 1988). Previous researches used volatile matter and selected intrinsic factors of coal to predict the spontaneous combustion liability (Banerjee (2000) and Panigrahi & Saxena (2001)). Kim (1977) indicated that the effects of blasting and improper cleaning of highwalls can contribute to the event of spontaneous combustion. The exposure of open pit walls to the air and run-off-mine for long periods before loading and poor clean-up techniques may result to self-heating. Geological discontinuities such as faults and fractures in the overburden allow water and oxygen to infiltrate within a coal seam and cause self-heating (Kim, 1977).

Detailed petrographic studies of coal have been carried out by Dai *et al.* (2003), Falcon (1978), Falcon (1987), Holland *et al.* (1989), Mastalerz *et al.* (2010), Snyman & Botha (1993), Stefanova *et al.* (2003) and Steyn & Smith (1977). The concentration of the liptinite group of South African coal is very low. A similar study is reported on some Indian coal (Choudhury *et al.,* 2007). Holland *et al.* (1989) examined the major macerals and mineral matter of Basal four seams in the south of Middelburg, Witbank Coalfield and found that the coal seams have an inertinite content greater than 55% in most cases, but usually ranges from 20% to 80%. The major macerals (inertinite, liptinite and vitrinite) are liable to self-heating and weathering with respect to time, temperature and environmental conditions. According to Beamish & Blazak (2005), Falcon (1989) and Ivanova & Zaitseva (2006) vitrinite is known to be more liable among the major macerals, while the inertinite and liptinite macerals are rarely liable to spontaneous combustion. Misra & Singh (1994) indicated that a high amount of inertinite

maceral groups may accelerate spontaneous combustion liability because of their porosity and affinity to absorb gas. The type and quantity of macerals and the degree of coalification have been shown by several researchers to be reliable factors to predict the spontaneous combustion liability of coal (Benfell *et al.,* (1997), Chandra & Prasad (1990) and Morris & Atkinson (1988)). The types of maceral found in structured inertinite (semifusinite, resin-inertinite and fusinite, bands of inertinite) have larger microscopic and sub-microscopic openings which may provide pathways for the ingress of oxygen in the air within coal seams than in vitrinite (Didari (1988) and Falcon (1987)). It was reported that the amount of exinite, liptinite and vitrinite contents influence the spontaneous combustion liability of coal and, with an increase in the vitrinite reflectance, the liability towards spontaneous combustion gradually increases (Avila (2012), Chandra *et al.,* (1991) and Pattanaik *et al.,* (2011)). The difference in the degree of coalification is found to be influenced by temperature variation, periods of exposure and the conditions of the surrounding environment. Kruszewska & du Cann (1996) and Pattanaik *et al.,* (2011) reported that as the oxidation rate increases, the vitrinite content increase and as the oxidation rate decreases, the degree of coalification increases.

Studies reported by Ogunsola & Mikula (1990), Raju (1988), Ren *et al*. (1999) and Smith *et al.* (1988) indicated that the higher the vitrinite reflectance, the lower the spontaneous combustion liability, while the lower the degree of coalification, the higher the spontaneous combustion liability. A study reported by Ogunsola & Mikula (1990) indicated that spontaneous combustion should not be based only on coal rank but other properties should be evaluated. Singh *et al.* (2013) reported that spontaneous combustion liability varies between bands of coal seams, a seam of low-rank coal may be resistant to oxidation, while higher rank coal may be more liable to spontaneous combustion. Kim (1977) indicated that as coal rank decreases, the liability toward spontaneous combustion increases.

It has been generally assumed that only coal can cause spontaneous combustion. However, limited studies have been reported on coal-shale to be capable of undergoing self-heating potentially leading to spontaneous combustion (Kim & Chaiken (1990), Restuccia *et al.,* (2017) and Rumball *et al.,* (1986)). This study provides a detailed experimental investigation of selected intrinsic properties that contribute to the spontaneous combustion liability of coal and coal-shale. The influence of these properties on coal and coal-shale collected between selected bands of coal seams and higwalls were investigated. This was done to evaluate the causes,

establish relationships between the spontaneous combustion liability index and intrinsic properties and recommend reliable methods to minimize the occurrence.

2.5 Prediction methods for spontaneous combustion liability of coal

The measure of the spontaneous combustion liability necessitates the identification of various coal based on their inherent properties since no specific standard method is recommended to evaluate the spontaneous combustion liability of coal. The techniques described below are generally used to measure the spontaneous combustion liability or self-heating characteristics of coal.

(i) Differential scanning calorimetry (DSC): Studies reported by Mahajan & Walker (1971) and Mohalik *et al.* (2009) used DSC to measure the spontaneous combustion liability of coal. This method determined the changes in energy inputs provided to a substance and a reference material as a function of temperature when both materials are kept at a controlled temperature. The experimental investigations of this technique on coal spontaneous combustion liability were restricted to the evaluation of only four Indian coal samples (Mahajan & Walker, 1971). This method indicated that there was no uniformity on the laboratory variables reported by different studies in carrying out the DSC (Mohalik *et al.,* 2009). Mohalik *et al.* (2010) suggested a correction of the adjusted experimental variables used. The method does not give a better understanding to correlate the liability between two coal samples to spontaneous combustion.

(ii) Thermogravimetric analysis (TGA): This method has been used to predict coal known to be liable to spontaneous combustion (Vaan Graan & Bunt, 2016). This method estimates the loss in weight of coal samples at variable temperatures due to self-heating. A specific weight of coal sample is heated through a programmed heating process and plotted against the time/temperature. The results are termed thermogravimetric (TG) curves. The created TG curve is referred to as the differential thermogravimetric (DTG) curve and is the difference between the curve of the coal and the curve of the inert material.

(iii) Russian U index: This method estimates the amount of oxygen absorbed by individual coal samples over 24hrs. The gases obtained under experimental conditions may be quantified by evaluating the gas composition. Banerjee (2000) reported that the oxygen absorbed during testing is directly proportional to the spontaneous combustion liability of coal. The constraint in this method is that the volume of oxygen absorbed by a coal sample during testing is not

reproducible when the experiment is repeated on the same coal. This method fails to provide the specific measure of spontaneous combustion liability for coal with a high moisture content.

(iv) Differential thermal analysis (DTA): Reports by Whitehead & Breger (1950) and Mohalik *et al.,* (2009) have used this technique to predict the spontaneous combustion liability of coal. The method involves heating a small coal sample at a constant rate and keeping records of the temperature differences within the material and a similar inert material as a function of temperature existing in the inert medium. This reveals the changes in the physical and chemical properties of a material at the particular temperature and the properties of the material used (Mohalik *et al.,* (2009) and Whitehead & Breger (1950)). It was found that there was no uniformity on the laboratory parameters selected in the use of this method.

(v) Crossing point temperature (XPT): Many studies have used this method to measure the spontaneous combustion liability index of different coal with respect to their ignition temperature which is similar to the XPT (Gouws (1987) and Humphreys (1979)). This method has been used as a liability index in countries such as South Africa, Turkey, Poland and India to categorize the spontaneous combustion liability of different coal. The experimental tests involve heating coal in an oxidized condition to cause low-temperature oxidation either at an automatic heating rate or at a certain temperature from the ambient temperature to the ignition point temperature of the coal. Studies reported using this method indicated that coal with a lower liability index have higher XPT and vice versa. Humphreys (1979) reported that this method is not suitable because it has not considered some inherent properties of coal and the design of the testing equipment. Barve & Mahadevan (1994) indicated a relationship between ash (dry), moisture and XPT as seen in Equation 2.9 which is not in-line with Humphrey's opinion.

$$XPT = 168.8 - 10.3M + 0.12A + 0.69M^2 - 0.06MA + 0.01A^2 \quad (2.9)$$

where, **M** is the moisture content, **A** is the ash content and **XPT** is the crossing point temperature.

(vi) X-Ray Diffractometer: Limited studies have been conducted by examining the quantity and identification of minerals with the use of X-ray diffraction method in a specific coal to predict spontaneous combustion liability (Gong *et al.,* (1998), Kelemen *et al.,* (1990) and Kok (2008)). The results obtained from this method were compared with the results obtained from petrographic properties (macerals) and other thermal techniques to confirm the validity of this

method. Cole (1975) indicated that the variations in iron minerals within coal during low-temperature oxidation may be caused by the catalytic reactions of the iron minerals with oxygen. This technique showed that pyrite (iron-content mineral) among other mineral matter may be easily altered during coal oxidation and the oxidation alteration of pyrites vary with the forms of sulphur and its mineralogy (Ribeiro *et al.,* 2016)**.**

(vii) Olpinski index method: This technique is generally used in Poland to categorize the spontaneous combustion liability of coal. The liability index is referred to as the Szb Index. A pellet of powdered coal (0.4g) is heated at a constant rate in a quinolone steam bath with the flow of air through the coal pellet. The time against the temperature curve is recorded until a temperature of 235^{o}C is reached. The temperature increase of the coal sample at this stage is used to measure the spontaneous combustion liability (Banerjee, 1985). This method has been used to provide a better definition of the spontaneous combustion liability index of Indian coal by establishing relationship between XPT, flammability temperature (FT), wet oxidation potential (WOE), proximate and ultimate analysis (Nimaje & Tripathy (2016), Nimaje & Tripathy (2015) and Nimaje *et al.* (2013)).

(viii) Adiabatic calorimetric method: This method is usually used in South Africa, New Zealand, Australia, UK and USA to reproduce the original condition of self-heating characteristics of coal (Cliff *et al.,* 1996). The rate of temperature increase, ignition temperature and the kinetic constant of coal (rate of chemical reaction at a given temperature of coal) are used to determine the proneness of coal to spontaneous combustion (Ren *et al.,* 1999). This technique involves placing a coal sample in a reaction vessel either in an adiabatic oven or oil bath, such that the heat is not dissipated from the vessel. The temperature of coal in the reaction vessel is controlled at particular intervals relative to the increase in the coal temperature. The reacting air or oxygen flows through the reaction vessel. A study reported by Cudmore (1988) investigated the effect of air humidity and moisture under adiabatic conditions but not the extent to which it affects the spontaneous combustion liability of coal.

A number of studies carried out showed relationships between this method and some inherent properties of coal (proximate and ultimate analysis and petrographic analysis) to re-examine previous spontaneous combustion records (Moxon & Richardson (1985) and Ren *et al.,* (1999)). The test is referred to as R_{70} in New Zealand and Australia (Beamish *et al.,* (2000), Beamish *et al.,* (2001) and Humphreys *et al.,* (1981)). Some constraints (such as time to

complete a test, design of reaction columns and required amount of samples to be used, exact particle sizes and the required flow rate of air/oxygen) have been observed in using this method.

Gouws *et al.* (1991) created an adiabatic calorimeter that monitors and keeps accurate records of data with respect to the rising temperature and incubation mode of coal samples with the use of a computer. The apparatus identified the various self-heating potentials such as minimum self-heating temperatures, total temperature rises, kinetic constants and initial rates of heat to allow the propensity of coal to self-heating to be initiated. The method created a reliable spontaneous combustion liability index from the results of the DTA. The XPT are determined with the adiabatic calorimeter and are referred to as the Wits-Ehac Index as seen in Equation 2.10.

Wits-Ehac Index = (Stage II slope/XPT) * 500 (2.10)

This index investigates two characteristics, the stage II slope and the XPT. The index is based on the fact that coal highly liable to spontaneous combustion have a steeper stage II slope and a lower XPT than coal less liable to spontaneous combustion. When the temperature difference between the inert material and the coal sample is plotted against the inert temperature, the part of the graph where the coal is heating faster than the inert sample (where an exothermic reaction occurs) is referred to as Stage II. According to Wade *et al*. (1987), coal with a spontaneous combustion liability index below 3 are known to be low risk, 3 to 5 medium risk, and coal with values greater than 5 are highly liable to spontaneous combustion. Studies reported by Genc *et al.* (2018) and Genc & Cook (2015) used this index to predict spontaneous combustion in the South African Coalfields.

2.6 National and global position of research on spontaneous combustion of coal

The international findings of the industries, researchers and scientists related to the present study are briefly described below:

Tideswell & Wheeler (1925) investigated the reaction of coal and fusain circulated with oxygen in a closed system. The study indicated that the oxidation rate of fusain slightly increased with temperature than coal.

Schmidt *et al.* (1936) investigated an increase in the total weight of coal samples during the oxidation reaction. It was found that the weight of oxygen left in the coal is higher than the weight of carbon and hydrogen dispersed as gaseous products.

Mehta *et al.* (1952) investigated the influence of oxygen absorption and adsorption on some Indian coal. The study indicated that coal known to be liable to spontaneous combustion has the highest rates of oxygen absorption. Furthermore, a linear relationship between the total oxygen absorbed in 40-hrs and the bed moisture contents occurred.

Nandy *et al.* (1972) investigated the differences in the XPT with the percentage of volatile matter, moisture content and the oxygen content of coal. It was indicated that the XPT of the coal samples decreases as each parameter increases.

Chandra *et al.* (1983) carried out a preparatory study on the rate of development of fire with reference to spontaneous combustion liability in different coal seams of the Raniganj coalfield. The vitrinite reflectance, amount of vitrinite and exinite macerals of the coal seams affects the spontaneous combustion liability of the coal seam. The liability towards spontaneous combustion decreases consistently as the coalification increases. It was found that the spontaneous combustion liability of coal could be linked to coalification.

Nordon & Bainbridge (1984) confirmed that the distribution of free gaseous oxygen in the coal stockpile agrees with an existing model for predicting the spontaneous combustion liability of the stockpile. A simplified, isothermal model of the study predicted a hyperbolic cosine distribution, termed the Thiele Modulus. The model depends on the stockpile characteristics such as coal properties, size and shape of the stockpile, intra-particle porosity, oxygen diffusivity, coal density and reactivity towards oxygen. The Thiele Modulus is determined from two measurements of oxygen concentration at different depths of a stockpile. It was indicated that the changes in coal reactivity caused by prolonged exposure to atmospheric oxygen were reflected in the magnitude of the Thiele Modulus.

Boyapati *et al.* (1984) developed a suitable and basic experimental technique to evaluate the relative weathering liability of coking coal in the laboratory. Three typical Australian coking coal were used for the study. The weathering characteristics of the coal samples were examined in adjacent similar stockpiles under testing conditions. A gas flow test was applied to investigate the relative weathering conditions. The results of the experimental stockpiles identified oxidized coal by the rate of degradation in coke strength. It was found that the relative weathering propensity of coal never increased as the particle size decreased.

Mahadevan & Ramlu (1985) determined the spontaneous combustion liability of Indian coal based on the correlation between spontaneous combustion test results and the mine

environment index (including geological and mining factors). The mine environment index and risk were evaluated based on Indian geological and mining conditions. The technique was successfully adopted to evaluate the risk of spontaneous combustion liability of coal. The study indicated that the results of the risk classification agreed with the field experience.

Riley *et al.* (1987) investigated the variables which may give rise to the spontaneous combustion in barges. The determinants that cause coal to self-heat in the barges were indicated with a data bank consisting of analytical data and barging information on 2283 barges. Tests that were carried out on the two barges over two summer periods in the Ohio and Mississippi rivers were studied. Fifteen barges of coal were in the first test and 10 barges in the second test. Four different river ports were used to evaluate high volatile bituminous coal in the barging experiment. Two separate loading procedures were used in the study, loading with a diffuser and single chute loading. The technique of compaction shows a notable variation between the results obtained with closely one-third of the temperature readings (105^{o}F) above the lightly compacted coal. The variation in temperature range can be due to the uniform particle size distribution formed while loading with the diffuser. It was indicated that the uniform distribution of coal particle sizes produced a steady airflow in coal which leads to low spontaneous combustion. Furthermore, compacting coal in barges provides significant safety against spontaneous combustion.

Singh & Demirbilek (1987) carried out statistical analysis to evaluate the relationship between intrinsic properties and spontaneous combustion tests of 47 coal samples. The prediction of the spontaneous combustion liability was established with respect to the initial rate of heating and total temperature rise in an adiabatic oxidation test. A multiple regression statistical analysis between spontaneous combustion tests and 13 independent variables produced a set of practical equations to predict the spontaneous combustion liability of coal. The equations derived by subdividing the data set based on the coal rank classification enabled the determination of coal liable to spontaneous combustion. It was found that the contribution of various coal intrinsic properties to spontaneous combustion was evaluated using the isolated factor analysis methods.

Tognotti *et al.* (1988) described a simple experimental technique to determine the ignition temperature of different cylinder-shaped coal beds of South African coal. The application of thermal ignition models in interpreting and extrapolating the small-scale results was reported. Measurement of the rate of heat generated in isothermal conditions was reported and compared with the results of ignition tests. The coal sample was found to undergo spontaneous

combustion in small-scale tests at a temperature of between 120 and 220^{o}C. The thermal ignition model gave a good correlation for critical ignition temperature of various coal sizes. It was found that the correlation proved to be relatively insensitive to the reaction (oxygen inter-particle diffusion) and the physical change in the material (reactant consumption). The effect of coal particle sizes indicated the existence of intra-particle diffusion limitation. It was found that comparison between ignition and thermal data yields a satisfactory agreement in predicting the critical dimension by means of a thermal ignition model.

Morris & Atkinson (1988) attempted to establish a comprehensive study of seam factors that affect spontaneous combustion liability of coal. The study was used in conjunction with earlier works of Morris & Atkinson (1986) to support the phenomenon of spontaneous combustion as a collective effect of seam factor, geological factor and mining factor.

Brooks *et al.* (1988) formulated a mathematical model to measure spontaneous combustion liability of coal stockpiles. The model was derived by breaking up the coal lumps into three different segments. The model incorporated a-one dimensional chimney region as used by Brooks & Glasser (1986), where reaction flow and heat transfer occur. It was assumed that the gas entering the bottom of the chimney was depleted by the flow of oxygen through the entire coal bed. The depletion was modelled with the use of a residence time distribution approach. The steady state equations describing the system was assumed and solved. The study classified stockpiles as SAFE, UNSAFE or CONDITIONALLY SAFE. The particle size at which the average temperature in the coal bed assumed its worst depth was determined. Furthermore, the parameters affecting the safety of the coal bed, such as rate of reaction, coal particle size and the coal bed porosity were indicated.

Itay *et al.* (1989) determined the low temperature (25 to140^{o}C) oxidation rate of a single coal in a number of ways. The coal used was a sample of the middling from the Grootegeluk coal mine. A well-enclosed vessel was used as an approach to follow the temperature-time history of the coal sample while at the same time the gas analyses were evaluated. It was observed that oxygen absorption was the dominant cause of the exothermic reaction. The portion of gaseous products formed (CO, CO_2 and H_2O) were at lower temperatures and smaller than the amount of oxygen absorbed. The existence of CO_2 and CO hindered the chemical reaction, while the presence of water appeared as a catalyst. An overall oxidation technique similar with experimental findings was proposed due to the experimental results.

Kim & Chaiken (1990) investigated the spontaneous combustion liability of carbonaceous shale, coal and roof coal through a revised model of a differential thermal analyser. The apparatus was developed to evaluate the increasing influence of heat changes within the shortest time. The degree in which the sample temperature surpasses the furnace temperature was presumed to be an indication of spontaneous combustion. Three ovens were used to evaluate the XPT and a composite index was obtained. The extent of self-heating changes was determined by the release of hydrocarbon and analysis of O_2, CO, CO_2 depletion. The results of coal and other carbonaceous materials generated a large data which have not been reported to determine spontaneous combustion in Coalfields. The study indicated that experimental data could be linked to possible self-ignition.

Gouws *et al.* (1991) created an adiabatic calorimeter to evaluate the spontaneous combustion liability of coal. The parameters of self-heating potential such as the total temperature rise, the starting rate of heating, the minimal self-heating temperature and the kinetic constants were examined. The calorimeter was designed to replace the ignition temperature apparatus and simplify the test procedure. It has the advantage of very little preparation time. It is fully automatic, requiring no supervision at all. Results obtained from adiabatic tests for some selected coal were correlated with the results obtained from the XPT and DTA tests. This was done with a view to create a reliable spontaneous combustion liability index.

Chen (1992) investigated the effect of moisture due to temperature increase in a coal stockpile with the use of a simplified one-dimensional differential equation that described the mechanism of self-heating. An analytical solution of the maximum temperature was reached with the use of well-established coefficients. The solution suggests that, if the saturation of gas stream in a coal stockpile moisture is assumed, the estimated temperature of the stockpile may be below 100^{o}C. The relative humidity of the gas stream drops as the estimated maximum temperature increases up to 100^{o}C. It was found that the analytical solutions supports the use of the equilibrium relationship between the relative humidity of the gas and the coal moisture content into the model.

Sen (1992) evaluated the properties of Indian coal using petrographic composition. It was found that the content of vitrinite, exinite and vitrinite reflectance were used as parameters to predict spontaneous combustion liability.

Carras & Young (1994) evaluated and identified the limitations of the absorption tests used to predict the spontaneous combustion liability of coal. The direct measurement of oxygen intake was noticed to be the easiest to interpret the physical terms. This provides a reliable method to compare the spontaneous combustion liability of different coal. Combined with an isothermal calorimeter, the absorption method enables the use of an empirical oxidation law for a particular coal. It was found that the spontaneous combustion liability of coal depends on several properties and no single property can be used to evaluate the spontaneous combustion liability of coal.

Akgun & Arisoy (1994a) examined the effects of particle size on the spontaneous combustion liability of coal. The study investigated coal with different particle sizes (between 2 and 50mm in diameter) that was oxidized in a cylinder of 3m in length and 0.3m in diameter. A critical range of particle was observed below which self-heating leads to flaming combustion. The Arrhenius equation was applied to the experimental results. It was observed that the apparent activation energies decrease (52.64 to 33 kJ/mol) with increasing particle size for the selected ranges of particle sizes. The results of temperature and oxygen consumption of the coal bed indicated that the influence of particle size on the oxidation reaction occurred at a greater depth. The determination of the operating regime for the coal-oxygen reaction was achieved by defining an empirical exponent for the particle diameter in the rate equation. It was found that the empirical exponent indicated the oxidation regime for a change from chemical to pore diffusion at successively higher temperatures.

Carras & Young (1994) analysed the degree of some numerical models established to assess the spontaneous combustion liability of coal and mine wastes. The frequently used industrial tests to anticipate spontaneous combustion liability of coal were investigated and their limitations were identified. It was found that further investigation needs to be conducted to determine the influence of water on the oxidation of coal and other carbonaceous materials. Furthermore, a validation of numerical models on the commercial size of coal stockpile was required.

Arief & Gillies (1995) modelled the characteristics of coal under conditions that may cause self-heating with a developed apparatus. The apparatus considered the influence of moisture content and coal particle sizes toward spontaneous combustion liability under field conditions. The study requires the needs to investigate the impacts of varying airflow conditions on coal oxidation in order to obtain different degrees of compaction.

Bhat & Agarwal (1996) developed a mathematical model that reviews moisture deposition and removal within a coal particle. The model developed investigated the effect of moisture migration on the spontaneous combustion liability of coal. The study indicated that partial wetting of coal employs two competing influences on oxidation processes. In the first case, as a portion of the coal fills up with liquid, the oxidation rate becomes negligible. This is because the oxygen dissolved in the moisture before infiltrating into the coal surface. Secondly, condensation caused the release of the latent heat of vaporization. This heat elevates the coal particles temperature and increases the oxidation rate in the dry region of the coal. The degree of the rate of these competing factors determines the possibility of spontaneous combustion to occur. A criterion was created to determine the conditions under which condensation of moisture may result to spontaneous combustion based on the developed model. It was found from field observations that moisture condensation can abate the likelihood of spontaneous combustion. It was concluded that the model is the first mathematical equation that correlates moisture interaction to the possibility of spontaneous combustion.

Krishnaswamy *et al.* (1996) created a two dimensional model to predict the spontaneous combustion liability of coal stockpiles by neglecting the impact of moisture content. The influences of bed porosity, side slope, wind velocity, coal reactivity and bed particles size were investigated. A correlation was established for safe long-term storage of coal stockpiles. It was indicated that wind-driven forced convection is the major cause of airflow in a stockpile.

Vance *et al.* (1996) studied the influence of moisture content on the spontaneous combustion liability of coal under oxygen conditions. A sample of the Renown coal seam was collected from the Huntly East mine in New Zealand. The coal lumps were reduced to 210µm size in three stage operations (such as jaw and cone crusher and hammer mill) in order to simulate the behaviour of the freshly crushed coal particles. The self-heating was maximum at a moisture content of approximately 7% and most pronounced at temperatures below 80^{o}C. The study indicated that a maximum oxidation process begins at that moisture content in the same temperature range.

Pis *et al.* (1996) evaluated the spontaneous combustion liability of fresh and oxidized coal with the use of a DTA. Six (6) different coal samples which ranges from a high bituminous coal to semi-anthracite were used for this study. The coal samples were pulverised to particle sizes of 150µm. The coal oxidation was carried out at 200^{o}C over different periods of time (0 to 72hrs) in an oven with forced-air circulation. The self-heating and temperature at the end of

combustion increased with the coal rank. It was found that an increase in the self-heating temperature and a decrease in the temperature at the end of spontaneous combustion occurred due to the oxidation process.

Ibarra & Miranda (1996) studied the process of natural oxidation of two low-rank coal that were exposed to the atmosphere with the use of a Fourier Transform Infrared (FT-IR) spectroscopy. The study was conducted on two Spanish low-rank high sulphur coal (Endesa and Mequinenza). A considerable sample size of over 2500T of each coal was stockpiled for 11 months at the Andorra coal-fired power station. Samples for analysis were taken from the piles at different time intervals from zones where self-heating and ignition were observed. The variables deduced from the curve fitted bands of selected regions of the FT-IR spectra allowed the evolution of the structural changes in low-rank stockpiled coal to be known. The carboxyl groups of the samples tend to increase slightly under conditions of low pile activity. The aliphatic hydrogen content of the samples tends to decrease with increasing the time of storage. The structural changes are more important when oxidation and self-ignition processes take place. Aliphatic structures decreased drastically. The net production of the oxygen-containing structure was observed. It was found that the aliphatic hydrogen content evaluated from the integrated absorbance measurements and the CO/H ratio is sensitive in detecting signs of weathering at low levels of activity.

Panigrahi *et al.* (1997) conducted experimental tests to evaluate the Russian U-Index. A sample of coal from ten (10) different sections of the Jharia Coalfields were collected and analysed with the same method. The XPT of the coal samples were investigated and the carbon, hydrogen, nitrogen and sulphur contents for the coal samples were studied using Fenton's method of ultimate analysis. The Russian U-Index shows a similar relationship with these elements as shown by the XPT with respect to spontaneous combustion.

Chen & Stott (1997) investigated the oxidation rates of a number of coal samples from New Zealand at a temperature below 100^{o}C. The oxidation rates occurred under varying airflow rates using an adiabatic (2m long) column containing 100kg of crushed coal particles. The scatter in the kinetic plot was due to the experimental technique and a variety of complications such as moisture transfer, shrinkage of the coal bed and oxidation ageing. It took 14 days for the tests to reach a temperature higher than 100^{o}C for coal liable to spontaneous combustion, while up to a month for coal less prone to spontaneous combustion. The study indicates the

problem of comparing the results of adiabatic laboratory tests to the spontaneous combustion liability results obtained in coal stockpiles.

Correa *et al.* (1997) investigated the influence of atmospheric conditions on the run-off-mine coal exposed for ten (10) months in the stockpiles. The petrographic composition and chemical properties variation were investigated. The calorific value reduced from 29018 to 27402J/Kg in the run-off mine. Factors such as volatile matter content and vitrinite reflectance experienced small changes in their values. It was found that the stockpile samples were petrographically distinguished by an increase in oxidation rims and cracks due to long time storage.

Uzun & Ozdogan (1997) investigated the total and combustible sulphur, CaO, ash and sulphur forms of 19 different Turkish coal according to the American Society for Testing and Materials (ASTM) standards. The coal samples were carefully chosen to span a similar range with respect to proximate analysis and total sulphur content. The correlations developed were proved for combustion conditions when compared with the coal intrinsic properties determined according to the ASTM standards. The results indicated that sulphate sulphur give rises to SO_2 emission during spontaneous combustion of coal.

Ren & Edwards (1997) evaluated the delay of spontaneous combustion liability of coal at the Department of Mineral Resources Engineering, University of Nottingham. Standardized adiabatic oxidation tests have been used to identify coal liable to spontaneous combustion. Coal intrinsic properties, geological settings and mining techniques were used to evaluate the proneness of coal spontaneous combustion with a computerized apparatus coupled with an expert system. This approach was explored for airflow behaviour, nitrogen inertisation processes and simulation within the goaf. It was indicated that the model can be used to locate areas most prone to spontaneous combustion and improve the use of nitrogen as a retarding agent.

Monazam *et al.* (1998) created a transient one-dimensional spontaneous combustion model to describe spontaneous combustion liability at a relatively low temperature. The model consists of three differential equations which described the temperature, moisture and oxygen concentration in a char stockpile. The equations were solved numerically by a finite difference method. The effects of char particle size, moisture content, initial char temperature and char reactivity during self-heating were evaluated. The initial temperature of the stockpile had a strong impact on the ignition time. It was indicated that sensitivity of these factors are the

variables influencing the spontaneous combustion liability of coal. The model developed was validated against data obtained from large-scale spontaneous combustion tests.

Ren *et al.* (1999) investigated the spontaneous combustion liability of eighteen (18) pulverised coal in Australia, UK, US, Indonesia, South Africa and South America using the adiabatic calorimeter. All the samples were tested at a starting temperature of 40^{o}C except for three samples tested at 60^{o}C. Their liabilities toward spontaneous combustion was ranked on the basis of their initial rate of heating (IRH) and total temperature rise (TTR) values. The study used air humidity as a major factor to determine whether heating will increase rapidly or not. The particle size distribution of the coal retards the IRH and TTR values with relatively smaller particles prone to be more reactive. It was found that aged and pre-oxidised coal have higher IRH and TTR values.

McManus *et al.* (1999) discussed the parameters that influence spontaneous combustion liability in coal stockpiles. The parameters are coal rank, surface area of exposed pile, ambient temperature, oxygen content of coal, moisture in coal and configuration of the coal. The study demonstrates that the effects of these variables goes a long way in predicting the spontaneous combustion liability of coal.

Sujanti & Zhang (1999) investigated the effects of inherent organic matter on coal spontaneous combustion. Freshly picked raw coal was crushed and sieved to 0.25 to 1.00mm size fraction. Fourteen samples were prepared; the raw coal, water- washed coal, acid-washed coal and acid-washed coal doped with eleven additives. Each sample was tested in an isothermal reactor to obtain its critical ambient temperature above which spontaneous combustion occurs. The relative effectiveness of the additives was determined by comparing their critical ambient temperatures with those of the raw coal and acid-washed coal. Potassium chloride, montan powder and sodium chloride were indicated to be the most effective inhibitors followed by magnesium acetate and calcium chloride. The presence of sodium nitrate and ammonium chloride in the coal samples did not show any significant influence on the spontaneous combustion. However, calcium carbonate, potassium acetate and sodium acetate promote spontaneous combustion liability. The inhibiting effects of NaAc and KCl on spontaneous combustion liability enhanced an increase in their loadings. It was found that the critical ambient temperature showed a linear correlation with the reactor external surface area. This indicates the role of external heat transfer in the spontaneous combustion.

Fierro *et al.* (1999) investigated the effect of self-heating of fire stockpiles (2000 to 3000T) built at the ENDESA Power Station in Spain. The effect of several measures to reduce the heat losses was tested with the application of a low angle slope, periodic compaction, protection of the coal stockpiles with an artificial barrier and covering with an ash-water slurry made with fly ash from the same power station. Wind tunnel tests were used to design the wind barrier which was found to be very effective. The results indicated that the best way to guide against the heat losses is the use of ash-water slurry to cover the coal pile. The coefficients of heat losses and total losses were measured by the development of a direct method to determine the coefficient of total losses. The consistency between the thermocouples and temperature measured by infrared thermograph showed the approach is reliable to evaluate heat emissions from coal stockpiles.

Garcia *et al.* (1999) studied the initial stages of coal oxidation using differential scanning calorimetry. Three Columbian low-rank coal which were weathered under ambient conditions for a period of 105 days were used. Samples were collected directly from the mines and stored in plastic containers with de-aerated water. Two of the coal samples used (Vanesia and Amaga,) were known to be liable to spontaneous combustion. The Titiribi coal that belongs to the same coal basin as the other two is more stable to reaction with oxygen at ambient temperature conditions. It was demonstrated that the overall oxidation enthalpies reduced with increasing oxidation. The random behaviour was due to weight losses in the course of the experiments. The initial temperature of oxidation increases with increasing coal rank. It was found that the initial temperatures are a better sign of the propensity of the coal to spontaneous combustion.

Moghtaderi *et al.* (2000) incorporated a mathematical model of self-heating into a Computational Fluid Dynamic (CFD) package called FLUENT. This was used to simulate the transport of heat and mass due to spontaneous combustion in a coal stockpile. The effects of wind-driven flow field on spontaneous combustion liability of a coal stockpile were investigated. The calculations of heat and mass transfer processes within a realistically shaped coal stockpile were integrated into a wind flow model. The wind was presumed to blow in a fixed direction relative to the pile. It was indicated that the wind flow changes the dynamics of the flow in the stockpile and affects spontaneous combustion liability. The combined effect of the natural convection processes inside the stockpile and the external wind flow was found to form a different flow order.

Erdogan & Vedat (2000) derived some equations between the results of linear and multiple regression analysis to determine the relationship between spontaneous combustion liability and coal properties. The linear regression analysis showed that ash, volatile matter, carbon, hydrogen, exinite, and inertinite are the major factors affecting spontaneous combustion liability. The multiple regression analysis indicated that volatile matter, carbon, hydrogen, nitrogen, oxygen, sulphur and inertinite are the principal factors affecting the spontaneous combustion liability of coal.

Akgun & Essenhigh (2001) investigated the effect of the coal moisture content, pile height, coal particle diameter and slope angle of the bed on the spontaneous combustion liability of a coal stockpile. It was observed that the physically realistic values of the ignition time (>15 days), temperature (60 to 80^{o}C) and hotspot locations can be predicted for a packed coal bed. The model shows the significance of bed porosity and coal type. The probability of ignition increases with decreasing rank. It was found that spontaneous combustion occurs when two primary conditions are satisfied. Firstly, when the height exceeds some critical value (2m) and secondly if there is sufficient time where spontaneous combustion can occur within 15 days or more. The results have significant practical applications and support the standard practice of compression as coal stockpiles are created.

Rosema *et al.* (2001) developed a numerical simulation model "COAL TEMP" to study the oxidation and possible spontaneous combustion of coal subjected to the atmospheric conditions. The equations illustrating the exchange of heat, radiation, oxygen and the differential equations of oxidation, heat flow and oxygen flow in the coal matrix were investigated. The model was used to study the spontaneous combustion liability of coal in the Rujigou coal basin and other Coalfields. The results showed that the Rujigou coal basin has low spontaneous combustion liability and also found that self-heat may be detected as a hot spot, by means of thermal infrared viewing systems at the initial heating stage.

Blazak *et al.* (2001) used an adiabatic oven test to provide a full temperature of oxidation rates up to the self-sustained process to determine the spontaneous combustion liability of New Zealand. The studied sub-bituminous coal had the highest propensity of 14.91 to17.23^{o}C/h. It was confirmed from experimental analysis that resin bodies from the Rotowaro coalfield showed no propensity to spontaneous combustion. This may be necessary due to structural changes and different unknown mineral matters affecting the spontaneous combustion liability of coal.

Fierro *et al.* (2001) applied a one dimensional model to stockpiles containing a mixture of lignite coal that was investigated to be unsafe due to spontaneous combustion. The coal selected for the study was collected from the Mezcia coalfield in the Teruel basins. The mathematical model developed for the coal sample indicated that the stockpile porosity and wind speed played a significant role on spontaneous combustion. Voidage is critical and controls the effect of the wind velocity. Five coal stockpiles (2000 to 3000T) were tested. A number of factors (such as the use of a low angle slope, periodic compaction, protection of coal stockpiles with artificial barriers and covering of the coal bed with ash-water slurry) were investigated on four of them. This was done to lower the negative effects of wind velocity and stockpile porosity. The heat losses were experimentally investigated.

Beamish *et al.* (2001) determined the spontaneous combustion liability of sub-bituminous coal from the Callide Coalfields with a 2m adiabatic column. A coal block 60cm x 40cm x 40cm was collected from the Dunn Creek mine operating in Callide Coalfields, Queensland in May 2000. The coal samples were preserved in the laboratory freezer till July 2000. The coal was divided into two parts. One part was cut into portions and stored in the freezer, while the other half was kept intact at room temperature. The two portions of the sample were pulverized into the size of -75mm and placed in the 2m adiabatic column for testing in September 2001. It was observed in the study that;

- Due to moisture adsorption, the early heating evolves at the peak of the column;
- At 60cm from the air inlet, an ultimate hotspot evolved with a higher spontaneous combustion liability index at some points; and
- The heating that finally evolved in the column is equivalent to the effects of sub-bituminous black thunder coal described by (Li & Skinner, 1986).

It was concluded that different Australian coal should be tested to allow the high accuracy of a finite difference model and evaluate the effectiveness of small-scale adiabatic tests.

Kaymakci & Didari (2002) carried out linear and multiple regression analysis to determine the relationship between the spontaneous combustion liability index (derived from temperature curves found from laboratory tests) and coal parameters (obtained from proximate, ultimate and petrographic analysis). The additive regression analysis showed that ash, volatile matter, carbon, hydrogen and mineral matter are the main factors affecting spontaneous combustion

liability. The major factors such as volatile matter, carbon, hydrogen, nitrogen, oxygen, sulphur and inertinite are the multiple regression analysis results.

Ward (2002) evaluated the total proportions of unknown mineral matters in coal samples by low-temperature oxygen-plasma ashing. The study determined the contents of individual constituents using the Computer Controlled Scanning Electron Microscopy (CCSEM) and Retrieved Based X-ray Powder Diffractometer. The study observed a scope for standardization in coal that showed the chemistry of mineral composition obtained from the mentioned methods. This is consistent with the chemical composition of the coal ash obtained from the same samples by direct analytical methods. It was found that the knowledge of the mineral matter in coal is essential to study the organic matter in the processes of a coal formation. The trace elements, minerals and another inorganic substances present in coal are important in practical applications of coal geology such as, correlating coal seams and seam sub-sections and identifying the mode of occurrence.

Beamish *et al.* (2003) upgraded a 2m self-heating column constructed at the University of Queensland to subdue the constraints observed with the earlier large-scale device. The device has improved confidence in detailed experimental testing than any earlier work and permits periodic data to be generated in days rather than months. The system offers a broad understanding of the phenomenon of spontaneous combustion on different coal samples. The study requires more findings on the impacts of physical variables like porosity, airflow rate and particle size distribution on the spontaneous combustion liability of coal.

Kucuk *et al.* (2003) studied the effects of the gas flow rate, the moisture of the stockpiles and the particle size with the use of XPT on Askale lignite in Turkey. It was found that the spontaneous combustion liability of the lignite increases with decreasing particle size, increases the moisture content of the coal and decreasing humidity of the air.

Krajciova *et al.* (2004) evaluated the role of meteorological conditions at varying temperatures in a coal stockpile. A mathematical model of experimental values measured by meteorological points was used to define the periodic boundary conditions. The energy from the sun has a strong effect on the temperature profile in a coal stockpile. Four different coal types were studied on the basis of reactivity. The influence of several factors (coal reactivity, coal matrix, porosity and meteorological conditions on the temperature profile in two types of coal stockpiles) were observed. The results showed the influence of meteorological conditions on

the stockpile temperature. The effect of coal reactivity illustrated a strong influence on the maximum temperature in the stockpile. The simulations of a heap-like stockpile (two dimensional) indicated good results as the temperature profile is not symmetric. The hot spot occurs on one side due to solar radiation distribution on two different oriented slopes.

Sahu *et al.* (2004) evaluated the spontaneous combustion liability of coal using the DSC. Thirty coal samples from seven (7) different Indian Coalfields were investigated by this method. The initial temperatures for the samples were determined. A comparative study of the initial temperature and the XPT was comprehensively discussed. It was found that the temperature increase in the coal indicated spontaneous combustion liability.

Humphreys (2005) reported a study that combines the comprehensive knowledge of the coal oxidation and heat loss mechanisms during spontaneous combustion. Numerical modelling was used to simulate laboratory tests and the spontaneous combustion behaviour of coal in stockpiles. A direct link was established between the results obtained from the laboratory tests and the predicted field behaviour of coal.

Beamish *et al.* (2005) established a relationship between the R_{70} values of coal and the effect of the coal rank, moisture content and mineral content. The study provides a better understanding of the spontaneous liability of coal. It was found that the intrinsic reactivity of coal towards oxygen can be accurately examined by the R_{70} test procedure. The value of this parameter is strongly affected by the coal rank and mineral matter. The study suggested the need to review the way in which other scale index parameters like the XPT and minimum self-heating temperature are used in spontaneous combustion risk assessment.

Sahu *et al.* (2005) evaluated the spontaneous combustion liability of some Indian coal with the use of the XPT, DTA and DSC. The study shows that the spontaneous combustion liability of coal varies from one seam to another. It is essential to measure their level of spontaneous combustion liability in order to plan advanced precautionary measures. It was observed that the initial temperature obtained from the DSC method is more reliable than the XPT.

Smith & Glasser (2005a) measured the spontaneous combustion liability of coal stockpiles. The relative importance of various coal intrinsic properties and the factors that influence the rate of low-temperature oxidation reaction were discussed. A regression analysis was carried out to relate the measured initial rate of oxidation, as determined on ash free basis with their petrographic composition, proximate and ultimate analyses. It was found that the multiple

regression analysis showed that the variables in the petrographic, proximate and ultimate analysis are the main factors affecting the spontaneous combustion liability of coal.

Smith & Glasser (2005b) investigated the effects of the moisture content of air on the spontaneous combustion liability of coal. The effect of the moisture content on self-heating is dependent on the coal rank and temperature. The outcome of the research demonstrates good agreement reached by Smith & Lazzara (1987) in a report of investigations for the US Bureau of Mines (USBM) founded in 1910. It was found that adsorption of water vapour does not compete with the low-temperature oxidation in terms of heat generation but appears to facilitate the oxidation rate and possibly plays a catalytic role.

Ward & French (2005) determined the amounts of various mineral matter and glass in fly ash samples using the Rietveld-based X-ray diffraction analysis. The studies were carried out using an ash sample spiked with a known amount of different minerals (Corundum and ZnO). The mineral compositions in the individual feed coal and fly ash showed significant correlation with each other. This indicated that a considerable amount of quartz in the coal have the tendency to move directly into the fly ash, without significant alterations. The iron minerals in the fly ash and coal indicated a more dispersed but roughly linear relationships between the number of iron oxide minerals in the fly ash and the quantity of siderite with pyrite in the low-temperature ash of the feed coal. This shows the iron oxide minerals in the fly ash are obtained from the iron minerals existing within the coal.

Arisoy *et al.* (2006) investigated both theoretically and experimentally the spontaneous combustion liability of coal stockpiles using a dimensional test column. A detailed computer model to simulate a full scale one-dimensional test column was developed. Predictions from this model were used to simulate full-scale storage conditions. The test carried out closely matches the model prediction characteristics of moisture transfer. The hotspot migration in the column was clearly visible in both tests. It was shown that a sub-bituminous coal reaches thermal runaway in 4.5 days in the study. The result was confirmed with the observations made at the mine site, where hotspots occurred within this time frame.

Ozdeniz & Sensogut (2006) developed a computer-controlled measurement of spontaneous combustion in the coal stockpile of the Western Lignite Corporation, Turkey. The objective of a computerized measurement system was to measure temperature changes existing in a coal stockpile. Twenty temperature sensors were placed at certain points inside the stockpile to

sense the electrical signal conversation of temperature. The electrical signals transferred the temperature data into a computer media. This was done by using analogue-digital conversion after applying filtration and upgrading processes. The graphs of the time-temperature data were plotted to evaluate the behaviour of coal stockpiles with respect to spontaneous combustion and take necessary precautions against spontaneous combustion in advance.

Yildirim *et al.* (2006) investigated the various developments in determining the spontaneous combustion liability of coal. The study emphasised that most methods of concern purely depend on the coal inherent properties. It was found that the relationship between the XPT and electrical resistance of coal can be used to predict spontaneous combustion liability.

Uludag (2007) studied the spontaneous combustion liability of South African coal with a small scale laboratory testing apparatus. The coal inherent properties with the DTA and XPT methods were determined. The relationships between DTA obtained after testing were compared with the actual inherent moisture. It was observed that as the Wits-Ehac Index increased, the inherent moisture content of the coal increased. It was deduced that the liability of spontaneous combustion increases with increased inherent moisture. The volatiles has a catalytic effect on self-heating immediately after the inherent moisture was reduced. In addition, as carbon content increases, the Wits-Ehac Index value increases, while the ash content decreases the possibility of spontaneous combustion.

Choudhury *et al.* (2007) investigated the influence of maceral and mean reflectance (R_{max}) on the spontaneous combustion liability of coal. The spontaneous combustion liability of some selected coal samples was investigated with the TGA. The amount of vitrinite and liptinite show high spontaneous combustion liability. The study showed no significant correlation between vitrinite reflectance and TGA parameters. The TGA parameters were observed to have good correlation with the petrographic composition that combines the effect of coal rank and mineral matter. The research identified the need to consider the role of low-rank inertinites, while evaluating the contributing behaviour of inertinite rich non-coking coal.

Singh *et al.* (2007) studied the phenomenon of spontaneous combustion in opencast coal mines. It was found that in opencast mines, coal directly oxidized and begins to self-heat. This was confirmed to be due to the coal rank, high moisture and volatile matter, presence of sulphur in the form of pyrites, low XPT, ignition point temperature values and less incubation period.

Sensogut *et al.* (2008) determined the temperature variation movements within a stockpile of dimension (10m by 3m with a height of 3m) in the Western Lignite Corporation (WLC) in Tuncbilek, Turkey. A computer system was used to record the heat data received from 20 resistance temperature detectors. The measurements were taken from twenty (20) different interior points of the coal stockpile formed to predict spontaneous combustion liability. The interior temperature distribution maps of the stockpile together with the maximum and the minimum temperature values were expressed visually and numerically with the assistance of the data obtained. It was found that the subsequent stockpile's location, direction, height and its position according to the sun and the selection of region considering wind directions needs to be taken into consideration.

Chelgani *et al.* (2008) analysed the effects of coal properties on some Kentucky coal from the caloric value of 4320 to 14960 (BTU/Ib) on Hardgrove Grindability Index (HGI) using the Artificial Neural Network (ANN) and multivariable regression. The stepwise least square mathematical method demonstrated the relationship between:

(i) Exinite, semifusinite, macrinite, micrinite and resinite;
(ii) Moisture, ash, volatile matter and total sulphur; and
(iii) In (total sulphur), ash in [(oxygen + nitrogen)/ carbon] and coal rank with HGI and obtained correlation coefficients of 0.77, 0.75 and 0.81 respectively.

The ANN predicted the HGI with correlation coefficients of 0.89, 0.89 and 0.95 respectively. It was found that in (exinite), semifusinite, micrinite, macrinite, resinite and coal rank can be used to predict the estimation of HGI on multivariable regression (R^2 =0.81) and also by ANN methods (R^2 = 0.95).

Yuan & Smith (2008) modelled the effects of coal properties on spontaneous combustion of coal in longwall gob (mined out) areas with a CFD study. The coal has different spontaneous combustion liability based on their minimum self-heating temperatures. Simulation results signified that under typical longwall mining conditions, the spontaneous combustion liability of different coal are strongly dependent on mine ventilation conditions and coal properties. The porosity and permeability profiles for the longwall gob were produced from a geotechnical model. They were used as inputs for the three dimensional CFD modelling. The spontaneous combustion liability was modelled as the low temperature oxidation of coal in the gob using kinetic data obtained from previous laboratory scale spontaneous combustion studies. The

effects of the coal surface area and heat of reaction on the spontaneous combustion liability were evaluated.

Beamish & Arisoy (2008b) developed an extensive database for spontaneous combustion liability of coal under adiabatic conditions at the University of Queensland. The experimental results established relationships and trends on the effects of various intrinsic properties on the spontaneous combustion liability of coal. It was found that an effective rating scheme can be developed to evaluate coal liable to spontaneous combustion and identify appropriate mining approaches for spontaneous combustion management planning.

Nawaz *et al.* (2009) investigated the presence of mineral matter in the form of kaolinite, pyrite and a small amount of hematite under microscopic study of five polished Makerwal coal samples. The mineral matter determined by the X-ray diffraction technique is in agreement with the mineral constituents observed under microscopic study.

Ozdeniz & Yilmaz (2009) investigated the effects of temperature due to atmospheric conditions in a coal stockpile in Konya, Turkey. The stockpile has a total tonnage of 120T with dimension of 5m by 10m and a height of 3m. Seventeen (17) temperature sensors were placed inside the stockpile at certain locations and the values were recorded. Electrical signal conversion of temperature sensed by 17 temperature probes inserted at certain locations within the stockpiles transferred the electrical signals into computer media. This was done by using the analogue-digital conversion unit after necessary filtration and upgrading processes were applied. The spontaneous combustion was observed inside the stockpiles having a 10mm to 18mm coal grain size. This is because the smaller the grain sizes, the larger the surface area and the more contact with oxygen, making the heat accumulate continuously. The measurement values relating to the air pressure, wind speed, wind direction, and air temperature were used for training and testing of the ANN model. Accuracy rates of training and testing were found to be 99.5% and 99.17% respectively due to the comparison of the ANN and equipment results. It was concluded that the spontaneous combustion liability of the coal stockpile can be estimated with an ANN model.

Ejlali *et al.* (2009) investigated the effects of a coal particle size on spontaneous combustion liability. In addition to the increased oxidation rate of smaller particles, the rate of heat transfer between particles increases as their size decreases. The rate of oxidation increases with decreasing particle size until a critical diameter is reached. The critical diameter corresponds

to a size at which oxygen penetrates the particle without experiencing any mass-transfer resistance. This depends on factors such as the reaction conditions and the porosity of the coal. It was found that the spontaneous combustion liability increases as the coal particle size decreases.

Zhang *et al.* (2009) determined precisely the oxygen concentration and temperatures inside coal stockpiles. The detail on self-heating conditions and spontaneous combustion liability of coal was acquired with designed experimental equipment. The equipment composed of a heating system, a column for testing coal and a monitoring system for the simulation of spontaneous combustion of coal blocks (23 to 25mm), covered with fine coal (0 to 3mm). The self-heating system is made up of an AC power, an adjustable transformer and a heating panel. Temperature sensors, conversion modules, a computer and a gas chromatograph constitute the monitoring system. A BP-ANN with 150 training samples was established over the course of the experiment. The oxygen concentration and temperature of the coal column were selected as the output variables. The trained network was applied to estimate the trend on the untried experimental data. Based on the prediction of the ANN, the average errors of oxygen concentration and temperature were 0.5% and 7^{o}C respectively, which meet actual tolerances. It was found that the application of this method can provide a practical guide to understanding the course of spontaneous combustion of coal stockpiles.

Yuan & Smith (2009) carried out a CFD simulations to model the spontaneous combustion liability of a large scale coal bed chamber. The temperature history of the coal bed was recorded with the use of thermocouples arranged in seven (7) vertical arrays of nine (9) thermocouples, 0.3, 0.9, 1.5, 2.1, 2.7, 3.4 and 4.0m respectively from the front of the coal bed. Spontaneous combustion was modelled as the low-temperature oxidation of coal using kinetic data obtained from previous laboratory scale spontaneous combustion studies. The CFD model was validated by comparing simulation results with test results obtained from the U.S. Bureau of Mines experiments conducted in the coal chamber. The model predicted lower temperatures in the early stage but proved well on the induction time for spontaneous combustion. The effects of the airflow rate and order of reaction during spontaneous combustion were determined. The calibrated CFD model is found to be useful for predicting the induction time for spontaneous combistion in underground coal mines.

Sahu *et al.* (2009) applied an experimental technique for the classification of coal seams based on their liability toward spontaneous combustion. Twenty-nine coal samples varying in ranks

were collected from the major Coalfields in India. The principal component analysis (PCA) was applied to classify the coal seams into three different classes using the XPT, ash, moisture content and volatile matter of the coal samples. The classification methods are equivalent to the spontaneous combustion liability of coal seams in the actual field conditions. It was found that the effectiveness of this method requires coal seams from other parts of the world to be tested.

De-Ming *et al.* (2009) indicated a new standard for the classification of spontaneous combustion liability of coal. The technique used two variables for different oxidation stages. Based on the comparative tests for comprehensive determination index with the adiabatic oxidation of varying coal and physical adsorption, the value of comprehensive determination index indicates the capacity of coal-oxidation reaction. It was found that the path of oxidation is more scientific and accurate to identify the spontaneous combustion liability of coal. The oxygen adsorption method was used to test the absorbed oxygen at 30^{o}C. The oxidation kinetics which considered the complete dynamic process was put forward by testing the oxygen concentration in the outlet of the sample vessel at 70^{o}C and the XPT. It was found that this technique can study spontaneous combustion liability of coal.

Khandelwal & Singh (2010) investigated various coal intrinsic properties to evaluate the accuracy of ANN and Multivariate Regression Analysis (MVRA). The concentration of macerals of Indian coal were determined using ANN by incorporating the proximate and ultimate analysis of the coal. Data sets from different Coalfields in India were used for training and testing of the ANN for the prediction of macerals. The network was trained by 149 datasets with 700 epochs. It was tested and validated by 18 datasets. It was confirmed that the correlation coefficient between measured and predicted macerals by the ANN was significant. The mean absolute percentage error was negligible as compared to the MVRA.

Xuyao *et al.* (2010) established an experimental technique to test the oxygen consumption of different coal ranks at programmed temperatures. The system heat rate was 0.8^{o}C/min and the coal sample sizes ranged from 0.18 to 0.42mm. The results indicated that for high rank coal, oxygen consumption increases with coal temperature as a non-linear process. Oxygen consumption at 100^{o}C under programmed temperatures can reflect the oxidation ability of coal perfectly. It was found that the oxidation process of coal is greater and its proneness to spontaneous combustion is easier if its oxygen consumption at 100^{o}C is more.

Xianliang *et al.* (2010) compared the effects of coal samples subjected to atmospheric conditions for a period of three months using the TGA. Three coal samples collected were crushed to a particle size less than 1.25mm. Each coal sample was divided into two coal stockpiles making a total of six stockpiles. The effect of Polyvinyl alcohol (PVA) oxygen insulating barrier on the spontaneous combustion of coal was investigated. The combustion activation energy, moisture loss activation energy and oxidation activation energy were determined using the coats Redfen formula. The results indicated that the spontaneous combustion liability of the three coal samples falls in the order of CYW>YJL>SW. The combustion activation energy and oxidation activation energy of samples protected by the PVA oxygen-insulating barrier both increased. The activation energy of coal samples enveloped by the PVA oxygen insulating barrier increased appreciably. It was found that the PVA oxygen insulating barrier can help prevent the spontaneous combustion liability of coal.

Ozdeniz *et al.* (2011) determined the conditions wherein coal spontaneous combustion may occur due to long exposure to atmospheric conditions. This was done in an industrial scale stockpile (10m by 5m with a height 3m) with a grain size of 10 to 18mm in Konya, Turkey. Seven temperature sensors (Pt 100) were placed at certain points inside the stockpile, while the temperatures were recorded. The effects of atmospheric conditions such as air temperature, air pressure, wind direction and wind speed were investigated by continuous measurements. The gravitational effect under the long term stay and the vibration resulting from heavy duty machines working closer to the stockpiles makes compaction necessary in coal stockpiles. The relationship between the calculated compaction rate and the spontaneous combustion liability was determined. It was observed that the inside temperature of the stockpile increased parallel to the compaction. This was because a decrease in the coal grain size caused the larger coal surface to be in contact with the confined oxygen for a long period of time. The heat inside the stockpile could not escape but accumulated continuously within the stockpile due to the effect of compaction.

Ejlali *et al.* (2011) studied the self-heating mechanisms of wet porous media coal stockpiles using a numerical approach. The heat, fluid flow and moisture transport within and around the reactive porous medium (a coal stockpile) were numerically investigated. This was based on the coal thermal non equilibrium approach. The results were compared with experimental results. It was found that the temperature within the porous medium increases at the start of the process due to spontaneous combustion. The rate of the temperature increase slows as the

simulations proceed due to a phase change from liquid to vapour within the medium. The last stage of self-heating involved an increase in the temperature which occurs when liquid moisture is fully vaporized and the phase change is no longer available to restrain the temperature rise. It was found that the required temperatures limit was produced numerically as a function of Darcy number D_a, porosity of the porous medium ε and moisture content W_t.

Rafezi *et al.* (2011) evaluated the intrinsic properties (proximate, ultimate analysis and gross calorific value) of 4540 US coal samples collected from twenty-five (25) States. The significant correlation between the parameters of proximate and ultimate analysis was used to predict gross calorific value using multiple regression analysis and the Adaptive Neural Fussy Inference System (ANFIS).

Khorami *et al.* (2011) applied multiple regression analysis (MRA) and the ANFIS on the results of petrographic composition, proximate and ultimate analysis of Kentucky coal to predict the Free Swelling Index (FSI). Three various input sets were applied for both techniques:

(i) Group- maceral analysis, mineral matter, moisture, sulphur and vitrinite maximum reflectance (R_{max});
(ii) Moisture, ash and volatile matter; and
(iii) Carbon, hydrogen, nitrogen, oxygen, sulphur and mineral matter.

With the same input sets, the ANFIS predicted the FSI with higher correlation coefficients of 0.46, 0.82 and 0.95, while nonlinear regression obtained the correlation coefficients of 0.38, 0.49 and 0.70 respectively. The study indicated that the input set (i) can be used to predict the FSI in both prediction methods. The ANFIS can be used to predict the FSI, while regression did not show any accuracy in the results.

Zhang *et al.* (2011) developed a nested spontaneous combustion furnace with better thermal insulation performance. The electrical sand bath was taken as a thermal insulation and gas preheating device. The temperature control device with a higher accuracy was selected in order to solve the problems of heat dissipation and temperature control. It was found that the oxidation process of the tested coal was simulated with the developed device after investigating the coal samples in the laboratory.

Pattanaik *et al.* (2011) investigated an Infrared Studies (IR), DTA, XPT, petrography and chemical analysis of different coal seams covering the entire stratigraphic sequence in the Chirimin Coalfields. The study established a significant relationship with the stratigraphic

disposition of the coal rank on the basis of the mentioned parameters. Low rank coal found at the top of the seams in the stratigraphic sequence are more liable to spontaneous combustion, while the higher coal found at the bottom of the stratigraphic sequence have low spontaneous combustion liability. It was found that the lower Karakoh and Sonawani seams have low spontaneous combustion liability while, the upper Duman and Kaperti seams placed at the top seams in the stratigraphic sequence are more liable to spontaneous combustion.

Eble (2012) investigated the organic composition of ten (10) samples of the Carbondale formation roof shale strata from the Western Kentucky coalfield. The shales were obtained in addition to thirty two (32) Carbondale formation coal samples that were studied for the coal bed methane content. The petrographic analysis showed variation in the macerals of the shales. The coal composed of a lesser amount of liptinite and inertinite than the Carbondale shale formation. The liptinite macerals were found to be rich in the shales but deficient in the coal.

Kim & Sohn (2012) studied the spontaneous combustion liability of coal stockpiles numerically for fire safety measures. The study simulated spontaneous combustion processes with physical and chemical models that are widely applied for high rank coal. The coal oxidation mechanism is analysed in thermal and fluid dynamic aspects. It was found from the numerical results that the inflow of air inside the coal stockpile is indispensable for long coal oxidation. The heat accumulation inside the stockpile causes the temperature to increase over a critical value of about 200^{o}C. Three methods were proposed in the study based on the self-heating mechanism. The methods adopted a dual barrier installed inside the stockpile with air blowing from the bottom of the stockpile. The three methods used have been verified to delay spontaneous combustion time more than the existing methods. The expected additional delay was about 10 to 30 days. It was found that more delays could be caused by a combination of the mentioned methods.

Sharma *et al.* (2012) evaluated the petrographic composition of some high sulphur tertiary coal samples in India and predicted their spontaneous combustion liability. Multivariable Regression Analysis (MRA) was used to study the relationship between macerals and the Gross Calorific value (GCV) of the coal samples. The macerals showed that the Northern Indian coal samples have a high vitrinite content (80.07% average), moderate to low liptinite (10.23% average) and low inertinite (9.3% average). The liptinite and inertinite contents were found to have strong linear relationships (R^2 =0.9283).

Zhu *et al.* (2012) developed a coal heating and oxidation device to evaluate the oxygen concentration for three coal samples at the inlet and outlet of the furnace. The device consisted of a heating and oxidation furnace, gas chromatograph, temperature control and data acquisition systems and other components. The oxygen consumption rates of the coal were determined during the heating process with respect to the airflow. The oxygen consumption rate increases with increasing temperature before the critical temperature (about 150^{o}C). The oxygen consumption rate increases steadily as the coal temperature exceeds the critical temperature. The relationship between the oxygen consumption rates and the temperature before and after the critical temperature shows linear relationships with a correlation coefficient exceeding 0.9. The result is significant to prevent the event of coal spontaneous combustion.

Nimaje *et al.* (2013) applied different techniques such as petrography, thermal and oxygen avidity to determine the spontaneous combustion liability of coal from Australia, USA, Czech Republic, China and India. The spontaneous combustion liability was investigated using the XPT apparatus. Statistical analysis was carried out between the XPT and proximate analysis parameters. It was found that the Mixture Surface Regression (MSR) model provides excellent residual values as compared to the polynomial and simple multiple regression models. The effect of Anderson-Drilling testing was evaluated on the basis of prediction results of the MSR model. The measured value of the XPT shows that the residual values followed normal distribution. This justified the suitability of the model for the prediction of spontaneous combustion liability of coal.

Morla *et al.* (2013) predicted the spontaneous combustion liability of thick coal seam using the XPT, DSC, spontaneous propensity test, gas revolution test and coal properties. Furthermore, the application of the CFD simulations was applied to investigate the goaf gas behaviour at the time of spontaneous combustion in the blasting gallery panels. The CFD simulation studies was conducted using ascentional and descentional ventilation systems with an inert injection at a single injection point, multiple inject points and various heat flow rates. The result indicated that the descentional ventilation system can be used for goaf inertisation. The multiple gas injections were not as effective as the single injection.

Zhang *et al.* (2013) investigated the low temperature oxidation of coal in an enclosed area. A cube of unused Shendong coal was cut from a newly worked face of the Bulianta coal mine. A chain saw was used after first removing a layer of coal about 25cm thick to ensure that samples were not prone to pre-oxidation. A batch reactor was used to simulate the coal oxidation under

ambient temperature. The time dependence of the rates of CO emission and oxygen consumption during oxidation of coal samples with varying particle size ranges were obtained concurrently from the amount of CO and O_2 concentration in the reactor. Oxygen consumption during coal oxidation under ambient temperature was observed to have five stages in terms of the reaction rate equation (chemical control regime, inhibitory control regime, transition state, direct burn-off reaction and diffusion control regime). The results showed that the reaction system changes during coal oxidation in enclosed area.

Wang *et al.* (2014) identified chemical inhibitors that may impede coal oxidation through the *in- situ* Fourier transform infrared spectrometer (FTIR) method. Lumps of coal obtained from different collieries were pulverised to produce particle sizes from 0.25mm to 0.8mm for the tests. Polyethylene glycol (PEG) 200 (99% purity) was chosen as the chemical inhibitor in the study. The coal samples were dehydrated in a vacuum for the duration of a night at 40^{o}C. The inhibitor with the raw coal particles were mixed in a container with an automatic stirrer to form coal samples containing 0, 1, 2, 5 or 10wt % of the PEG additive. The Nicolet 6700 FTIR was used to estimate the difference in concentration and distribution of the coal surface functional groups. The reaction vessel was stocked with ground coal samples with the dome installed. The oxygen consumption and the XPT in the low temperature oxidation (40 to 70^{o}C) and the combustion gases were determined with an apparatus. The apparatus consists of a spontaneous combustion simulation system, a data acquisition system to record the temperature in real time and a gas chromatograph. It was found that the XPT of the treated coal was greater than those of untreated coal. Their oxygen consumption rates were lower at oxidation temperatures between 40 and 70^{o}C. The quantities of CO released from the treated coal were much lower than the quantities produced by untreated coal. The study indicated that the preferred technique, which takes the structural features of specific coal into consideration, can successfully address the choice of chemical inhibitors to delay coal oxidation.

Taraba *et al.* (2014) studied the effects of wind on coal stockpile spontaneous combustion using a CFD. A two-domain model was developed to simultaneously solve the governing equations of a coal stockpile domain situated in a wind flow domain. Numerical simulations with fixed direction and real fluctuations of the airflow confirmed the role of wind in the development of sponatneous combustion. Under the conditions of the simulations, three possible shifts of a hot spot in the stockpile were distinguished for coal to undergo spontaneous combustion:

- Hot spot moved inwards to the stockpile as the wind speed increases;

- Transfer of the hot spot from the upper part of the stockpile to the lower part when the self-heating progresses. The speed was estimated as $\leq 3ms^{-1}$ and clearly is mainly connected with the effect of buoyancy; and
- The shift of the hot spot to the stockpile surface when self-heating of coal is in progress.

It was found that the study supports the use of protection against blowing air on the windward side of coal stockpiles as a method of reducing the risk of spontaneous combustion.

Avila *et al.* (2014) investigated the textural features of different coal after oxidation reaction with the use of immersion microscopy and image analysis techniques. Petrographic composition (vitrinite reflectance and maceral analysis) provides information about the textural characteristics of each coal type. These parameters provide partial information about the potential of samples to undergo spontaneous combustion. The petrographic analysis of particles subjected to control heating revealed a number of thermal alterations including a change in vitrinite reflectance and the formation of particular morphotypes. Changes in vitrinite reflectance during oxidation correlates to the spontaneous combustion liability. Six different thermal-morphotypes were identified and classified (as primary or secondary alterations). The six was simplified as particles with homogeneous reflectance change produced by a kinetic control of the oxidation and particles with oxidation rims, produced when the reaction was controlled by oxygen diffusion. The magnitude of changes in vitrinite reflectance and the morphological characteristics were related to the spontaneous combustion liability of coal. This provides an additional source of information to identify coal prone to spontaneous combustion.

Taraba & Pavelek (2014) used pulse flow calorimetry (PFC) to determine coal liable to spontaneous combustion based on the values of oxidation heat q^{30} (wKg^{-1}). Around 300 varied coal samples (mainly from the Czech Republic and Slovakia, India, Germany, Turkey, Poland, Ukraine, Indonesia, Venezuela and Kazakhstan) were investigated to determine their spontaneous combustion liabilities. Heat from both physical and chemical interactions of the oxygen with the coal was estimated (denoted as q^f and q^{30}) from the calorimetric measurements. The maximum value of oxidation heat q^{30} was observed to be $10.5wKg^{-1}$. The classifications of coal spontaneous combustion liability were ranked on the basis of the control values of q^{30}. The variance between the initial rate of heating (IRH) and oxidation heat q^{30} of adiabatic tests was observed. It was found that with bituminous coal the PFC provide similar information on coal liable to spontaneous combustion as the adiabatic oxidation test.

Hooman & Maas (2014) theoretically analysed the problem of spontaneous combustion liability of a coal stockpile under free convection (no wind) conditions. The study considered stockpiles with geometry represented by two spatial coordinates. Scale analysis was used to derive the stockpile temperature and inflection point, as functions of the key parameters (coal type, particle diameter, moisture content and as well as the ambient air temperature). The theoretical predictions were successfully compared with experimental data. It was found that the model involves many simplifications and could be improved by including in more detail, such as the underlying physics.

Panigrahi & Ray (2014) determined the spontaneous combustion liability of seventy-eight (78) coal samples collected from 13 different mining companies in the Indian Coalfields using the Wet Oxidation Potential (WOP). Nine hundred and thirty-six tests were carried out under various experimental conditions to standardize the method for extensive practice. The results of the WOP method were correlated with the coal intrinsic properties (proximate, ultimate and petrographic analysis of the samples). The correlation analysis was conducted with the use of a Design Expert 7.00 software. The ANN analysis was used to ensure the best combination of experimental conditions were applied for obtaining results. It was found that the experimental conditions should be a 0.2-N $KMnO_4$ solution with 1-N KOH at 45^oC to achieve optimum results to obtain the spontaneous combustion liability of coal.

Ozdeniz *et al.* (2014) measured the behaviour of temperature distribution at certain locations within a longwall using temperature sensors. The temperature changes during the longwall operations were used to evaluate the event of spontaneous combustion. The temperature of the exhaust air and concentration of some of the mine gases (CO, CH_4, O_2 and CO_2) were determined. The Graham's ratio was calculated and used to determine the spontaneous combustion liability of the coal. During the investigation, as the temperature of the longwall increased with time, the coal production in the longwall was stopped. After eliminating the contact of the air with coal, the temperature was recorded and the temperature changes in the inner part of the longwall were recorded continuously.

Diacoru *et al.* (2014) simulated the physical processes involved in the event of self-heating with the use of a software package knows as ANSYS CFX. The transient regime simulation was conducted using numerous assumptions for the stockpile characteristics. The two-dimensional temperature profiles in the cross-section of the stockpile were recorded. A steady temperature increase caused by the heat generated due to low oxidation in the coal bed was

observed in the stockpiles. The heat generated in the vicinity of the side slopes greater than the rest of the stockpile was due to the low-temperature oxidation of coal. The continuous supply of oxygen to the side slope of the exposed coal stockpile increases the rate of the low-temperature oxidation. It was found that the low-grade coal with high humidity content are more liable to spontaneous combustion.

Jun *et al.* (2014) developed a formula to determine the concentration at which temperature increased in a temperature programmed experiment. The temperature changes during the low-temperature oxidation. The changes in the indicator gas CO_2/CO and the alkaline ratio of the coal of mixed particle sizes at various mines were investigated. The indicator gas analysis and growth rate analysis method were used to obtain the characteristics temperature points. It was indicated that the characteristics temperature points for different coal samples varied due to the coal grades. The internal structure of coal molecules reflects the difficult degree of coal spontaneous combustion and oxidation.

Chandralal *et al.* (2014) investigated the effects of high moisture on low-rank coal stockpiles in order to understand the possibilities of lowering their total moisture content under ambient conditions. A significant reduction in the moisture content of high moisture low-rank coal was observed from the small-scale dry tests. The experimental tests indicated consistent losses over time with an average weight loss of 27% for 22 days in which the test was run. The test showed the maximum possible natural drying potential with no impediments to drainage and additional moisture from rainfall. Dry conditions allowed the stockpiles to drain free moisture at a loss rate of between 0.7 and 1.7% per day. It was found that a natural drainage period of between 18 and 25 days assist in reducing the moisture content without considering the effect of atmospheric conditions on the stockpile temperature and moisture.

Wang *et al.* (2014) determined the critical self-heating temperature with the use of a thermal diffusivity and internal temperature inside a coal stockpile. The study used samples of two lignite coal samples (one raw and one thermally dried) and a sub-bituminous coal sample. The samples were pulverized into the mesh size of 0.25 to 0.75mm. The outline of temperature at the centre of a coal stockpile was measured at various ambient air temperatures (40 to 140^{o}C) in order to estimate the critical self-heating temperature values for the coal stockpiles. The critical self-heating temperature of the coal stockpile was determined from conceptual equations for scaling. It was found that the critical self-heating temperature as a function of stockpile size or volume was estimated on the basis of the Frank-Kamenetski model.

Song *et al.* (2014) proposed a comprehensive evaluation system for spontaneous combustion liability of coal stockpiles. The trapezoidal and the triangular extent Fuzzy Analytic Hierarchy Process (FAHP) methods were used to study the uncertainty of the factors affecting the spontaneous combustion liability of coal stockpiles. Four stockpiles were investigated to demonstrate the validity of the index system and the effectiveness of FAHP. It was found that the proposed evaluating methods were valid in determining the spontaneous combustion liability of coal stockpiles. The triangular extent FAHP method is more effective to evaluate the factors affecting spontaneous combustion liability of coal stockpiles than the trapezoidal FAHP method. The study can be used to minimize the risk of spontaneous combustion from a holistic point of view.

Qi *et al.* (2014) determined the thermal behaviour of different coal ranks with the use of a micro-calorimetry. Two coal samples (a lignite coal sample from YiMa mine and an anthracite coal sample from KA-BU-Liang mine) in an exothermic condition during the oxidation process from room temperature to 200^{o}C were studied. The kinetic parameters were consistently changing in the coal oxidation process, rather than a fixed value. Kinetics based simulation method was used to determine the temperature increase under adiabatic conditions. It was found that the liability of the YiMa coal sample to spontaneous combustion is extremely high, while that of KA-BU-Liang coal sample shows low spontaneous combustion liability. The variation between the predicted curve and tested curve obtained by the adiabatic test was not enough to be accepted. The oxidation process in the gob was evaluated with the heat balance equation of the residual coal and kinetics based simulation techniques. The time required for coal to reach 70^{o}C and the lower limit of the advancing speed of the working face was obtained from the predicted curve. It was found that the kinetics based simulation can be used to predict the spontaneous combustion liability of coal.

Pak *et al.* (2015) established a heat generation simulation for coal stockpiles. The simulation developed consists of unsteady analyses that links the heat transfer, flow and reaction in order to emulate the heat behaviour of a coal stockpile. Priority was given to the evaporation, adsorption and desorption characteristics of moisture in coal. Furthermore, the low-temperature oxidation of a coal stockpile and the flow behaviour of air inside the stockpile were taken into consideration. The airflow distribution was evaluated by measuring the grain size distribution of the coal samples at various locations within the stockpile. The pressure drop for each grain size distribution was determined. It was found that the simulation allows the prediction of

spontaneous combustion liability of different coal samples during their storage using data obtained from their physical properties without conducting large-scale measurements. The study indicated that the stockpile of sub-bituminous coal exhibits rapid temperature increases than that of bituminous coal.

Ozdeniz *et al.* (2015) investigated the effects of atmospheric conditions on an industrial scale stockpile (6m by 10m with a height 3m) of grain size of ± 18mm in Tuncbilek, Turkey. The industrial-scale coal stockpile was kept for 15 days in an open stock under atmospheric conditions. The change in temperature due to atmospheric conditions and sun rays were taken and recorded. A statistical model was developed using multi-nonlinear regression analysis to predict spontaneous combustion liability. The correlation coefficients were calculated around 0.95 levels. It was found that the model established can predict the effects of atmospheric conditions on the spontaneous combustion of coal stockpile.

Deng *et al.* (2015) evaluated the effects of pyrite contents on one of the factors affecting spontaneous combustion liability of coal. Coal samples with pyrite contents of 0%, 3%, 5%, 7%, and 9% were produced by blending coal and pyrite. The DSC was used to determine the intensity of heat discharged during coal oxidation for the different pyrite contents. The study indicated that the presence of pyrite can accelerate the propensity of coal spontaneous combustion. The DSC shows that the coal sample with a 7% pyrite content has the highest rate of heat flow. Samples with pyrite contents of 5% to 7% have the most significant influence on spontaneous combustion liability within the range of the study.

Chen *et al.* (2015) investigated the low-temperature oxidation characteristics and kinetic behaviour of some individual and blended coal. The blended coal was mixed in the ratio of 50% (50A50B, 50A50C and 50B50C respectively). The thermo-chemical kinetic characteristics and interaction during low-temperature oxidation of blended coal were studied with the use of a METTLER TGA thermogravimetric simulations thermal analyser. The blended coal with high volatile bituminous coal showed a synergetic interaction at a temperature range of 45 to 180^{o}C during low-temperature oxidation. The interaction at low-temperature oxidation of coal could be attributed to low moisture and some potential minerals that could increase the chemical inertness. The reaction model for the oxygen absorption and desorption stage was illustrated as a third order reaction mechanism model. The study provides a detailed observation on low-temperature oxidation of blended coal during low-temperature oxidation.

Gurdal *et al.* (2015) investigated the spontaneous combustion liability of the Can Basin coal (Canakkale, Turkey) and determined the composition of the gas produced from the coal during the combustion. The intrinsic properties and the gas composition of burned and partly burned coal samples were analysed. Furthermore, the mineralogical composition of the burning coal samples was investigated. It was found that the pyrite content is a significant factor that promotes spontaneous combustion liability in addition to the rank and moisture content. The use of X-ray diffraction (XRD) and scanning electron microscope (SEM) indicated the contents of pyrite, quartz, tridymite, kaolinite, amorphous matter, and gypsum in the sample. Fumarolic minerals (sulphur blooming and ammonium chloride) forming on the surface of the coal seams were monitored. Elements including beryllium, fluorine, scandium, cobalt, nickel, copper, zinc, arsenic, selenium, lead, titanium, molybdenium, zirconium and uranium were found to be higher than the world average.

Genc & Cook (2015) analysed and classified 119 coal samples from different coal seams and production Coalfields in South African collieries through a series of laboratory tests carried out between 2008 and 2012. A comprehensive data of the results is available to provide the basis for an improved risk evaluation method for spontaneous combustion. From the analysed results, it is very possible that the incidence of spontaneous combustion will increase from the current levels. This is due to factors such as an increased rate of mining, re-working of previously mined seams, more stopping and total extraction for underground mines and higher stripping ratios for surface mines leading to spoil. The results indicated that spontaneous combustion liability depends on the coal seam properties. Additionally, most of the collieries located in the Witbank and Highveld Coalfields have a high risk of spontaneous combustion. Fifty-two out of 119 collieries have a high risk spontaneous combustion liability rating, while most of the selected collieries possess medium risk ratings. The findings indicate the tendency for risk of spontaneous combustion without investigating the intrinsic properties of the coal.

Arisoy & Beamish (2015) evaluated the spontaneous combustion liability of coal using the data on the kinetics of coal oxidation reaction from an adiabatic oven testing. A time to temperature influence on the reaction kinetics was indicated based on repeated tests on the coal at different starting temperature (20 to 57^{o}C). The initial reaction rate was primarily non-Arrhenius at the low temperatures but crosses to Arrhenius behaviour at temperatures above 70^{o}C. A new rate model equation that considered the changes in kinetic behaviour of low temperature regions were generated. The influence of the starting temperature was reviewed in the rate equation for

fresh coal modelling studies. The new modelled equation allows for convergence in the Arrhenius equation at peak temperatures (>70^{o}C).

Chen *et al.* (2016) studied the oxidation dynamic variables of some coal samples treated by inhibition foam with the use of thermal analysis. The synergetic influence of water-based foams inhibitors to delay coal oxidation was examined. Sodium dodecyl sulphate (SDS) was used as the foaming agent. $MgCl_2$, $CaCl_2$, NH_4Cl and NH_4HCO_3 were chosen as chemical inhibitors. The chemical reagents employed in the study were analytically pure. An automatic surface tensiometer, BZY-3B was used to determine the surface tension of the solution. Coal samples (lignite) collected from the Malan colliery in the Shaxi province were pulverized to produce a particle size of 0.1 to 0.125mm. The selected chemical inhibitors aid the surface action of the SDS solutions to lower the surface tensions and critical micelle concentration (CMC). It was envisaged that the extent of chemical inhibitors to reduce the solution of CMC are in descending order of $MgCl_2$, $CaCl_2$, NH_4HCO_3 and NH_4Cl. The oxygen adsorption attained from the treated coal samples is lower than that obtained from the untreated coal. The apparent activation energy increases by adding inhibition foams. It was found that the inhibition foams could be used to delay coal oxidation.

Nimaje & Tripathy (2016) investigated the effects of coal intrinsic properties and spontaneous combustion liability indices on selected coal properties. Forty-nine *in situ* coal samples were analysed using the XPT, DTA, flammability Temperature (TP), Olpinski Index, wet oxidation potential (WOE) and intrinsic properties to measure the spontaneous combustion liability of coal. It was found form the statistical analysis that the parameters of ultimate analysis showed significant correlation with the Olpinski index as compared to the other liability indices.

Goede *et al.* (2016) evaluated the influence of moisture content in coal stockpiles of different particle sizes. Three particles sizes, fine (6 to 7mm), total range (-13.2mm) and coarse (-13.3+ 6.7mm) were used. In the first experiment, samples were loaded into sectioned 12 PVC pipes for each size range and exposed to air for 10 to 12 days to facilitate the process of evaporation. In the second experiment, samples were placed in three shallow containers and exposed to the air to determine the rate at which moisture evaporates from a stockpile surface. It was observed in the stockpile consisting of fine particles that the moisture initially evaporates at a higher rate than the coarser particles. The high rate of evaporation is restricted to the surface of the fine coal stockpile. The porous nature increases the depth at which evaporation occurs in the coarse coal. The steady state was obtained after the evaporation of moisture was observed up to the

4th day of each experiment. It was found that water evaporates from stockpile surfaces into the body of the stockpile depending on the void spacing and the particle size distribution of the coal.

Zhang *et al.* (2016) developed a two-dimensional numerical model to predict the spontaneous combustion liability of coal stockpiles. The model used chemical kinetic parameters (activation energy, E and pre-exponential factor, A2) for low-temperature oxidation measured with a tube reactor system. The kinematics parameters and other factors affecting coal stockpiles under various conditions were evaluated with the developed model. In the model, the evaporation rate of moisture from coal was limited by the heat from oxidation and the extent of moisture saturation in the bulk gas mixture. From the simulation results, the process of self-heating could be divided into three stages; temperature-rise stage, constant-temperature stage and ignition-stage. Twelve tests were conducted to examine the impacts of different factors. Among these factors which affect the temperature rise includes, wind velocity, coal type, heat loss from the bottom, stockpile height and initial temperature of the stockpile, while air humidity and particle size have small effects. It was found that good ventilation, low stockpile height and ground material with a high thermal conductivity can be used to enhance the heat loss and prevent the spontaneous combustion of the coal stockpile.

Kim & Sohn (2016) used a numerical method to investigate the effects of wind barrier design and closed storage on spontaneous combustion liability of coal stockpiles. A single and a dual wind barrier were selected, their suppression effects were investigated based on the distance between the barrier and the stockpile. The correlations between the ignition delay and the distance were observed. The distance promotes self-heating slightly with a single barrier. The self-heating advances as the distance increases with a dual barrier. A dual barrier with longer distance than a critical one has no retarding effect as compared with the stockpile without any barriers. It was found that a closed stockpile is proposed for a method to minimize self-heating.

Qi *et al.* (2016) investigated the thermodynamic features of three different coal samples to spontaneous combustion under low oxygen concentration. The experiments were conducted under varying ranges of oxygen concentrations, 5%, 7%, 10%, 13%, 16%, 19% and 21% respectively. The mechanism function, pre-exponential factor A and activation energy E under different conditions were analysed. The results indicated varying temperature distribution during coal spontaneous combustion. The kinetic parameters depend on the coal ranks and reaction stages. The changing trends varied at various reaction stages for the same and different

coal samples. This was due to different effects of oxygen concentration on the micro-reaction pattern during coal spontaneous combustion. This study may be useful to understand the actual development process of coal spontaneous combustion.

Ajrash *et al.* (2016) evaluated the spontaneous combustion behaviour of coal dust layers in a humid environment with a relative humidity of >80%. The Minimum Auto-Ignition Temperature (MAIT) of four different coal dust samples (Australian) with particle sizes below 212µm were used. The dust layer auto-ignition temperature apparatus in accordance with the ASTM E-2021 standard was used to measure coal dust layer thickness of 5, 12, and 15mm. The MAITs for coal dust layers increases with increasing particle sizes. The MAIT for the coal samples with a smaller diameter of 50mm sizes were observed to be lower in comparison with samples with a larger diameter of 50mm size. It was found that the MAIT increased proportionally with the increasing thickness of the coal dust layer. Exposure of the coal bed to relative humidity had no significance on the coal's MAIT.

Ward (2016) evaluated the techniques used to identify and quantify the mineral matter within a coal sample. The techniques used were optical microscopy, low temperature-plasmashing, quantitative evaluation of x-ray diffraction data, a combination of observation and chemical analysis. A comprehensive knowledge of mineral matter is vital in understanding the mode of occurrence, origin, environment effect and potential economic value of trace elements in particular coal seams. The study provides a refine base for understanding coal formation and evaluating the reaction of specific coal to different combustion, gasification and cooking processes.

Restuccia *et al.* (2017) investigated the spontaneous combustion liability of shale rock samples produced from coastal cliffs of Kimmeridge Bay (UK) at different ambient temperatures. The Frank-Kameneski theory of ignition was used for various particle sizes of the samples. The kinetic parameters for two particle sizes of shale were evaluated to upgrade the results of the stockpiles and the deposits in varying bands. The Frank-Kamenetski theory of ignition indicated that the Arrhenius reaction applies as the energy was noted to be persistent for the temperature variation. It was found that the particle size distributions have a great influence on reactivity, with coarse particles being less reactive than finer particles. The study showed that shale rocks are reactive depending on the particle diameter and that self-heating is possible for small particles in stockpiles and geological deposits exposed to atmospheric conditions.

2.7 Effects of chemical inhibitors on spontaneous combustion of coal

The influences of an additive on the start of coal oxidation are considered from theories which explain the phenomena in purely physical perception with the idea of developing a rule to investigate suitable chemical inhibitors to manage the occurrence of coal spontaneous combustion. The risk constituted by coal fires involves air pollution, great loss of precious resources, ground surface subsidence and methane leakage (Bo-tao *et al.*, 2009). A variety of historical techniques such as injecting inert gases, spraying resistant agents and infusing gelatum has been developed and used globally to prevent spontaneous combustion (Fu-bao *et al.*, (2007) and Michalski (2004)). The use of techniques such as nitrogen infusion, air leakage, pressure ventilation and grouting delays the process of self-heating by reducing the oxygen concentration (Wang *et al.*, 2013). Inhibitors and retarding foams have the influence to isolate the coal material from oxygen. In addition, it gives a depressive catalytic impact on the combustion process and can successfully slow down spontaneous combustion (Luo, 2003). Chemical inhibitors are generally used to prevent coal mine fires and have been applied in coal mines to minimize coal oxidation either by reducing the production of active groups or preventing the free radical reactions (Qi *et al.*, (2012) and Wang *et al.*, (2013)). Such chemicals comprise of antioxidant and inorganic salts (Smith et al., (1988), Sujanti & Zhang (2000), Watanabe & Zhang (2001) and Sujanti & Zhang (2001)). These chemical inhibitors hinder coal oxidation simply by generating a check to oxygen and by the adsorption of water.

Wang *et al.* (2012) investigated the influence of ionic liquids to retard coal oxidation. The liquid solvated a variety of functional groups and reduced the exothermic oxidation of coal. Zhan *et al.* (2011) found that Na_3PO_4 plays a vital action to change the disintegration pathways of hydroxyl radicals, thus making better the thermal stability. Sujanti & Zhang (2000) and Watanabe & Zhang (2001) investigated the influence of additives on the self-heating by evaluating the critical ambient temperature of a coal blended with a variety of inorganic salts. The analyses indicated that $CaCl_2$, $Ca(Ac)_2$, $Mg(Ac)_2$, $MgCO_3$, NaCl and NaOH might retard the coal oxidation process. Smith *et al.* (1988) indicated that $NaNO_3$, NaCl and $CaCO_3$ could delay the spontaneous combustion of coal. The influences of inorganic compounds on spontaneous combustion of coal are dependent on changes in thermal development of self-heating and the estimation of apparent activation energies for coal oxygen adsorption (Smith *et al.*, (1988), Sujanti & Zhang (2001) and Watanabe & Zhang (2001)).

Spontaneous combustion is one of the major challenges in the coal mining industry. Irrespective of the detailed studies that have been carried out since the beginning of coal mining, spontaneous combustion is still a problem at present. Serious incidents of spontaneous combustion were reported as a result of spontaneous combustion of coal-shale. Coal-shale can start spontaneous combustion depending on the various organic matter within the material and together with the nature of the associated coal (rank). Limited studies have been conducted to investigate factors affecting the spontaneous combustion liability of coal-shale in the coal mines, therefore establishing reliable methods to predict and control the occurrence of spontaneous combustion is necessary but yet to be investigated. The characteristics of coal and coal-shale intrinsic properties between bands of coal seams affecting spontaneous combustion is yet to be studied. There were conflicting views whether coal-shale can initiate spontaneous combustion or not after a visit to four (4) selected open cast mines in South Africa. The indication by the mines proved that self-heating of coal-shale has an effect on spontaneous combustion in mines. Studies reported by Kim & Chaiken (1990), Restuccia et al. (2017) and Rumball et al. (1986) showed that carbonaceous shale can cause spontaneous combustion but the inherent properties of this material with associated coal properties is yet to be investigated. The intrinsic properties of coal, coal-shale and other related carbonaceous materials can be a parameter to investigate the cause of spontaneous combustion in coal mines. Therefore, self-heating of coal-shale can be reasoned to cause spontaneous combustion. Considering these concerns raised, a detailed investigation of the major factors causing spontaneous combustion of coal and coal-shale is required.

2.8 Summary

This chapter has reviewed the mechanism of spontaneous combustion, areas of spontaneous combustion, various factors affecting spontaneous combustion, and different experimental studies to predict spontaneous combustion liability. The prediction of the spontaneous combustion liability of coal is a difficult task due to various factors involved. There are several competing effects and mechanisms in coal oxidation that decides whether spontaneous combustion can occur. The whole result can be overseen by the heat balance, which is a function of the relationship between coal intrinsic properties and other external factors. Regardless of the extensive studies that have been conducted over time, the cause of spontaneous combustion is still a major challenge in the coal mines. Spontaneous combustion of coal is a well-known phenomenon around the globe. Apart from the coal itself, burning coal-

shale is becoming a problem in some South African coal mines which is yet to be evaluated. The experimental techniques used in this study are discussed in the next Chapter.

3 EXPERIMENTAL METHODS

3.1 Introduction

This Chapter describes the nature of the Witbank Coalfields as well as the experimental procedures and spontaneous combustion test results obtained for the selected coal and coal-shale samples. The origin of the coal seams is discussed in section 3.2. In section 3.3, the techniques used for the sample collection are described. A description of the procedure used for the overall geochemical tests is discussed in section 3.4. The Wits-Ehac tests used to measure the spontaneous combustion liability of the samples is described in section 3.5. Section 3.6 consists of a newly developed device (Wits-CT apparatus) and the testing procedure to predict the spontaneous combustion liability of coal and coal-shale. In section 3.7, the statistical method used to establish some fundamental relationships and trends between selected the intrinsic properties and the spontaneous combustion liability index of coal and coal-shale are described. Section 3.8 describes an experimental approach of selecting chemical inhibitors to minimize the spontaneous combustion liability of coal and coal-shale and section 3.9 contains a summary of the experimental procedures. Applied experimental methods at various stages in this research are illustrated in Figure 3.1. The test results of the samples investigated contain the following sections:

1. The spontaneous combustion liability index of the coal and coal-shale using a newly developed medium-scale test (referred to as the Wits-CT apparatus) and small-scale test (the Wits-Ehac apparatus);
2. Comparison and validation of results of the Wits-CT Index and the Wits-Ehac Index with the incidence of spontaneous combustion in the mines;
3. Geochemical tests (proximate, ultimate, total and sulphur forms, petrographic composition, XRF and XRD analysis), their influences and relationships on the spontaneous combustion liability tests;
4. Develop models using statistical analysis to predict the incidence of spontaneous combustion in coal mines; and
5. Investigate selected chemical inhibitors to minimize spontaneous combustion liability.

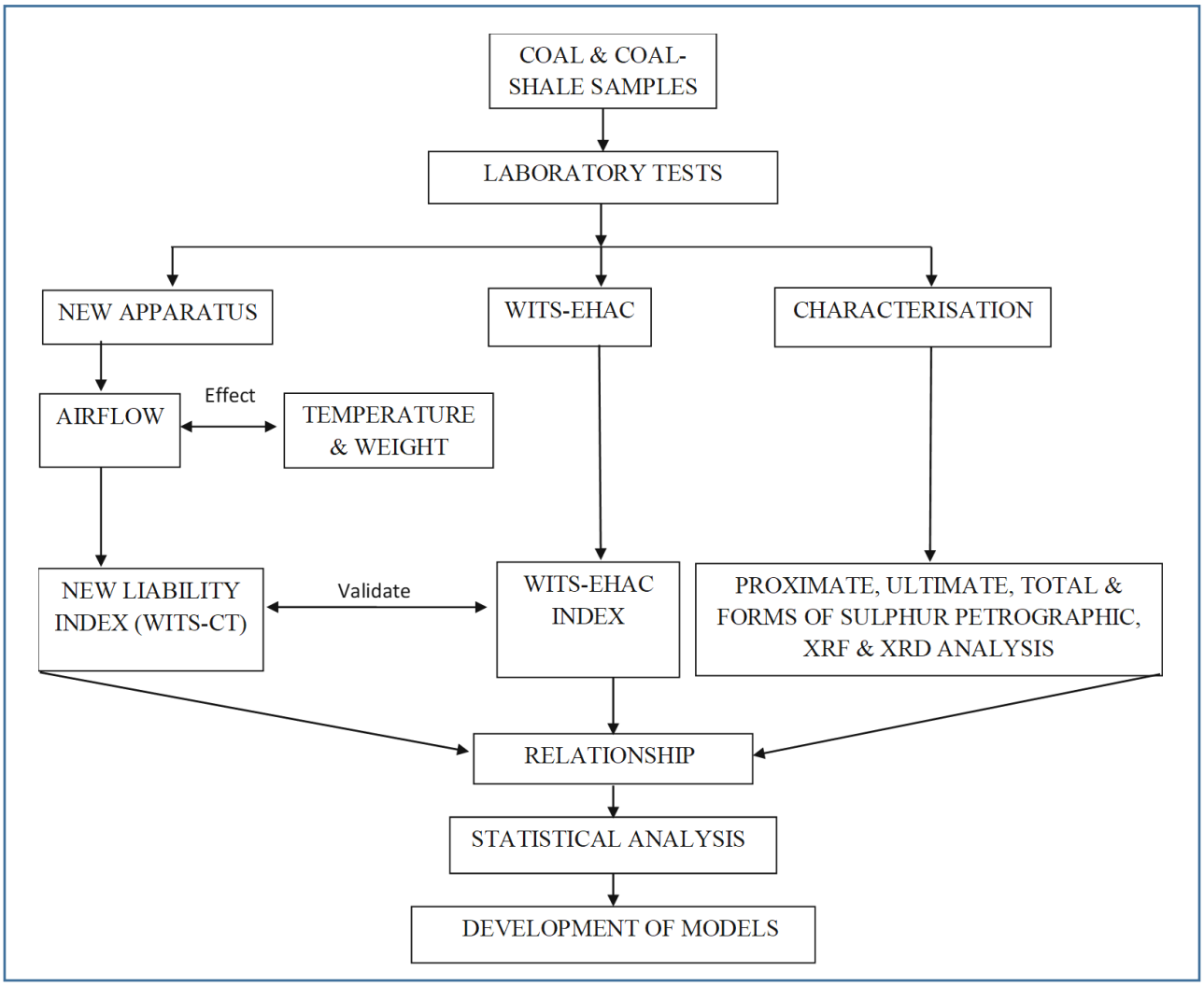

Figure 3.1 Experimental flow chart

3.2 Coal seam description of the study area

The Witbank Coalfields seams have different characteristics that makes it one of the commercial markets in the power generation, metallurgical, liquefaction, domestic and chemical sectors. The coalfield consists of five (5) seams with a thick sedimentary sequence (shale, mudstone, sandstone, and siltstone) as shown in Figure 3.2. Following the order from the base upward, the seams are numbered from 1 to 5. The distribution of the No.1 and 2 seams is dominated by the Pre-Karoo topography, while the No. 4 and 5 seams is eroded over a wider area (Snyman, 1998). The parting thickness, which is roughly 8m thick between the seams is usually constant over the entire Coalfields except towards the northern limits where the stratigraphic units are thinner (Smith & Whittaker (1986) and Pinheiro (1999)). The Witbank seams are briefly discussed below and the generalised stratigraphic column for the Vryheid Formation in the Witbank Coalfields (individual coal seams are numbered from No. 1 at the base to No. 5 at the top) is shown in Figure 3.2

i. The No.1 coal seam is a high grade seam coal suitable for export after beneficiation (Smith & Whittaker (1986) and Snyman (1998)). It comprises primarily of lustrous to dull coal with shaly sandstone partings which resulted in localised No. 1 lower seam around the Arnot area. The seam has low phosphorus content and in most cases, it is mined separately as metallurgical feedstock (Pinheiro, 1999).

ii. The No.2 coal seam is the main source of high-yield export quality steam coal (Pinheiro, 1999). The seam is the major seam of the Witbank coalfield. It comprises of various sandstone and shale partings, with the thickest parting existing locally in the upper part of the seam. This results in the splitting of the seam into No.2 and No. 2 upper seam (Smith & Whittaker (1986) and Snyman (1998)). It has a varying thickness and reaching maximum thickness of 8m in the Ogies area but thinning to approximately 3m towards Arnot Colliery in the East. It is mined primarily for the production of low-ash metallurgical coal and export steam coal. The upper part of the seam is usually shaly and unmineable. The better quality lower part of the seam is exploited by selective mining operations (Smith & Whittaker, 1986).

iii. The No. 3 seam is inconsistently developed and thin, rarely exceeding 0.5m thick in most areas. It is usually evaluated to be uneconomic because of its thickness. It is locally of high quality which makes it possible for the seam to be mined by opencast methods where a thickness of 0.8m is reached (Smith & Whittaker, 1986).

iv. The No. 4 seam has varying thickness of between 2.5m and 6.5m in the central Witbank area (Smith & Whittaker, 1986). It consists of different sandy mudstones and siltstone partings. This splits the seam into No.4 seams, occasionally called No.4 lower, No.4 upper and No.4A seams. The partings are typically not too thick for selective mining (Pinheiro, 1999). The seam is of poor quality and contains primarily of dull to dull lustrous coal with the upper portion being of poor quality.

v. The No.5 seam rarely exceed 2m. It comprises of a mixture of mainly bright, banded coal with thin shale and mudstone partings. The seam has been broadly eroded over wide areas (Smith & Whittaker, 1986). The seam has been used by the metallurgical industries for the local steel production (Bell & Spurr, 1986a).

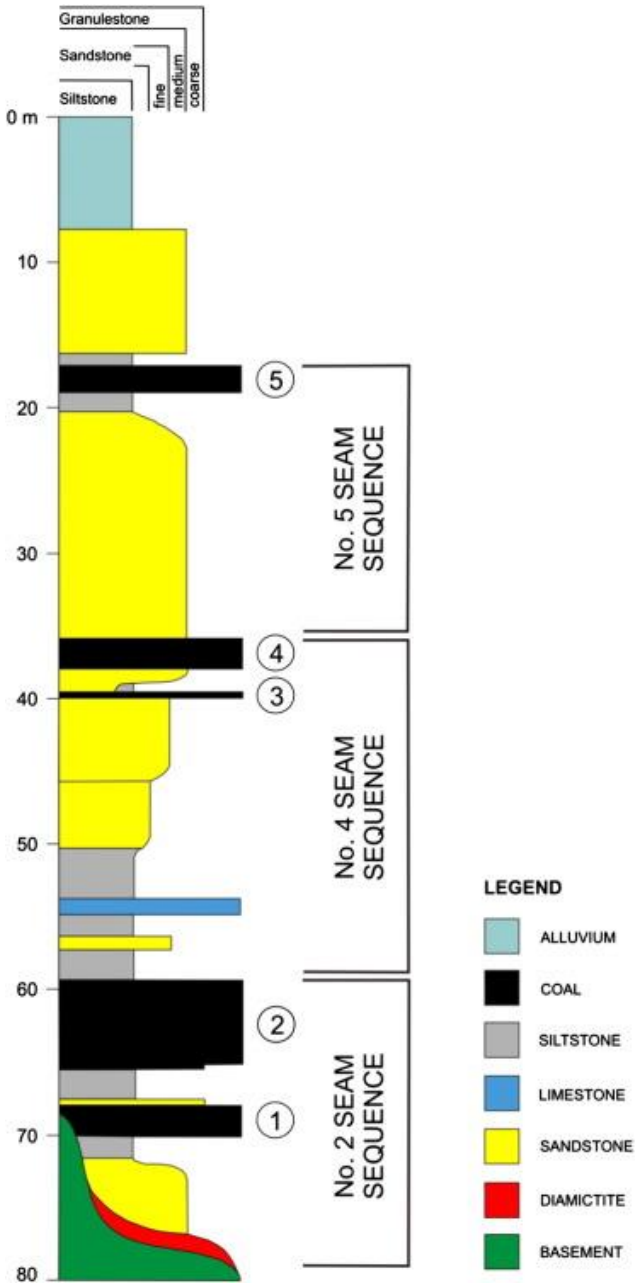

Figure 3.2 Generalised stratigraphic column for the Vryheid Formation in the Witbank Coalfield. Individual coal seams are numbered from No. 1 at the base to No. 5 at the top (Hancox & Gotz, 2014).

3.3 Sample collection

The coal and coal-shale used in this study were collected from four different coal mines (iMpunzi Mine, Goedgevonden Colliery, Khwezela Mine, and Tweefontenin Mine) in Witbank Coalfields, South Africa using the ply sampling technique. Fourteen (14) samples representative of *in-situ* coal (bituminous) and 14 coal-shale samples were collected from areas

known to be self-heating, highwalls and selected bands between the coal seams (above and below). The samples were kept in airtight bags to avoid moisture loss and further oxidation; the bags are made of aluminium-coated polyester which allows minimum reduction of vapour and oxygen permeability. Each ply sample bag was clearly labelled with a chosen number. The samples were stored and prepared under laboratory conditions (room temperature).

3.4 Sample preparation and Geochemical tests

Sample lumps were reduced using a ROCKLABS MK III crusher as shown in Figure 3.3 to suitable sizes (>10mm). The samples were milled, blended and divided using a centrifugal mill (Retsch ZM 200) as shown in Figure 3.4 to generate different sub-samples as required for each test, -250µm for geochemical tests, -212µm for spontaneous combustion tests, and -1000µm for petrography analysis). Samples for characterisation tests were prepared in accordance with the ASTM, the International Standard and Organisation (ISO) and the South African National Standard (SANS).

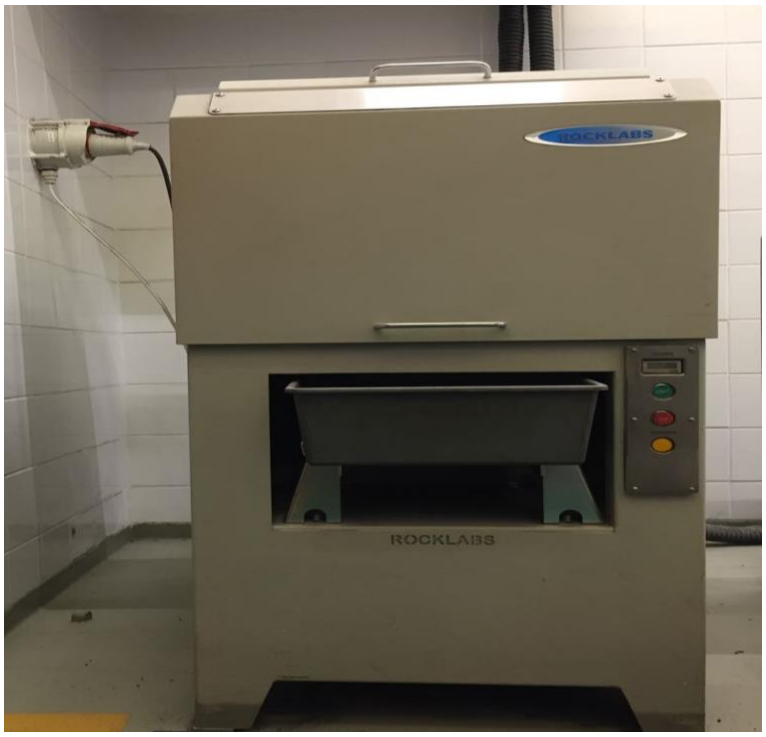

Figure 3.3 Crusher (ROCKLABS MK III)

Figure 3.4 Centrifugal mill (Retsch ZM 200)

3.4.1 Proximate Analysis

The determination of the proximate analysis (moisture, ash and volatile matter contents) were carried out in accordance with the ASTM (D3173-17a), ASTM (D3174-11) and ASTM (D3175-17) standards. Approximately one gram (1g) each of the coal and coal-shale samples were used for the analysis. Fixed carbon was obtained by subtracting the sum of the percentage of volatile matter, ash and moisture from 100. The analytical technique for each method is described below;

(i) Moisture content as determined by ASTM (D3173-17a)

The moisture content was determined by measuring the weight loss in each sample that has been pulverized to pass through -250µm, expressed as a percentage. An accurately weighed sample (1g) was maintained under controlled conditions of 110^{o}C in an air-oven as shown in Figure 3.5 for 1hr. The sample was allowed to cool to room temperature and weighed as shown in Figure 3.6. The moisture content is calculated from the loss in mass as indicated in Equation 3.1.

$$M\ \% = \frac{W_1 - W_2}{W_1} \tag{3.1}$$

where, $\boldsymbol{M}\%$ is the percent moisture; $\boldsymbol{W_1}$ is the initial weight; and $\boldsymbol{W_2}$ is the final weight.

Figure 3.5 Oven (EcoTherm)

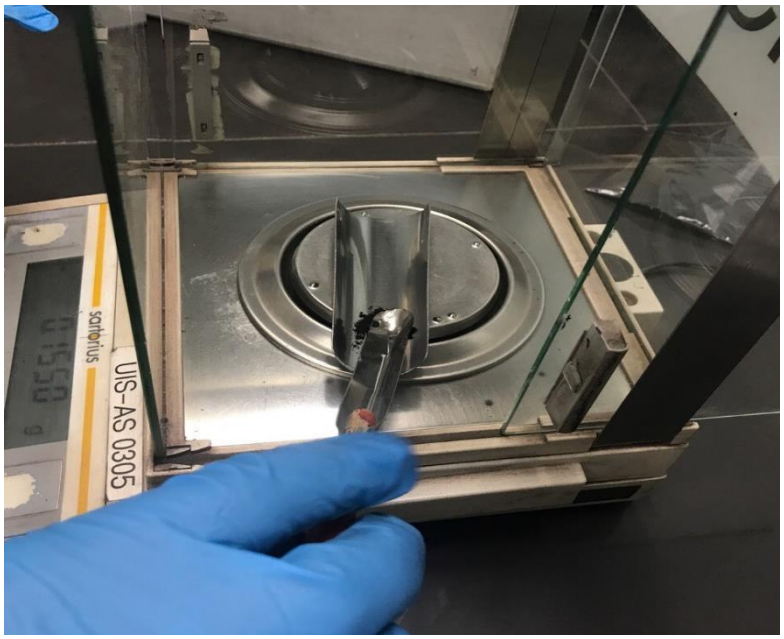

Figure 3.6 Weighing balance (SHIMADZU TXB2201L)

(ii) Volatile Matter content as determined by ASTM (D3175-17)

The volatile matter is the percentage of volatile products excluding moisture vapour that is released during the heating of coal under controlled conditions. In this test, an accurately weighed sample (1g) was placed in a pre-weighed platinum crucible (10 to 20mL) with a close-

fitting cover. The crucible was then suspended at a definite height in the furnace chamber as shown in Figure 3.7. The temperature in the region where the crucible was suspended in the furnace was maintained at 950°C. The cover of the crucible was tapped to confirm the lid was well sealed to avoid the ingress of air after the rapid release of the volatile matter was shown by the disappearance of the luminous flame. The crucible was removed from the furnace and cooled after heating for seven (7) minutes. The crucible was weighed immediately after it was cooled. The volatile matter was determined by the percentage loss of weight minus the percentage moisture as shown in Equation 3.2.

$$VM\ \% = \frac{W_1 - W_2}{W_1} - Moisture\ \% \quad (3.2)$$

where, $\boldsymbol{VM}\%$ is the percent volatile matter; $\boldsymbol{W_1}$ is the initial weight; and $\boldsymbol{W_2}$ is the final weight.

Figure 3.7 UltraFurn Muffle Furnace SAF 11(SPT) with J4 Controller

(iii) Ash as determined by ASTM (D3174-11)

An accurately weighed sample (1g) was placed in a pre-weighed platinum crucible (10 to 20 mL) with a close fitting cover in a ventilated muffle furnace at 750°C for 4hrs. The crucible was removed from the furnace and cooled after heating. The crucible was weighed immediately after it was cooled. The ash content was determined by the ratio of percentage loss of weight to initial weight of sample as shown in Equation 3.3.

$$A\ \% = \frac{W_3 - W_1}{W_2}\ x\ 100\% \quad (3.3)$$

where, $\boldsymbol{A}\%$ is the percentage of Ash content; $\boldsymbol{W_1}$ is the weight of the crucible; $\boldsymbol{W_2}$ is the weight of the sample and $\mathbf{W_3}$ is the weight of the ash and crucible.

(iv) Fixed carbon

The percentage of fixed carbon (FC%) is derived by subtracting the moisture, volatile matter and ash from the value 100 as shown in Equation 3.4.

$$FC\% = 100 - (\%M + \%VM + \%A) \quad (3.4)$$

where, $\%\boldsymbol{M}$ is the percentage of moisture (air-dry basis); $\%\boldsymbol{VM}$ is the percentage of volatile matter (air-dry basis); and $\%\boldsymbol{A}$ is percentage of ash (air-dry basis).

3.4.2 Ultimate and Total Sulphur Analysis

The carbon (C), hydrogen (H), nitrogen (N) and sulphur (S) content of coal and coal-shale samples were carried out according to ISO standards, 12902:2001, for CHN and S content respectively using a LECO TruSpec CHN and LECO C & S analyser for the total sulphur as shown in Figure 3.8 and Figure 3.10. Approximately 0.25g of the samples were used for the analysis at a temperature of up to 950^{o}C with an analysing time between 60-300 seconds.

(i) Carbon, Hydrogen, Nitrogen and Total sulphur analysis

The Dumas method of combustion was used for the analysis and provides the results of the elemental constituents with an optimized detector within four minutes. Three phases were involved in the experiments, purge, burn and analysis. An accurately weighed sample (1g) was placed into a 502-186 Tin-Foil Cup and covered as shown in Figure 3.9. The sample weight and identity were saved into the sample login. The sample was placed into the carousel and the analysis proceeded. In the sample-drop purge phase, the sample was placed in the loading head, sealed and purged of any atmospheric gases that were present during the sample loading. The sample was dropped into the primary furnace (950^{o}C) and flushed with pure oxygen for quick combustion at the burning stage. The combustion gases in the ballast became homogeneous by means of passive mixing during the analysis stage. A series of infrared detectors determined the evolved gases of carbon and hydrogen. In addition, a 3cc aliquot was captured in a loop before the ballast piston was forced down to evacuate the ballast. The sample aliquot gases were swept through hot copper to eliminate the oxygen and changed NOx to nitrogen (N). The

Lecosorb and Anhydrone were used to remove carbon dioxide and water. A thermal conductivity detector was used to determine nitrogen. The equipment is shown in Figure 3.8.

(ii) Oxygen content as determined by ASTM (D3176-15)

The percentage of oxygen content (O%) on an air-dried basis is derived by subtracting the percentage moisture, ash, carbon, hydrogen, nitrogen and sulphur from the value 100 as shown in Equation 3.5.

$$O = 100 - (\%C + \%H + \%N + \%S + \%M + \%A) \quad (3.5)$$

where, **%C** is the percentage of carbon, **%H** is the percentage of hydrogen, **%N** is the percentage of nitrogen, **%S** is the percentage of total sulphur, **%M** is the percentage of moisture and **%A** is the percentage of ash.

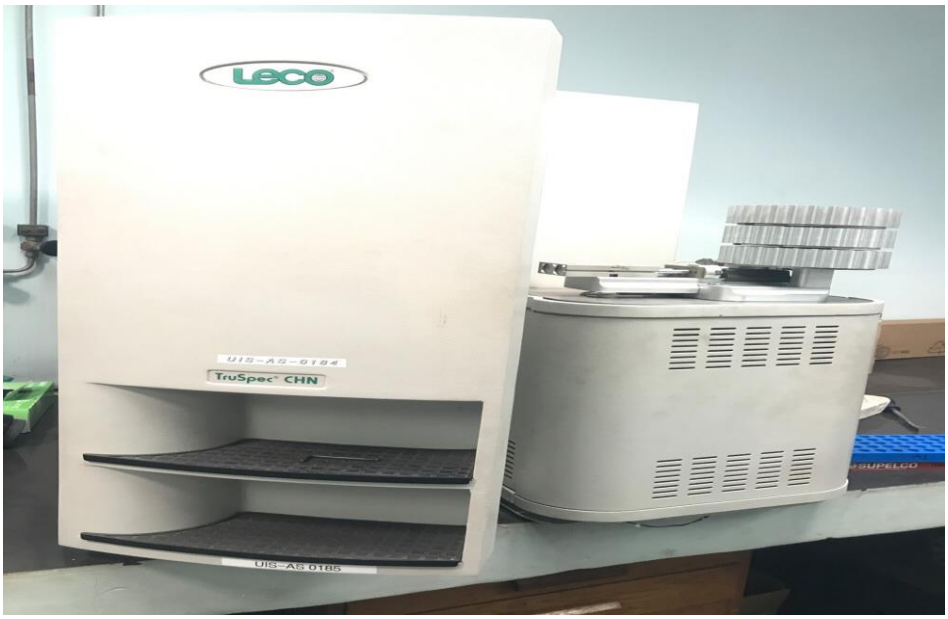

Figure 3.8 LECO TruSpec CHN Analyser

Figure 3.9 A 502-186 Tin-Foil Cups - LECO TRUSPEC

(iii) Total sulphur

Coal and coal-shale samples were melted in a pure oxygen atmosphere, making carbon to react to a mixture of carbon monoxide (CO) and carbon dioxide (CO_2) while sulphur reacts to sulphur dioxide (SO_2) inside the induction furnace. The combustion gases were purified after passing through a dust filter and moisture absorber. Thereafter, the infrared cells detected the sulphur dioxide. After the sulphur measurement, the oxidation of both carbon monoxides to carbon dioxide and sulphur dioxide to sulphur trioxide (SO_3) followed. The SO_2 gas was removed with cellulose wool and the carbon content is detected by infrared cells.

Figure 3.10 LECO C & S (CS23OSH)

(iv) Determination of the forms of sulphur by ASTM (D2492-12)

The pyritic sulphur of the samples were detected in selective reaction when diluted with hydrogen nitrate (HNO_3). The sulphide in the solution was analysed by Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES) as shown in Figure 3.11. The solution is filtered to separate the sulphates from the pyrites. Pyritic sulphur was obtained by determining the soluble iron (pyritic iron) in the precipitate by ICP-OES and converting it to percentage sulphur as pyritic. The sulphate sulphur of the samples was detected in selective a reaction with hydrogen chloride (HCl) and analysed by the photometric method with barium sulphate ($BaSO_4$). The organic sulphur is expressed as the difference between the total sulphur and the sum of the sulphate and pyrite sulphur (ASTM, D2492-12).

$$\%\, Organic\ sulphur = \%Total\ sulphur - \%sulphate\ sulphur \qquad 3.6$$

Figure 3.11 ICP-OES Spectrometer (Perklin Elmer Optima 5300DV)

3.4.3 Petrographic analysis

This study used the petrological microscope to study the relative amounts of various maceral groups and mineral matter present in the coal and coal-shale samples. The macerals are categorized into three groups; vitrinite, liptinite and inertinite. The classification of macerals is based on the classification of the International Committee for Coal and Organic Petrology (ICCP 1998 and ICCP 2001). Vitrinite describes a group of macerals with a grey colour and whose reflectance is usually between that of the related darker liptinite and lighter inertinites. The inertinite group of macerals are readily recognised due to their high reflectance among the whole maceral group. The inertinite group shows a grey to white colour subject to the degree of metamorphism and demonstrates a higher reflectance than vitrinite of the same sample. They are known as high carbon and low hydrogen contents because of aromatization and condensation processes. The classification of macerals in the inertinite group is dependent on the intensity of alteration, the degree of preservation and vegetation origin. The liptinite group macerals display a deep grey colour and is the darkest among the entire maceral group. Random vitrinite reflectance analyses were conducted to evaluate the petrographic composition and rank of the coal but not on the coal-shale due to the limited amount of vitrinite. The technique used was based on the procedure laid down in accordance to the SANS, 7404 Parts 1-5, under oil, using a reflected light microscope.

(i) Preparation of petrographic blocks

The properties of a given coal are determined by the amount and distribution of the minerals and macerals present including the coal rank. The petrographic blocks used for the petrographic analyses were produced by mixing 2 to 3g of -1000µm grain size sample, with epoxy resin in a 3.2cm diameter phenolic ring form moulds. The moulds were allowed to cure, and ground with the use of 320, 400 and 1000 grit papers. Once ground, they were polished using a 0.3 and 1.0µm alumina slurries. Final polishes were achieved with the use of colloidal silica, which provides a relief free and scratch-free surface for analysis by reflectance microscopy as shown in Figure 3.12.

Figure 3.12 Petrographic blocks

(ii) Maceral Analysis

Maceral analysis including mineral matter was conducted to evaluate the organic and inorganic content of the coal and coal-shale. The maceral analysis was carried out on the polished sections as shown in Figure 3.12 under white incident light and fluorescent light under the microscope. A Zeiss Axio Imager M2M reflected light petrographic microscope fitted with an oil immersion lens at a total magnification of X 500 and Diskus software was used at the Department of Geology, University of Johannesburg, South Africa, as shown in Figure 3.13. The point count analysis was conducted with the help of a mechanical stage that advances the sample laterally in equal steps with a single count per particle. The spacing between the counts was maintained at 0.4mm. Over five hundred (500) counts were taken on each sample and twenty-four (24) mineral groups were quantified. The macerals results are reported as volume percent (vol.%), inclusive of mineral matter or mineral matter free basis (mmf).

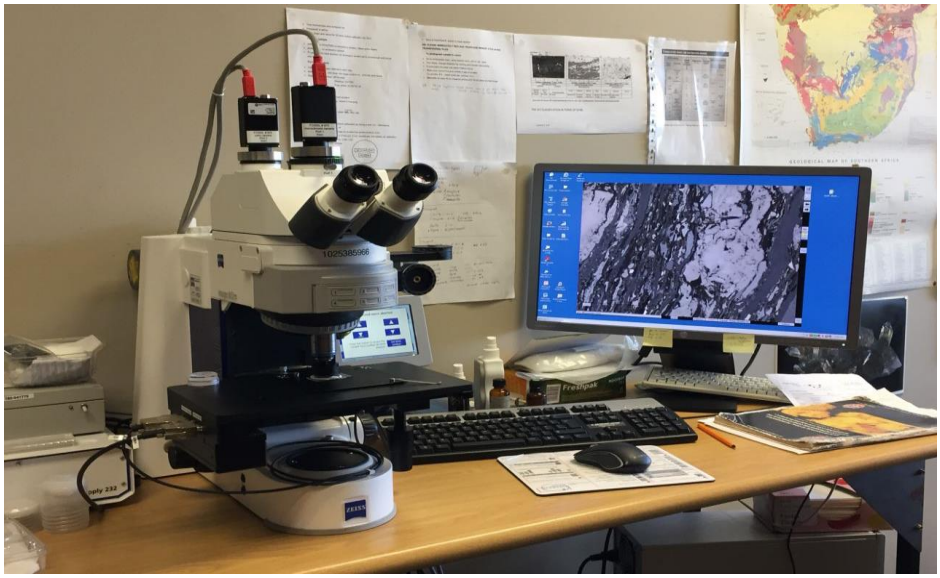

Figure 3.13 Zeiss Axio M2M reflected light petrographic-microscope house at the Department of Geology, University of Johannesburg, South Africa

(iii) Rank analysis

Vitrinite reflectance was conducted under the same microscope using the same sample and the Diskus Fossil software. The system was calibrated using a YAG (0.900) calibration standard (calibration services used for laser power and energy calibrations and wavelengths). Where there was a suitable population of collotelinite, over one hundred (100) counts were taken on each sample. In certain samples where the collotelinte and total vitrinite was less than 10%, a limited amount of particles was available for the vitrinite reflectance analysis. Vitrinite reflectance measurements on the coal samples were carried out on vitrinite particles lacking microfractures, impurities and oxidation rims. Due to the low organic content in the coal-shale samples, no reflectance analysis was conducted.

3.4.4 X-ray fluorescence analysis as determined by ASTM (D4326-13)

The ash oxide and the inferred mineral matter were determined by the X-ray fluorescence (XRF) method. Coal and coal-shale ash were prepared by placing a weighed amount (5g) of -250µm in a cold muffle furnace according to the ASTM (D-3682-13). The temperature was gradually raised to 500^{o}C in 1hr and to 750^{o}C in 2hrs as shown in Figure 3.14. At the end of the heating period, the sample was removed from the furnace, covered, allowed to cool and then weighed. The ash was fused with a lithium tetraborate ($Li_2B_4O_7$) and cast into a glass disk. SuperQ software was used to determine the major elements. The instrument used for the various elemental concentrations is called AxiosmAX and was conducted at the School of

Geoscience, University of the Witwatersrand Johannesburg, South Africa. Figure 3.15 and Figure 3.16 show the sample glass disk and the AxiosmAX analyser. The major elements were measured in weight percentage (wt.%) and reported as the oxide. A brief description of the procedure used for the XRF analysis follows;

(i) Procedure to Prepare Fusion Disks for Major Analysis

- Clean platinum (Pt) crucibles and porcelain loss on ignition (LOI) dishes were placed in the furnace at 1020^{o}C for half an hour to remove any organic residues;
- Approximately 1g of each sample was placed into each LOI dish after been cleaned and cooled. Both the LOI dish identification number (ID) and the weight of the LOI dish before and after each addition were saved on the computer. It was ignited at 1020^{o}C for 40 minutes. It was removed from the furnace after the analysis and allowed to cool in a desiccator;
- Approximately 1.75g of flux (Spectroflux 105) was weighed into each platinum crucible. Both the Pt crucibles identification number and the weight of the Pt crucible before and after each addition were saved on the computer. It was ignited at 1020^{o}C for 40 minutes. It was removed from the furnace after the analysis and allowed to cool in a desiccator;
- The hot plate was on for a minimum of 1hr prior to pouring;
- Once both the LOI and the Pt crucibles cooled down, the after ignition weights of the LOI dishes were measured and recorded in order to determine the samples LOI. The weights of the Pt crucibles to obtain the actual flux weights were similarly measured and recorded after ignition;
- The weight of the Pt crucible after the addition was recorded. It was ignited at 1020^{o}C for 50 minutes. It was removed from the furnace after analysis and allowed to cool in a desiccator; and
- The Pt crucibles were weighed and recorded and returned to the furnace at 1020^{o}C. The fusion beads were poured and pressed on the hot plate. Once all samples were poured and the Pt crucibles were cooled, the Pt crucible IDs were mapped out in order to label the beads the following day. The beads were allowed to set for 90 minutes on the hot plate. The hot plate was switched off and allowed to harden overnight. The Pt crucibles were cleaned in dilute HCl for 90 minutes on the hotplate and allowed to cool overnight.

The remaining ignited material from the LOI dishes was transferred to vials and store for future use.

Figure 3.14 Muffle furnace FM 22

Figure 3.15 Sample glass disc

Figure 3.16 XRF-WD (Axios mAX)

3.4.5 X-ray diffraction determination

The mineral composition was determined using a back loading preparation method on the PANalytical Empyrean diffractometer as shown in Figure 3.17 with a PIXcel detector and fixed slits with Fe filtered Co-Kα radiation. The phases were identified using the X'Pert Highscore plus software. The relative phase amounts (weight %) were estimated using the Rietveld method (a method used in the characterisation of crystalline materials).

Figure 3.17 PANalytical Empyrean diffractometer

3.5 Wits-Ehac test

The Wits-Ehac Index was developed in South Africa in the late 1980's to test the spontaneous combustion liability of coal (Eroglu (1992), Genc *et al.,* (2018), Genc & Cook (2015), Gouws & Wade (1989a), Gouws & Wade (1989b), Gouws & Eroglu, (1993), Uludag *et al.*, (2001) and Wade (1989)). The apparatus consists of an oil bath, six cell assemblies, (three (3) for the coal and the other three for the inert material (calcined aluminium)), an oil circular, a heater, a flowmeter, an air supply compressor and a computer (Wade *et al.*, 1987) as illustrated in Figure 3.18. Freshly pulverized (-212µm) dried coal samples (under room temperature) of 20-25g is used. The test apparatus is used to test coal under predefined conditions and a spontaneous combustion liability index is obtained. The system incorporates the determined XPT and DTA from selected coal samples with periodic measurements of temperatures to obtain a consistent self-heating liability index termed as the Wits-Ehac Index. The coal temperatures are recorded every 30 seconds by the computer for the oil in the bath to be heated to 200^{o}C. When using the DTA, the difference in temperatures between the coal sample and an inert material sample is measured via a data logger, stored in a computer and plotted against the temperature of the inert material sample. When the temperature difference between the inert material and the coal sample is plotted against the inert temperature, the part of the graph where the coal is heating faster than the inert sample (where an exothermic reaction occurs) is termed Stage II. It is important to understand that during the DTA, three stages are obtainable. At first, the temperature of an inert material sample is higher than the temperature of the coal sample (Stage I), which is dependent on the cooling effect of the evaporation of the coal sample moisture content. Secondly, during the evaporation of the moisture content, the coal sample begins to heat up at a higher rate than the heating rate of the inert material (Stage II) and this is based on the liability of coal to self–heating and attempting to reach the temperature of the surrounding temperature (oil bath temperature). Lastly, the high exothermicity is reached at a point where the line crosses the zero base line and is referred to as the XPT. Uludag *et al.* (2001) indicated the three stages in a study and described that Stage I starts with minimal differential and progressively increases towards the XPT where the differential is zero. Stage II continues from the XPT to the point referred to as the kick–point. In addition, Stage II is one of the best indicators of spontaneous combustion liability. Stage III is when the coal starts to burn beyond the kick-point. The index makes use of the fact that coal highly prone to self-heating have a steeper Stage II slope and a lower XPT than coal not highly prone to self-heating. According to Wade *et al.* (1987), coal with a spontaneous combustion liability index below 3 are

considered to be low risk, 3 to 5 medium risk, and coal with values greater than 5 are considered to be highly liable to spontaneous combustion. The index is calculated from the formula in Equation 3.7. The thermogram in Figure 3.19 illustrates the stages and the XPT for a given coal sample.

Wits-Ehac Index = (Stage II slope/XPT) * 500 (3.7)

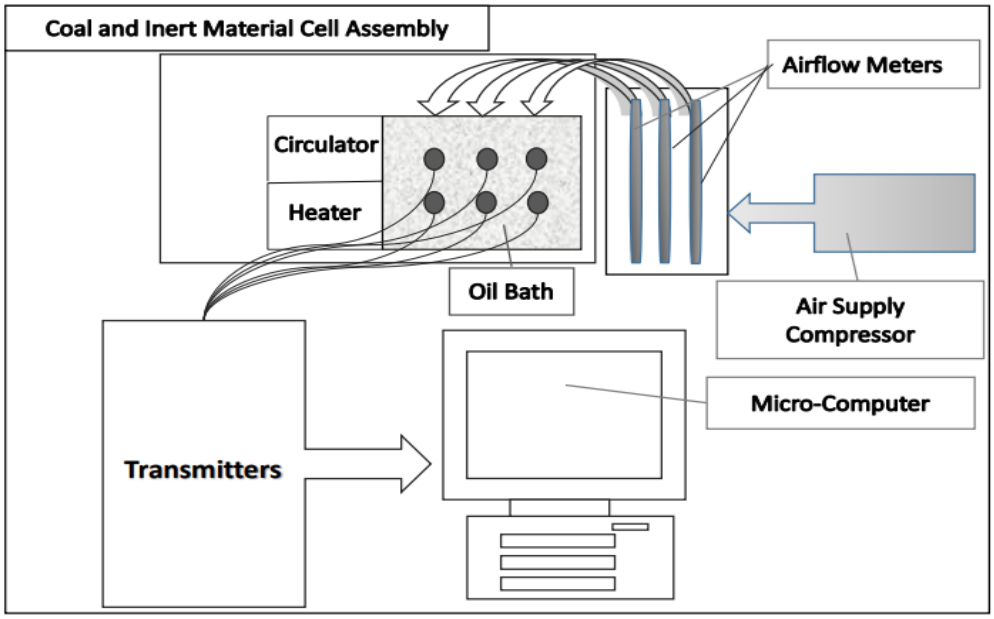

Figure 3.18 Schematic of the Wits-Ehac apparatus setup (Wade et al., 1987)

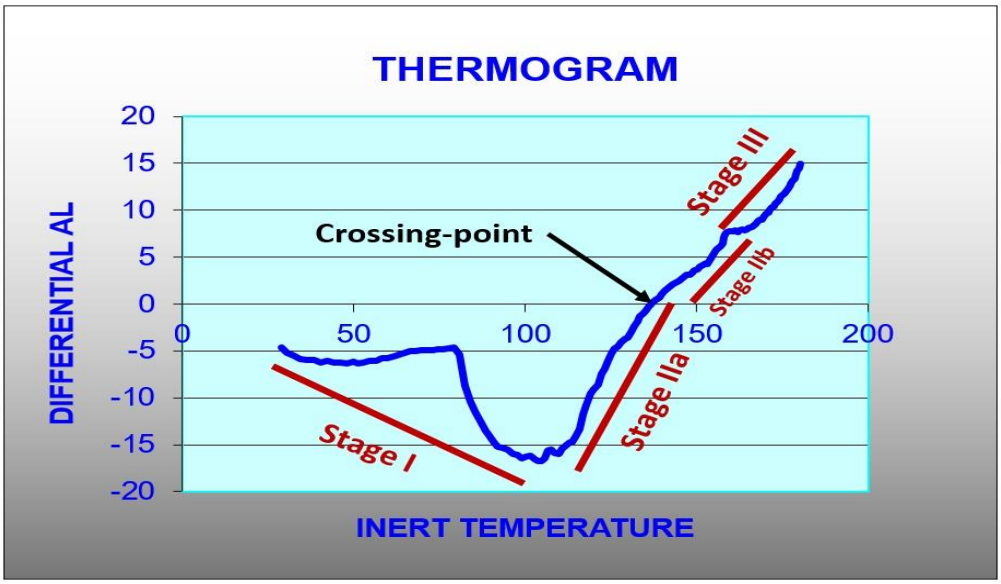

Figure 3.19 A typical differential analysis thermogram of a coal sample (Gouws and Eroglu, (1993), Wade *et al.*, (1987))

3.6 New apparatus (Wits-CT)

3.6.1 Wits-CT experimental setup

The Wits-Ehac Index was developed in South Africa to test the spontaneous combustion liability of coal. The Wits-Ehac Index failed to produce results when testing some coal-shale samples. To overcome this problem, a new apparatus was developed to test coal and coal-shale under chemical reactions with oxygen and a new index referred to as the Wits-CT Index was developed. This equipment was used to test the spontaneous combustion liability of South African coal and coal-shale samples. Most coal undergo spontaneous combustion under atmospheric conditions, i.e. when exposed to oxygen in the air. The equipment developed is based on this fact and it emulates the influence of oxygen adsorption on coal and coal-shale in a closed system. Therefore, the equipment does not involve heating of samples to a high temperature such as the adiabatic method (the R_{70}, Wits-Ehac test and others). The equipment prevents heat loss that may occur under testing conditions by ensuring the lid is fixed and fastened, and also a glass mineral wool/ceramic fibre blanket (thermal insulator type material) is placed inside the equipment in order to prevent any heat loss. The insulating material is selected due to the temperature that may be generated during testing and the heat transfer that may be involved. However, as no heating system is applied to the equipment to increase the temperature of the sample, the equipment is not expected to generate a high temperature. The equipment determines the temperature variation within coal particles with six temperature sensors under chemical reactions with oxygen for 24hrs. This scenario emulates the reaction of oxygen and coal leading to self-heating in mines. The influence of oxygen absorption on the sample leading to the temperature variations during the start and the end of the test is recorded by the computer via a data logger as illustrated in Figure 3.20 and Figure 3.21. The temperatures recorded and the total carbon content of the sample are used to obtain a spontaneous combustion liability index.

The equipment has a capacity to accept a 15kg sample, depending on the packing density. It oxidizes materials at a controlled air pressure and airflow. Three uniformly separated temperature sensors placed along the length of the equipment are used to keep record of the temperature measurements. The sensors are located at different points in the equipment (locations A, B, and C within the interior of the container). Each of the three thermocouples has two temperature sensors which measure temperature at different levels (i.e. locations A, B, and C in order to keep a continuous record of temperature distribution during testing). At

location A, sensors are located at levels of 13cm and 23cm within the equipment; location B, 53cm and 63cm, and location C, 33cm and 43cm. The equipment comprises of an upright mounted cylindrical tube of 0.75m long and with an internal diameter of 160mm. The cylinder wall thickness is made of 5mm thick mild steel. Oxygen is supplied and controlled at a constant flowrate (flowmeter) before being fed into a manifold attached below the lid of the equipment. The connection between the manifold and the equipment is achieved by means of a spiral copper tube. Oxygen gas is supplied from the lid through the manifold. The equipment is placed on a digitalize scale (Richterscale, Model W113) to measure any changes in weight during testing due to oxygen absorption. The experimental process emulates a well-insulated system relative to actual *in-situ* conditions. Sensogut & Ozdeniz (2005) and Ozdeniz *et al.* (2015) used a similar approach in a coal stockpile (using thermocouples and a data logger to keep record of temperature measurements under ambient temperatures and pressure).

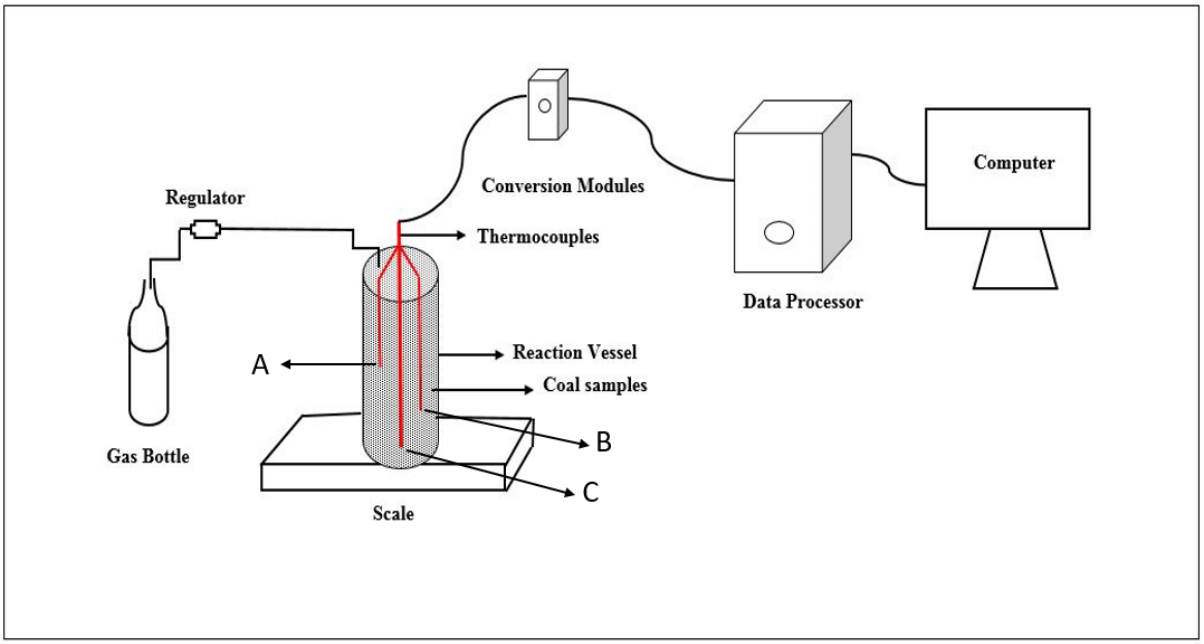

Figure 3.20 Schematic view of experimental setup

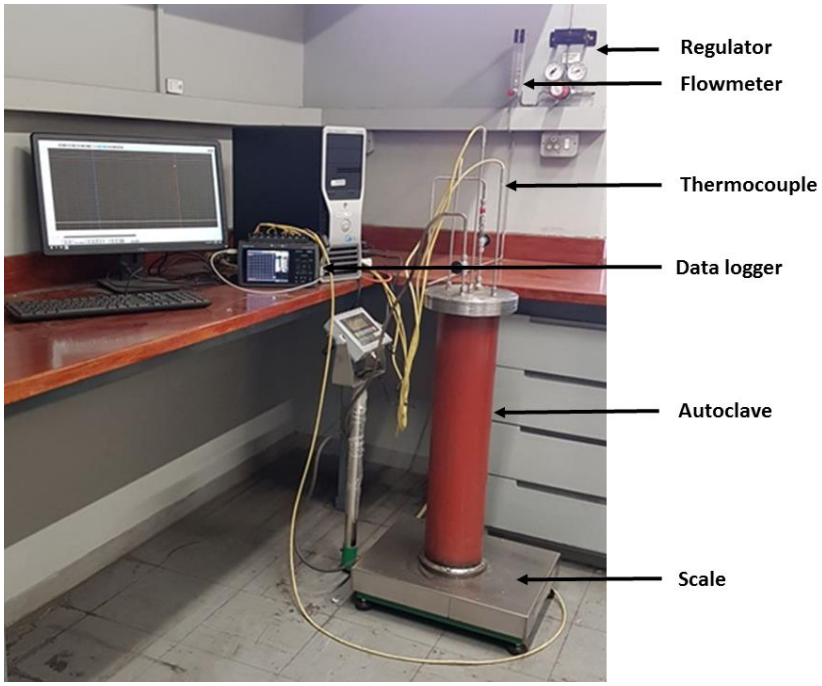

Figure 3.21 New apparatus setup

3.6.2 Wits-CT testing procedure

Coal samples of particle size (<6.39mm) is weighed and loaded in the equipment in order to determine the spontaneous combustion liability via self-heating. The particle size used is based on the study reported by Kunii & Levenspiel (1991) to determine the surface-volume area from the size distribution. The system is sealed at the base, placed on a scale and loaded from the top with the sample. The lid of the equipment is detached during the loading of the samples; the process is repeated until the equipment is filled to the marked point. The lid is fixed and fastened as soon as the equipment is full. Each temperature probe is inserted into the equipment as the correct sample level is reached. The position of each temperature probe inside the equipment is placed vertically and spaced uniformly to the center as soon as the sample level is reached. Oxygen is supplied and controlled at a fixed flow rate (20mL/min) by means of a flowmeter before being fed into a manifold attached below the lid of the equipment. The changes in temperature distribution are recorded by the data logger every minute for 24hrs. At the end of each test, samples are discarded and the equipment is cleaned. Twenty-eight (28) tests were conducted with this equipment to obtain a better definition for the Wits-Ehac Index of coal-shale samples that could not be determined.

3.7 Statistical Analysis

Linear (R-squared values and correlation coefficients) and multiple regression analysis were carried out using MS Excel to find out the best correlated parameters. Intrinsic properties and the spontaneous combustion liability index were considered as independent and dependent variables respectively. Linear regression analysis between intrinsic properties, i.e. parameters of proximate analysis (moisture, volatile matter and ash), elements of ultimate analysis (carbon, hydrogen, nitrogen and total sulphur), macerals of petrographic composition (total vitrinite, total liptinite and total inertinite), and spontaneous combustion liability indices (Wits-Ehac Index and Wits-CT Index) was carried out. Multiple regression was carried out on the combined parameters of the intrinsic properties and each spontaneous combustion liability index.

3.7.1 How to compute correlation coefficient in MS Excel

- Type the data into columns in MS Excel;
- Select any empty cell and check the function button on the ribbon;
- Type correlation into the search for a function box, click "GO'', CORREL will be highlighted and click "OK'' as shown in Appendix 9.2; and
- Type the location of your data into the "Array 1" and "Array 2" and click "OK'', the result will appear in the cell you selected in step 1 as shown in Appendix 9.2.

3.7.2 Procedure for regression data analysis tool in MS Excel

- Click the data analysis command button on the Data tab of the MS Excel; Select the regression tool from the analysis tool list and click OK;
- Select the Y-range. This is the predictor variable i.e. the dependent variable (the spontaneous combustion liability index); Select the X-range. These are the descriptive variables i.e. the dependent variables (intrinsic properties). These columns must be adjacent to each other;
- Check the labels; check in the Output Range box and select cell; check residuals; and click OK. The summary of the regression output will appear where designated.

MS Excel indicates a portion of the regression analysis results including three, stacked visual plots of data from the regression analysis. There is a range that provides some basic regression statistics, this includes the R-squared value, the standard error, and the number of observations.

The Regression tool also supplies analysis of variance (or ANOVA) data, such as information about the degrees of freedom, sum-of-squares value, mean square value, the F-value, and the significance of F. Aside the ANOVA information, the Regression tool supplies information about the regression line calculated from the data, including the coefficient, standard error, t-stat, and probability values for the intercept as well as the same information for the independent variables, which is the number of ads. The screen shots of section 3.7.1 and section 3.7.2 are illustrated in Appendix 9.2.

3.8 Inhibitor tests

Three coal samples and one coal-shale sample with high spontaneous combustion liability indices among the studied samples with three recently obtained coal samples reported from KCA, KCC and KCD Mines, South Africa with very high spontaneous combustion liability indices were added to inorganic salt (chemical inhibitors). Calcium carbonate ($CaCO_3$), magnesium chloride ($MgCl_2$), ammonium dihydrogen phosphate ($NH_4H_2PO_4$) and sodium metasilicate pentahydrate ($Na_2SiO_3.5H_2O$) were selected and investigated as the chemical inhibitors based on an assessment of the most suitable chemical inhibitors. The choice of selecting these chemical inhibitors was to prove the concept that inhibitors can minimise spontaneous combustion. All the selected chemical inhibitors are analytically pure. Each inhibitor was blended with the selected raw coal and coal-shale samples in a beaker and stirred mechanically to produce samples containing 20 wt.% additives (Chen *et al.,* (2016), Cheng *et al.,* (2016) and Wang *et al.,* (2014)). The particle size (-212µm) of the samples is the same as that used for untreated coal and coal-shale. All samples were sealed from air and stored in a desiccator prior to measurements. Subsequently, each component of the formulation was investigated to determine its effect on spontaneous combustion liability. The spontaneous combustion liability index of the treated coal and coal-shale were determined with the Wits-Ehac apparatus following the same procedure described in Section 3.5. Twenty-eight (28) tests were conducted on the treated samples.

3.9 Summary

This section summarises the experimental techniques used for fourteen (14) bituminous coal and 14 coal-shale samples. This section involves two major tests; geochemical and spontaneous combustion tests (the Wits-Ehac and the Wits-CT tests). The combination of these tests provides a new methodology to the understanding of the spontaneous combustion liability of

coal and coal-shale phenomena. The testing procedures used for each experiment were described. The results of the experimental tests conducted on the samples to evaluate the mechanism that control their liability toward spontaneous combustion are discussed in the next Chapter.

4 SPONTANEOUS COMBUSTION AND GEOCHEMICAL ANALYSIS

4.1 Introduction

This Chapter presents and discusses the results of the spontaneous combustion liability and geochemical tests for coal and coal-shale and their relationships with the spontaneous combustion liability index. The possible factors related to the cause of the spontaneous combustion were investigated from the geochemical tests carried out in Chapter 3. A detailed report on the equations, calculations and graphical representation of the XPT, Stage II slope, Wits-Ehac Index are discussed. Furthermore, a detailed report on the application of the Wits-CT apparatus to predict the spontaneous combustion liability of coal and coal-shale is discussed. The combination of these factors has provided a new approach to the understanding of the spontaneous combustion liability of different coal and coal-shale which will enable coal miners to have a better understanding of the principal factors contributing to the risk of spontaneous combustion in coal mines. Furthermore, it provides a new knowledge to measure the possibility of a material to undergo thermal runaway.

4.2 Spontaneous combustion liability testing

The relative propensity of various coal to undergo self-heating can be established in the laboratory by different methods. These methods are well established in their usage but the fact that no particular test method has become a standard indicates that doubt still exists as to the validity of all of them. The underlying principle of all the tests is that the more readily the coal undergoes exothermic oxidation, the more liable it is to self-heat.

4.2.1 Equations and calculations

Spontaneous combustion is a first order of oxidation reaction where heat loss can be assumed to be only due to conduction. The reaction is temperature dependant and can be expressed as follows (Banerjee, (1985), Gouws & Eroglu, (1993) and Wade *et al.*, (1987)):

$$wC_P \frac{dT}{dt} = WQZe^{-\frac{E}{RT}} \tag{4.1}$$

where, **w** is the density (Kg/m^3), **Cp** is the specific heat capacity of coal at constant pressure (J/Kg/K), **T** is the temperature (K), **t** is time (minutes), **Q** is the heat of reaction (J/Kg), **e** is the exponential (natural log), **E** is the activation energy (kJ/mol) and **R** is the molar gas constant (J/mol.K).

The expression in Equation 4.1 is referred to as the Arrhenius Equation and under adiabatic conditions where the conductivity term is negligible, it can be simplified as follows (Gouws & Eroglu, (1993) and Wade *et al.*, (1987)):

$$C_P \frac{dT}{dt} = QZe^{-\frac{E}{RT}} \tag{4.2}$$

or

$$\frac{dT}{dt} = Ae^{-\frac{E}{RT}} \tag{4.3}$$

where, $A = \frac{Q_z}{C_p}$ (4.4)

A is the rate coefficient

This can be further simplified using Equation 4.4 as follows:

$$In\frac{dT}{dt} = -\frac{E}{RT} + In\ A \tag{4.5}$$

which is of the form:

$$y = mx + c \tag{4.6}$$

where **x, y** are the coordinates of any point on the line x and y axes respectively, **m** is the slope and **c** is the intercept (where the line crosses the y axis).

The log of the rate of temperature rise ($In\frac{dT}{dt}$) can be plotted against the inverse of the absolute temperature of the coal ($1000/_T$). This plot is called the Arrhenius plot and is shown in Figure 4.1 (Gouws & Eroglu, (1993) and Wade *et al.*, (1987)). The slope of the straight line during the latter part of the experiment is equal to $\frac{E}{RT}$. The activation energy (E) of the coal can be calculated from this slope. The vertical intercept is the rate coefficient (also known as the pre-exponential coefficient or frequency factor) for the reaction, which is believed to be greater for coal more likely to self-heat (Banerjee, 1985, Gouws & Eroglu, (1993) and Wade *et al.*, (1987)).

Besides the characteristics already mentioned, another characteristic of the coal curve is immediately apparent (Gouws & Eroglu, (1993) and Wade *et al.*, (1987)). This is the temperature of the coal at which the coal curve cuts the $Log\ \frac{dT}{dt}$ = 0 line.

In order to investigate the characteristics of the Arrhenius plot, the DTA thermogram (from the ignition temperature tests) can be modified to show the inverse absolute temperature of the coal on the horizontal axis. The modified DTA thermogram for a sample is shown in Figure 4.2 (Gouws & Eroglu, (1993) and Wade *et al.,* (1987)).

After comparing Figure 4.1 and Figure 4.2, the point at which the coal curve cuts the $In\frac{dT}{dt}$ =0 line is the XPT for the coal (Gouws & Eroglu, (1993) and Wade *et al.,* (1987)).

After further comparing Figure 4.1 and Figure 4.2, relationships between the slopes of the coal curve during the latter part of the experiment and the Stage II slope of the DTA, the thermogram was established (Gouws & Eroglu, (1993) and Wade *et al.,* (1987)). The slopes of the coal samples tested in the calorimeter were compared with the Stage II slopes of the same sample and a linear regression showed a correlation coefficient (R) of 94 % and a coefficient of determination (R^2) of 89 % (Gouws & Eroglu, (1993) and Wade *et al.,* (1987)).

Since the calculation of the Wits-Ehac Index value of a coal sample requires only the XPT and the Stage II slope, it can be calculated from the calorimeter data that has been shown to correlate well with the ignition temperature data. It has also been pointed out that the slope is equal to E/R (Gouws & Eroglu, (1993) and Wade *et al.,* (1987)). Hence, the index can therefore be rewritten as shown in Equation 4.7 (Gouws & Eroglu, (1993) and Wade *et al.,* (1987)):

$$Wits - Ehac = \frac{E/R}{XPT} * \ constant \tag{4.7}$$

By plotting the inverse of the XPT on the y-axis, the Stage II slope on the x-axis as shown in Figure 4.3 and joining the two points, a triangular area is enclosed. This area now takes into account the two parameters measured (Gouws & Eroglu, (1993) and Wade *et al.,* (1987)).

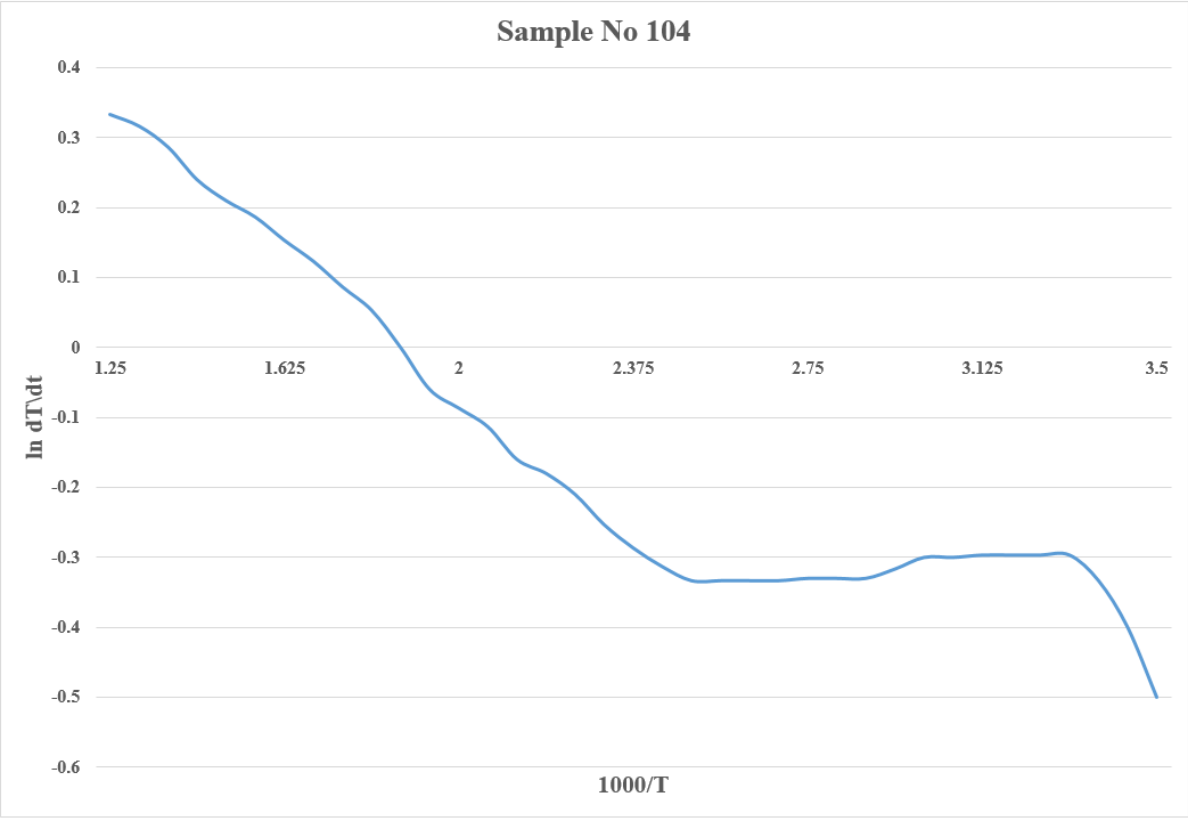

Figure 4.1 A typical Arrhenius plot (Gouws & Eroglu, (1993) and Wade *et al.*, (1987))

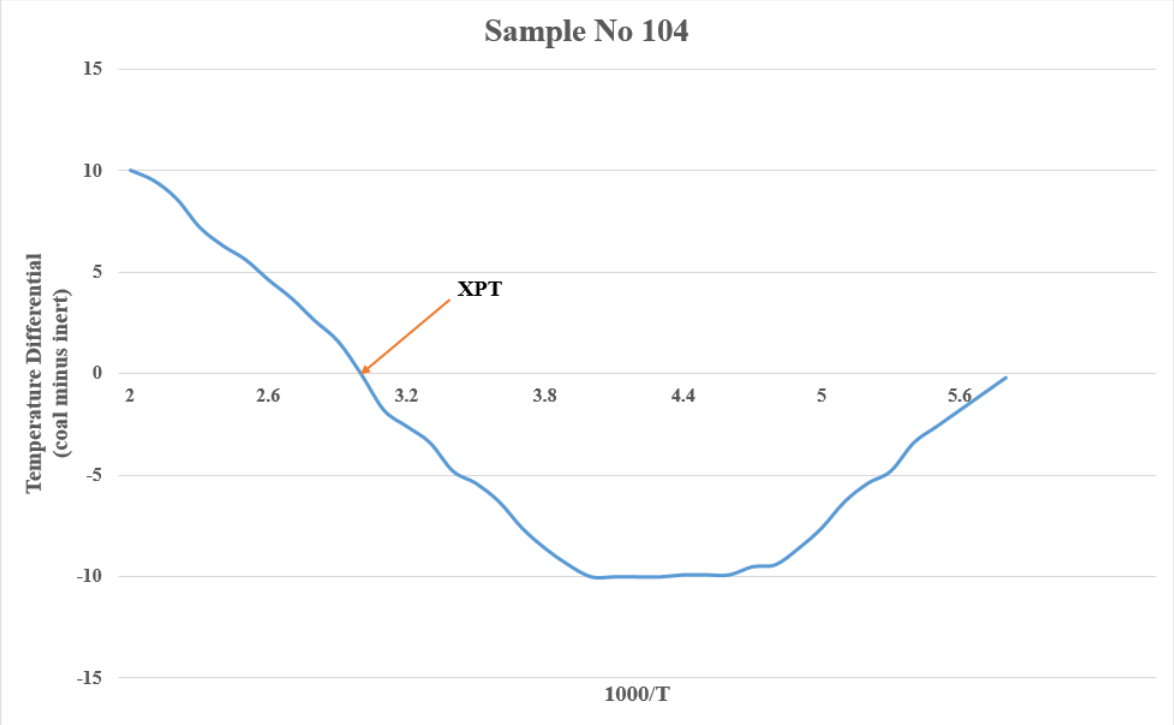

Figure 4.2 Modified DTA curve obtained using ignition temperature tests (Gouws & Eroglu, (1993) and Wade *et al.*, (1987))

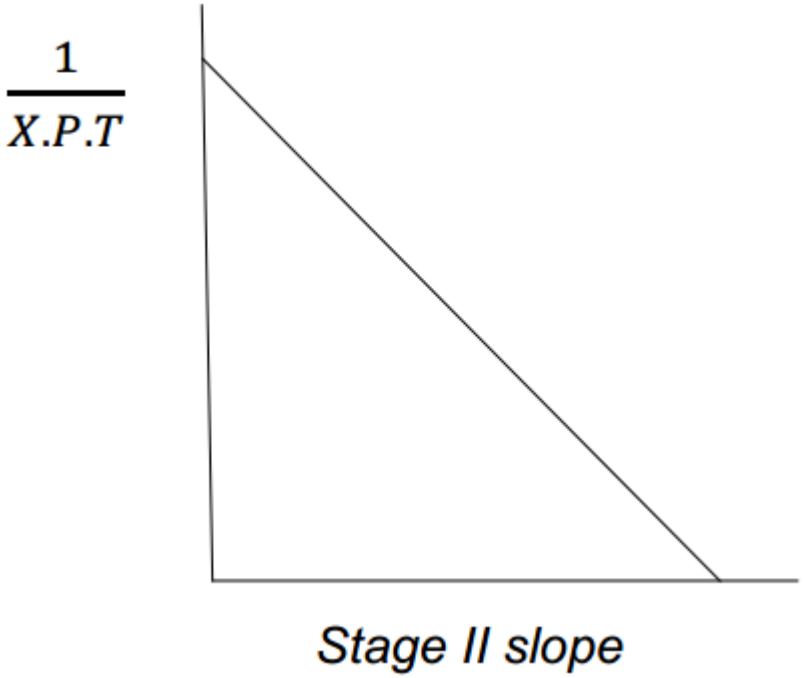

Figure 4.3 Triangle formed by joining the two characteristic values of a coal (Gouws & Eroglu, (1993) and Wade *et al.*, (1987))

Using simple geometry, this area and the Wits-Ehac are computed as follows:

$$Area = 0.5 * Stage\ II\ slope * \frac{1}{XPT} * 1000 \tag{4.8}$$

$$Wits - Ehac = \frac{Stage\ II\ slope}{XPT} * 500 \tag{4.9}$$

The determination of the XPT and Stage II slope from the Arrhenius Equation and under adiabatic conditions has been identified in this study as being a source for predicting the spontaneous combustion liability (Gouws & Eroglu, (1993) and Wade *et al.*, (1987)). Having established whether or not a given coal sample is relatively liable to spontaneous combustion, it becomes necessary to evaluate the degree of liability in the mining environment.

4.2.2 Prediction of spontaneous combustion propensity of coal and coal-shale using the Wits-Ehac Index

The Wits-Ehac Index value of a coal sample requires only the XPT and the Stage II slope as shown in the equations and calculations in Equation 4.9 (Gouws & Eroglu, (1993) and Wade *et al.*, (1987)). The index was proposed in 1987 by Wade and was based on characteristics of a given coal, as identified on a DTA thermogram as shown in Figure 3.19. The thermogram is

obtained by heating a coal sample and an inert material in identical sample holders in an oil bath as shown in Figure 3.18. The thermogram is obtained by plotting the temperature of the coal minus that of the inert material against the temperature of the inert material. The liability index proved to be successful, and was validated by an investigation of apparent anomalies (Eroglu, 1992, Gouw et al., 1989 and Wade, 1989) and a research project in which the liability of a seam to self-heat was contoured (Eroglu, 1992). As previously stated, the Wits-Ehac Index value of a coal sample requires only the XPT and the Stage II slope as shown in the equations and calculations in Equation 4.9 (Gouws & Eroglu, (1993) and Wade *et al.,* (1987)).

Table 4.1 XPT (^{o}C), Stage II slope and the Wits-Ehac Index results for the coal samples

Samples	**XPT**	**Stage II slope**	**WE**
CA	145.5	1.3492	4.64
CB	149.4	1.3841	4.64
CC	150	1.3879	4.52
CD	146.3	1.3448	4.60
CE	140.4	1.3622	4.76
CF	153.1	1.3737	4.49
CG	140.7	1.3815	4.91
CH	145.8	1.3927	4.69
CI	167.2	1.2763	3.82
CJ	146.7	1.3097	4.46
CK	146.9	1.3036	4.44
CL	148.6	1.4487	4.87
CM	145.4	1.3839	4.76
CN	139	1.3460	4.84

Table 4.2 XPT (°C), Stage II slope and the Wits-Ehac Index results for the coal-shale samples

Samples	XPT	Stage II slope	WE
SA	178.7	1.1050	3.09
SB	187.7	1.1471	3.06
SC	191.7	-	-
SD	186.4	1.1389	3.27
SE	164.1	1.2227	3.73
SF	186.6	1.1574	3.10
SG	188.7	-	-
SH	192.5	-	-
SI	184.1	-	-
SJ	194.1	-	-
SK	189	1.1268	2.98
SL	189.8	1.1337	2.99
SM	190.4	-	-
SN	161.8	1.2245	3.77

The results and the discussions in this section are based on experiments conducted on fourteen (14) coal and 14 coal-shale samples collected from four (4) selected coal mines in Witbank coalfield. For each sample, the XPT and Stage II slope were conducted simultaneously and the data was calculated as described in Section 4.2.1 to obtain their Wits-Ehac Indices. Simple liability indices are the characteristics obtained from the XPT and DTA experiments as illustrated in Figure 4.4 and Figure 4.5. The XPT had a range from 139°C to 167.2°C for the coal samples and a range of 161.8°C to 194.1°C for the coal-shale samples. The Stage II slope had a range from 1.2763 to 1.4487 for the coal samples and a range of 1.1050 to 1.2245 for the

coal-shale samples. The spontaneous combustion liabilities of the coal samples, according to the Wits-Ehac Index ranges from 3.82 to 4.91, while the coal-shale samples ranges from 2.98 to 3.77. Sample CN has the lowest XPT (139^{o}C), while sample CI has the highest XPT (167.2^{o}C) among the coal samples. Sample CL has the highest Stage II slope (1.4487), while sample CI has the lowest Stage II slope (1.2763). For the coal-shale samples, sample SN has the lowest XPT (161.8^{o}C) followed by sample SE (164.1^{o}C), while sample SJ has the highest XPT (194.1^{o}C). Sample SN has the highest Stage II slope (1.2245) followed by sample SE (1.2227), while the Stage II slopes of samples SC, SG, SH, SI, SJ and SM could not be determined due to their low reactivity. For the coal samples, Sample CG has the highest Wits-Ehac Index (4.91), while sample CI has the lowest Wits-Ehac Index (3.82). For the coal-shale samples, sample SN has the highest liability index (3.77), while the Wits-Ehac Index of samples SC, SG, SH, SI, SJ and SM could not be determined under the Wits-Ehac tests. Higher Wits-Ehac Index values represent the high risk of a coal to spontaneous combustion (Gouws & Eroglu, (1993), Gouws, (1987), Wade *et al.*, (1987)). Hence, the samples with high liability indices are more prone to undergo spontaneous combustion than those with lower liability indices.

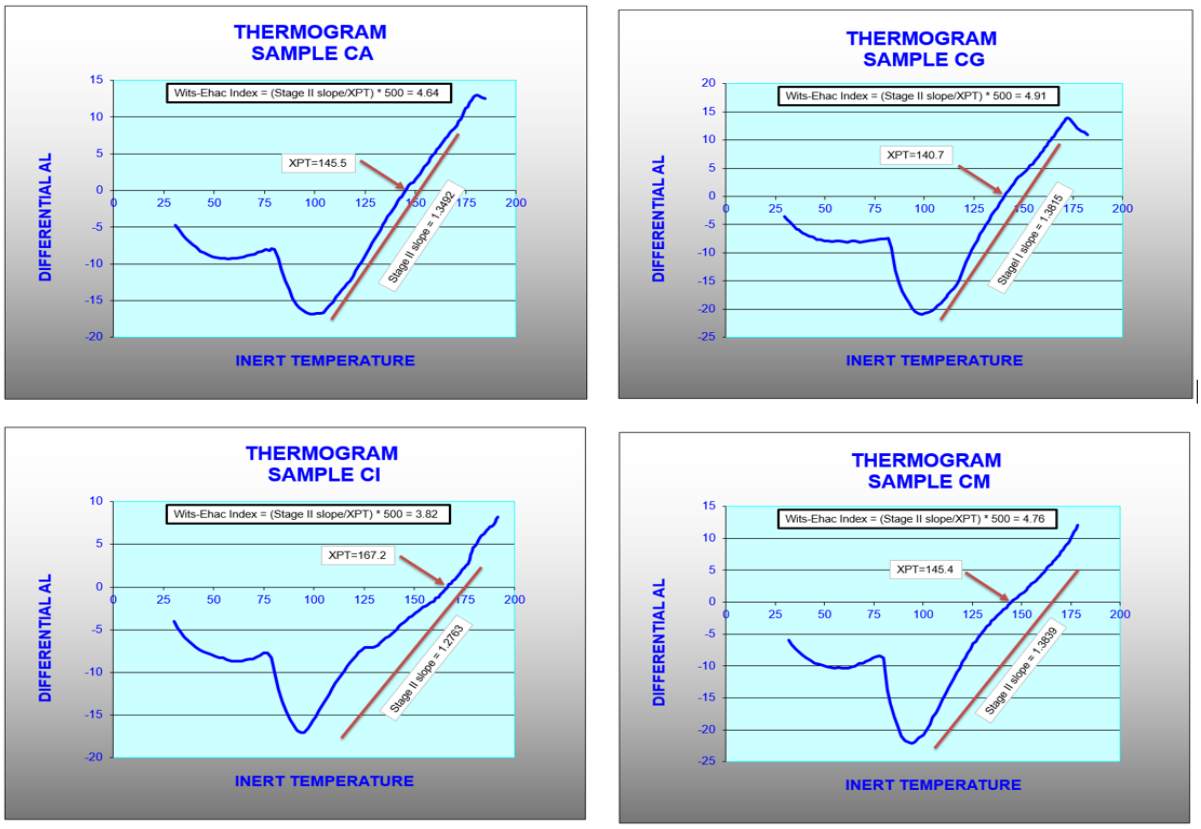

Figure 4.4 Differential analysis thermogram for untreated coal samples

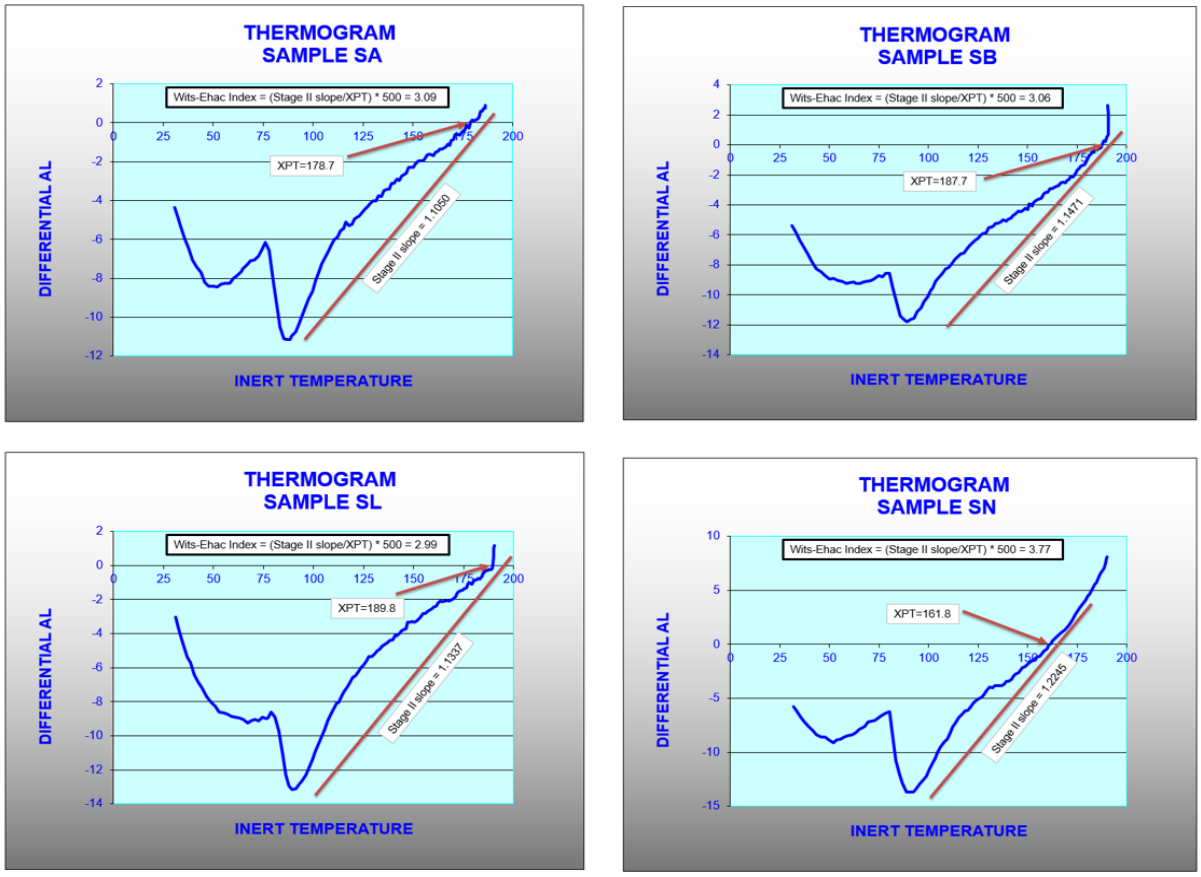

Figure 4.5 Differential analysis thermogram for untreated coal-shale samples

4.2.3 Prediction of the spontaneous combustion propensity of coal and coal-shale using a new apparatus (Wits-CT apparatus)

Coal oxidation occurs in an environment where there is sufficient oxygen. The oxidation rate of coal cannot be easily detected due to the reaction being very slow. It is known that coal, roof shales, spoil heaps and other carbonaceous materials become self-heated and liberate heat naturally when subjected to oxygen in the air for a long time. The experience of spontaneous combustion has approached into a phase that makes practical illustrations feasible as a result of the re-established awareness of self-heating of coal and coal-shale in mines. The spontaneous combustion liability of some coal-shale in this study could not be determined by the Wits-Ehac Index because of their low reactivity under the available experimental conditions. This necessitated the need to develop a new device that measures the spontaneous combustion liability of coal, coal-shale and other carbonaceous materials under chemical reaction with oxygen. The use of realistic medium experimental scale tests which emulates the influence of oxygen in the air to predict the spontaneous combustion liability of different coal, coal-shale and other carbonaceous material is important. The principal argument against

laboratory testing to predict the spontaneous combustion liability of coal is that the process is unlike the natural conditions under which self-heating takes place. On the basis of this criteria, a medium scale laboratory apparatus was developed to predict the spontaneous combustion liability of coal and coal-shale. The equipment considers a number of parameters such as the airflow rate and temperature variation. The need for a reliable method to capture the large volume of data used to predict the spontaneous combustion liability motivated the development of this new apparatus (Wits-CT apparatus). The method is reliable, accurate and provides the required information on the effects of oxygen on the spontaneous combustion of coal and coal-shale.

i. Wits-CT Index

The Wits-CT apparatus was developed to measure the spontaneous combustion liability of coal, coal-shale and other carbonaceous materials. The equipment monitors and records the temperature distributions within these materials with the use of thermocouples under chemical reactions with oxygen and a new spontaneous combustion liability index, referred to as the Wits-CT Index, was obtained. The testing process includes complex reactions between coal and coal-shale and oxygen, heat transfer and oxygen transmission inside the equipment. The oxygen adsorbed by each sample causes a slight increase in weight. Samples with a high Wits-CT Index have higher oxygen absorption than those with a lower Wits-CT Index. The temperature increase in each sample depends on the oxygen consumed. As this was based on tests carried out on only fourteen (14) coal and 14 coal-shale samples, to expand the database of the samples investigated, this new liability index was compared with the results of the Wits-Ehac Index. The results from the two liability indices are in-line with what is reported in the mines. The formula used for the calculation of the new liability index is shown in Equation 4.10:

$$\text{Wits-CT Index} = (T_M/24 + T_R) * \% C_{ad} \quad (4.10)$$

where, $\mathbf{T_M}$ is the difference between the sum of maximum temperatures of each thermocouple inside the equipment and the room temperature (22^oC), $\mathbf{T_R}$ is the difference between the maximum temperature and the initial temperature during the oxidation reaction in degree Celsius, $\mathbf{\%C_{ad}}$ is the air-dried percentage of the carbon content of the sample and **24** is the test duration (hrs) and is constant.

The Wits-CT Index is established based on the experience of spontaneous combustion observed in the mines and proved to be reliable in identifying coal prone to spontaneous combustion and coal-shale which is not determined by the Wits-Ehac Index. A new classification of the spontaneous combustion liability index for coal and coal-shale based on the experience in the mines is shown in Table 4.3. The Wits-CT Index uses the total carbon content and the temperature differences due to the reaction with oxygen to predict the spontaneous combustion liability index. Coal and coal-shale with a spontaneous combustion liability index below 2.5 are considered to be less reactive, 2.5 to 5 moderately reactive, 5 to 7.5 reactive, and those with values greater than 7.5 are considered to be highly liable to spontaneous combustion as shown in Table 4.3.

Table 4.3 Risk rating classification for coal, coal-shale and other carbonaceous materials to spontaneous combustion

Wits-CT Index	**Risk Rating**
< 2.5	Less Reactive
2.5-5	Moderately Reactive
5-7.5	Reactive
> 7.5	Highly Reactive

The spontaneous combustion liability of coal seams differs over a wide range. It will be significant to evaluate their degree of proneness in order to take reliable precautionary measures against the event of spontaneous combustion. Much research have been carried out on coal but limited research has been conducted on coal-shale. The need for proper evaluation of the spontaneous combustion liability of coal and coal-shale is important so that the mine management will be aware of their influences on the mine, will plan the production activities and maximize coal production within the incubation period. The risk rating classification shown in Table 4.3 provides mine operators with a better understanding of the spontaneous combustion liability of coal and coal-shale. The Wits-CT Index is suitable for rating the spontaneous combustion liability of a material like the Wits-Ehac Index. The fact that different coal and coal-shale have different spontaneous combustion liability indices makes it important to classify them according to their liability index. The Wits-CT risk classification is dependent on two principles:

1. Determination of the total carbon content of the material; and
2. Changes in temperature of the materials due to their reaction with oxygen.

With an improved knowledge and the accumulation of a large volume of data obtained during the spontaneous combustion tests, this study has established a liability index in-line with the experience of spontaneous combustion in the South African coal mines. Table 4.4 and Table 4.4 shows the results of the Wits-CT indices for both coal and coal-shale samples.

Table 4.4 Results of the Wits-CT Index for the coal samples

Samples	**Wits-CT Index**
CA	6.29
CB	6.96
CC	5.31
CD	6.80
CE	5.42
CF	3.97
CG	7.53
CH	7.51
CI	4.05
CJ	6.61
CK	9.10
CL	9.59
CM	7.29
CN	7.91

Table 4.5 Results of the Wits-CT Index for the coal-shale samples

Samples	Wits-CT Index
SA	1.33
SB	1.30
SC	0.91
SD	0.70
SE	1.60
SF	1.36
SG	0.67
SH	0.27
SI	0.95
SJ	0.42
SK	1.18
SL	1.34
SM	1.44
SN	3.99

The Wits-Ehac Index values obtained from the values of XPT and Stage II slope had a range from 3.82 to 4.91, while the coal-shale samples had a range from 2.98 to 3.77 as shown in Table 4.1 and 4.2. The investigation shows that samples with a higher Wits-Ehac Indices represent high risk to spontaneous combustion. Spontaneous combustion test results show that coal CN (4.84), CM (4.79), CL (4.87), CG (4.91) and CE (4.76) will undergo spontaneous combustion faster than the other coal samples due to their high liability indices. However, for the coal-shale, samples SN (3.77) and SE (3.73) with high liability indices will undergo spontaneous combustion than the other coal-shale samples with lower liability indices. The

Wits-CT Index values varies between 4.05 to 9.59 for the coal and 0.27 to 3.99 for the coal-shale respectively as shown in Table 4.4 and Table 4.5. Coal samples CL (9.59), CM (7.27), CN (7.91), CG (7.53) and CK (9.1) with higher liability indices will undergo spontaneous combustion faster than the coal samples with lower Wits-CT Indices. For the coal-shale, samples SN (3.99) and SE (1.66) with higher Wits-CT Indices will undergo spontaneous combustion faster than the coal-shale samples with lower Wits-CT Indices. The results obtained from the spontaneous combustion tests indicated that coal and coal-shale vary in spontaneous combustion liability indices between bands of coal seams. Hence, samples with higher liability indices will undergo spontaneous combustion faster.

The spontaneous combustion tests show that the coal and coal-shale samples with a high Wits-CT Index and Wits-Ehac Index are more liable to spontaneous combustion. The Wits-Ehac Index values for coal-shale samples SC, SG, SH, SI, SJ and SM could not be determined due to their low reactivity, while the Wits-CT Index successfully obtained the liability indices for the same samples. The coal-shale samples for which the liability index values could not be determined with the Wits-Ehac Index have very low liability indices with the Wits-CT Index. The results from the two liability indices were compared with respect to what is happening in the mines and proved that samples with higher spontaneous combustion liability indices are more prone to spontaneous combustion than those with lower liability indices. Most of the fire caused by spontaneous combustion in the affected areas in which samples were collected have occurred in the zones that have been identified as having high liability indices. The records of the mines revealed that an incident of spontaneous combustion happened in these coal seams. The investigation shows that coal and coal-shale varies in spontaneous combustion liability indices between bands of coal seams.

4.2.4 Relationship between weight gained and liability index of coal and coal-shale

Table 4.6 and Table 4.7 show that the weight gained due to oxygen absorption by each sample can be related directly to the spontaneous combustion liability.

Table 4.6 Results from the spontaneous combustion tests and weight gain for the coal samples

Samples	Wits-CT Index	Weight gained (g)
CA	6.29	32.9
CB	6.96	35.0
CC	5.31	32.1
CD	6.80	34.7
CE	5.42	32.6
CF	3.97	29.8
CG	7.53	38.7
CH	7.51	37.2
CI	4.05	30.2
CJ	6.61	32.7
CK	9.10	42.5
CL	9.59	45.2
CM	7.27	36.1
CN	7.91	40.7

Table 4.7 Results from the spontaneous combustion tests and weight gain for the coal-shale

Samples	Wits-CT Index	Weight gained (g)
SA	1.33	27.5
SB	1.30	25.7
SC	0.91	23.5
SD	0.70	23.7
SE	1.60	31.5
SF	1.36	30.3
SG	0.67	20.1
SH	0.27	15.0
SI	0.95	20.1
SJ	0.42	18.5
SK	1.18	28.1
SL	1.34	28.9
SM	1.44	30.5
SN	3.99	32.3

The weight gained by each sample was small and slightly increases (in grams) with the increasing Wits-CT Index as shown in Figure 4.6 and Figure 4.7. Coal and coal-shale with a high Wits-CT Index have a high oxygen absorption rate than those with a low Wits-CT Index. Studies reported by Mehta *et al.* (1952) indicated that coal with a high amount of oxygen adsorption are more liable to spontaneous combustion. Coal CL has the highest weight gained and the highest Wits-CT Index, while coal CF has the lowest weight gained and lowest Wits-CT Index. Coal-shale SN has the highest weight gained and the highest Wits-CT Index, while coal-shale SH has the lowest weight gained and the lowest Wits-CT Index. The study shows that the temperature variations within the samples depends on their characteristics to absorb oxygen. The purpose of the measured weight enables the determination of how much oxygen has been absorbed by each sample under experimental conditions and their relationship with the spontaneous combustion lability index. Findings show that the samples increase in weight after part of it absorbed oxygen. This may likely be the same when fresh coal is exposed to

oxygen in the air. The sharp fall in the coal-shale sample with the Wits-CT Index of 0.95 (sample SI) may be due to a varying number of microscopic organic and inorganic matter present in the sample.

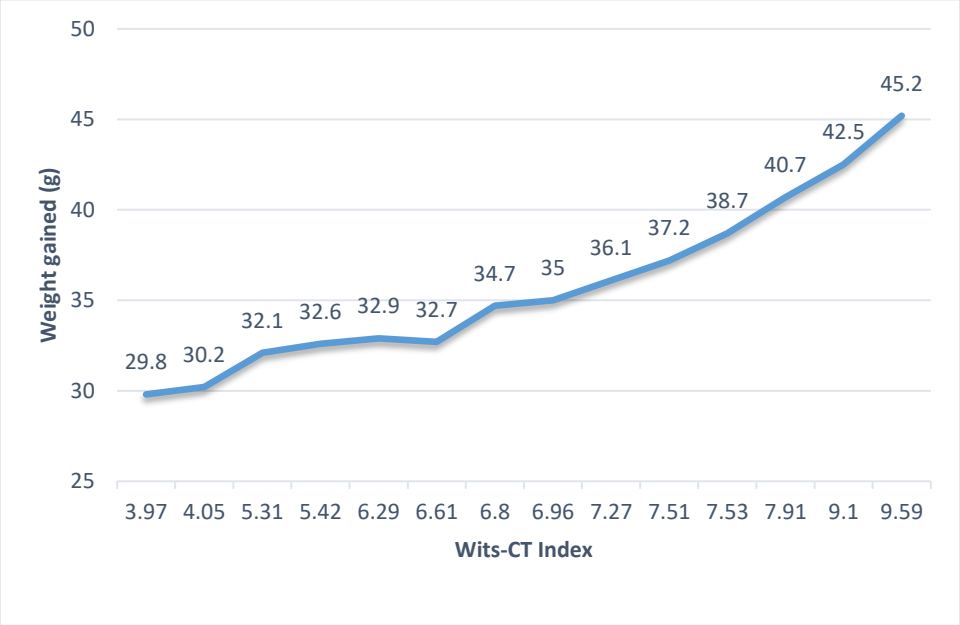

Figure 4.6 Relationship between weights gained and Wits-CT for the coal samples

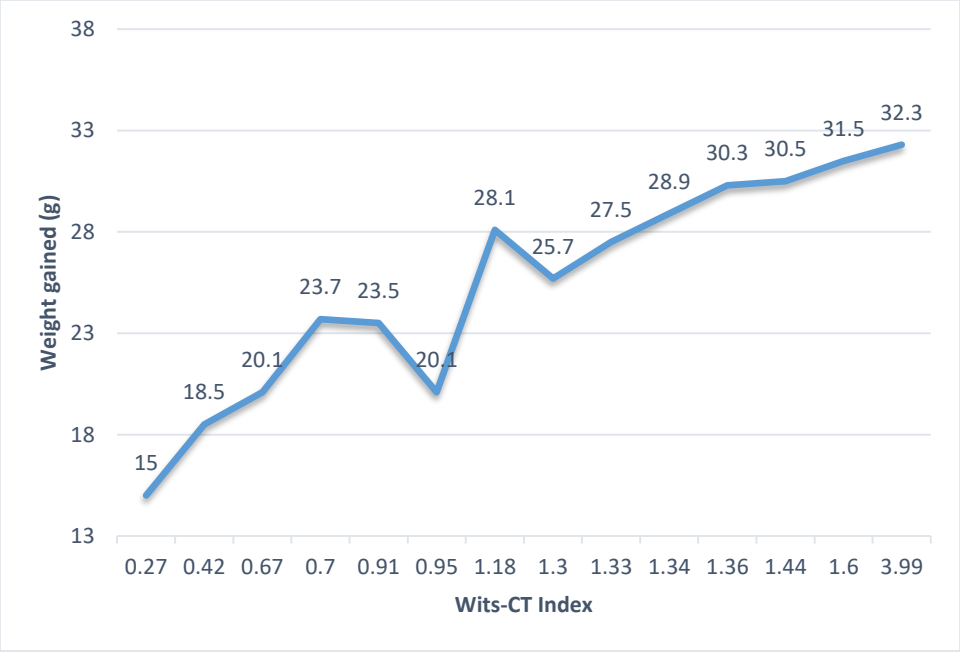

Figure 4.7 Relationship between weights gained and Wits- CT for the coal-shale samples

4.3 Characterisation results

The results for the geochemical data (proximate, elemental, petrographic, XRF and XRD analysis) and spontaneous combustion tests obtained from fourteen (14) bituminous coal and 14 coal-shale samples are presented in Tables 4.8 to 4.18 respectively. The coal samples are rich in carbon content and are more liable to spontaneous combustion based on the Wits-Ehac Index and the Wits-CT Index as shown in Table 4.8. Sample CK has the highest carbon content and the lowest ash content, while samples CF and CI have the lowest carbon content and the highest ash content as shown in Table 4.8. Sample CC also has a high ash content. Samples CF, CI and CC are rich in mineral matter as indicated by the high ash content as shown in Table 4.8 and high concentration of petrographically observable mineral matter as shown in Table 4.10. Sulphur occurs in all the samples, primarily as pyrite as shown in Table 4.8. Pyrite on exposure to the air and/or moisture will emit heat as a consequence of the exothermic reactions resulting from oxidation or heats of wetting. Sample CJ has the highest concentration of pyrite/ sulphur, while sample CM has the lowest concentration of pyrite/ sulphur as reported by all the analyses. Maceral group analysis shows a four-fold division, vitrinite, inertinite, liptinite and total observable mineral matter according to the ICCP (1998) and ICCP (2001). The coal samples show inertinite as the main constituent among the maceral groups as shown in Table 4.10 and Table 4.11. This is in-line with the studies reported by Falcon & Ham (1988), Hagelskamp & Snyman (1988), Holland *et al.* (1989), Snyman & Botha (1993) and Van Niekerk *et al.* (2008). On an mmf basis, the organic matter consists primarily of inertinite, dominated by inertodetrinite as shown in Table 4.11. All samples contained vitrinite and liptinite as well, with sample CF containing over 59 (vol.% mmf) vitrinite and samples CI, CL and CN containing over 4 (vol.% mmf) liptinite. Sample CF contained over 64 (vol.% mmf) of the total reactive maceral. The rank of the coal as determined by vitrinite reflectance places them in the Medium Rank C bituminous coal as shown in Table 4.12. The XRF data indicates that the samples are dominated by SiO_2 followed by Al_2O_3 as shown in Table 4.15 and the XRD data confirms the dominance of silicate minerals, specifically in the form of kaolinite followed by quartz as shown in Table 4.17.

The coal-shale samples are rich in mineral matter as indicated by the high ash content as shown in Table 4.9 and high concentration of petrographically observable mineral matter as shown in Table 4.14. All samples are hence classified as carbonaceous shale (ash above 50 wt%). Sample SN has the lowest ash content, highest carbon content and highest spontaneous combustion

liability index as shown in Table 4.9. Sulphur occurs in all the samples, primarily as pyrite as shown in Table 4.9. Sample SE has the highest concentration of pyrite / sulphur as reported by all the analyses. On an mmf basis, the organic matter consists primarily of inertinite, dominated by inertodetrinite as shown in Table 4.14. All samples contained vitrinite and liptinite as well, with sample SL containing over 39 (vol.% mmf) vitrinite and samples SD and SJ containing over 20 (vol.% mmf) liptinite. Sample SJ contains over 63 (vol.% mmf) of the total reactive maceral. The coal-shale occurred in association with medium Rank C bituminous coal. Reflectance analysis was not conducted on coal-shale samples due to their low vitrinite contents. The XRF data indicates that the samples are dominated by SiO_2 followed by Al_2O_3 as shown in Table 4.16. And the XRD data confirms the dominance of silicate minerals, specifically in the form of kaolinite followed by quartz as shown in Table 4.18. Coal-shale sample SN has the highest spontaneous combustion liability index (both for the Wits-Ehac Index and the Wits-CT Index), followed by coal-shale sample SE. It was found that samples SC, SG, SH, SI, SJ and SM displayed no liability indices for the Wits-Ehac Index. The relationships between the geochemical data and spontaneous combustion liability test results are discussed therein.

Table 4.8 Results from the proximate, ultimate, total sulphur, forms of sulphur analyses and spontaneous combustion test results for the coal

	Witbank Mines	**Proximate Analysis (ad)**				**Ultimate Analysis (ad)**					**Forms of sulphur (ad)**			**Liability Indices**	
Samples		**M**	**V**	**A**	**FC**	**C**	**H**	**N**	**S**	**O_c**	**P**	**SS**	**OS**	**WE**	**WC**
CA-4 seam coal	Mine 1	2.3	23.2	28.0	46.5	54.4	3.33	1.34	1.91	8.72	0.95	0.003	0.96	4.64	6.29
CB-2 seam coal		2.3	21.0	20.0	56.7	61.4	3.36	1.48	1.11	10.4	0.59	0.007	0.52	4.64	6.96
CC-2 seam coal		2.2	24.1	33.8	39.9	47.5	3.20	1.35	3.96	7.99	2.73	0.175	1.06	4.52	5.31
CD-2 seam coal	Mine 2	2.3	25.5	20.5	51.7	61.4	3.78	1.53	0.86	9.63	0.28	0.005	0.58	4.60	6.80
CE-4 seam coal		2.3	24.3	28.6	44.8	53.6	3.41	1.25	1.08	9.76	0.43	0.003	0.65	4.76	5.42
CF-2 seam coal		2.5	23.6	46.9	27.0	35.9	3.01	0.89	3.42	7.38	2.33	0.422	0.67	4.49	3.97
CG-2 seam coal	Mine 3	2.5	20.0	16.8	60.7	66.0	3.64	1.58	0.64	8.84	0.13	0.007	0.51	4.91	7.53
CH-5 seam coal		2.4	26.9	18.8	51.9	65.2	4.21	1.55	2.19	5.65	1.13	0.005	1.06	4.69	7.51
CI-4 seam coal		2.1	16.7	48.4	32.8	36.1	2.55	0.85	1.22	8.78	0.22	0.297	0.70	3.82	4.05
CJ-1 seam coal		1.9	25.7	28.1	44.3	52.4	3.13	1.35	5.30	7.82	4.13	0.084	1.09	4.46	6.61
CK-2 seam coal	Mine 4	1.6	22.1	13.7	62.6	69.7	4.02	1.60	0.76	8.62	0.36	0.009	0.40	4.44	9.10
CL-4 seam coal		1.6	26.1	22.5	49.8	58.9	3.57	1.45	3.88	8.10	3.22	0.008	0.66	4.87	9.59
CM-2 seam coal		1.6	22.0	17.0	59.4	66.7	3.77	1.57	0.59	8.78	0.30	0.006	0.28	4.76	7.27
CN-4 seam coal		1.6	23.9	17.0	57.5	65.8	4.20	1.63	2.92	6.85	2.30	0.005	0.62	4.84	7.91

where: **ad** is air dried, **M** is moisture (%), **V** is volatile matter (%), **A** is ash (%), **FC** is calculated fixed carbon (%), **C** is carbon (%), **H** is hydrogen (%), **N** is nitrogen (%), **S** is total sulphur (%), **P** is pyrite (%), **SS** is sulphide sulphur (%), **OS** is organic sulphur (%), **Oc** is calculated oxygen (%), **WE** is Wits-Ehac Index and **WC** is Wits-CT Index.

Table 4.9 Results from the proximate, ultimate, total sulphur, forms of sulphur analyses and spontaneous combustion test results for the coal-shale

	Witbank Mines	**Proximate Analysis (wt.%-ad)**				**Ultimate Analysis (wt.%-ad)**					**Forms of sulphur (wt.%-ad)**			**Liability indices**	
Samples		**M**	**V**	**A**	**FC**	**C_{ad}**	**H**	**N**	**S**	**O_c**	**P**	**SS**	**OS**	**WE**	**WC**
SA-4 seam shale	Mine 1	1.4	11.2	78.5	8.9	11.50	1.34	0.34	0.54	6.39	0.21	0.009	0.32	3.09	1.33
SB -2 seam shale		0.9	13.9	77.2	8.0	11.00	1.27	0.40	1.56	7.67	1.01	0.004	0.55	3.06	1.30
SC2- seam shale		1.1	12.7	74.6	11.6	13.80	1.60	0.42	0.35	8.13	0.15	0.005	0.20	-	0.91
SD -2 seam shale	Mine 2	1.6	13.3	77.3	7.8	10.80	1.43	0.32	2.53	6.02	1.43	0.343	0.76	3.27	0.70
SE -4 seam shale		1.7	15.9	68.4	14.0	15.80	1.78	0.41	6.90	5.01	4.26	0.450	2.19	3.73	1.60
SF-2 seam shale		0.9	13.5	76.9	8.7	11.80	1.40	0.43	0.46	8.11	0.19	0.009	0.26	3.10	1.36
SG -2 seam shale	Mine 3	0.8	10.7	84.3	4.2	6.02	1.04	0.29	0.73	6.83	0.22	0.007	0.50	-	0.67
SH -Upper shale		0.8	8.5	88.7	2.0	2.66	0.96	0.09	0.41	6.38	0.15	0.049	0.22	-	0.27
SI 2 -seam shale		1.0	11.9	79.6	7.5	9.12	1.41	0.26	0.22	8.39	0.05	0.005	0.17	-	0.95
SJ -Upper shale		0.9	11.9	86.9	0.3	3.42	0.75	0.08	0.75	7.19	0.43	0.004	0.32	-	0.42
SK -2 seam shale	Mine 4	1.0	11.7	79.1	8.2	9.75	1.73	0.41	0.16	7.85	0.10	0.005	0.05	2.98	1.18
SL -4 seam shale		1.0	16.0	74.0	9.0	10.50	2.14	0.39	0.12	11.85	0.04	0.003	0.08	2.99	1.34
SM -2 seam shale		0.8	11.7	76.9	10.6	12.50	1.61	0.52	0.24	7.43	0.16	0.015	0.06	-	1.44
SN -inseam shale		1.5	16.6	51.5	30.4	33.70	2.87	0.96	0.31	9.16	0.12	0.006	0.19	3.77	3.99

Table 4.10 Macerals analysis (vol.%) for the coal

Maceral Groups	Macerals (vol.%)	Sample no:													
		CA	**CB**	**CC**	**CD**	**CE**	**CF**	**CG**	**CH**	**CI**	**CJ**	**CK**	**CL**	**CM**	**CN**
Vitrinite macerals	Telinite	0.0	0.4	0.2	0.2	1.2	0.0	0.0	0.4	0.6	0.6	0.2	0.4	0.0	0.6
	Collotelinite	11.0	1.6	14.0	15.1	10.8	10.9	1.6	39.0	3.6	10.0	3.6	9.3	2.6	13.9
	Collodetrinite	8.2	2.8	12.0	9.8	13.3	5.9	6.8	9.0	3.8	8.2	6.2	12.1	3.2	11.7
	Corpogelinite	0.0	0.2	0.2	0.8	0.2	0.4	0.0	0.4	0.0	0.2	0.0	0.0	0.0	0.4
	Gelinite	0.2	2.0	0.0	0.0	0.0	0.0	1.0	0.6	0.0	0.8	1.6	1.2	1.2	1.0
	Pseudovitrinite	0.0	0.0	0.0	0.0	0.0	1.8	0.0	0.0	0.0	0.0	0.2	0.0	0.0	0.2
Total vitrinite		19.4	7.0	26.5	26.1	25.5	19.0	9.4	49.4	8.0	20.2	11.9	22.9	7.0	27.8
Inertinite macerals	Fusinite	2.0	2.2	7.2	6.4	7.6	1.4	5.6	6.6	0.8	4.2	3.8	4.2	7.2	5.8
	Reactive semifusinite	6.4	6.6	0.6	7.6	4.2	0.4	2.0	0.8	0.2	1.6	2.4	1.2	1.2	1.4
	Inert semifusinite	6.0	15.6	10.0	24.9	21.7	5.1	41.4	11.6	20.4	9.2	21.5	15.5	27.1	19.1
	Micrinite	0.6	0.4	0.0	0.8	0.0	0.0	1.6	0.0	0.0	0.8	0.2	0.8	1.2	0.8
	Macrinite	0.0	0.0	0.0	0.2	0.4	0.0	0.2	0.0	0.4	0.4	0.2	0.0	0.0	0.2
	Secretinite	1.8	6.0	1.4	4.6	2.2	1.6	3.0	1.2	1.4	3.0	2.8	0.8	5.2	1.4
	Inertodetrinite Reactive	4.6	4.6	0.8	0.8	0.6	0.0	0.4	0.0	0.0	0.6	0.6	0.4	0.8	0.8
	Inertodetrinite Inert	38.0	44.8	12.6	17.5	18.7	3.2	30.4	15.4	34.6	35.7	47.1	24.9	36.9	30.0
Total inertinite		59.4	80.2	32.7	62.9	55.4	11.7	84.6	35.6	57.8	55.5	78.7	47.9	79.7	59.6
Liptinite macerals	Sporinite	1.8	0.6	0.4	1.8	2.6	0.6	0.6	1.8	1.8	1.2	2.0	2.8	2.4	3.4
	Cutinite	0.2	0.2	0.2	0.4	0.2	0.4	0.0	0.0	1.2	0.0	0.0	0.2	0.0	0.4
	Alginite	0.6	0.8	0.0	0.0	0.0	0.0	0.0	1.0	0.0	0.0	0.0	0.0	0.0	0.0
Total liptinite		2.6	1.8	0.6	2.2	2.8	1.0	0.6	2.8	3.0	1.2	2.0	3.0	2.4	3.8
Total reactive macerals		33.0	20.0	28.5	36.7	33.1	20.4	12.4	53.0	11.2	23.6	16.9	27.6	11.4	33.8
Total mineral matter		18.6	11.0	40.3	8.8	16.3	68.4	5.4	12.2	31.2	23.0	7.4	26.2	10.8	8.9
Wits-Ehac Index		4.64	4.64	4.52	4.6	4.76	4.49	4.91	4.69	3.82	4.46	4.44	4.87	4.76	4.84
Wits-CT Index		6.29	6.96	5.31	6.8	5.42	3.97	7.53	7.51	4.05	6.61	9.1	9.59	7.27	7.91

Reactive maceral =total vitrinite + total liptinite + reactive semifusinite + reactive inertodetrinite

Table 4.11 Macerals analysis (vol.%, mmf) for the coal, (mmf=mineral matter free basis)

MACERAL Group	MACERAL (vol%)	Sample no:													
		CA	**CB**	**CC**	**CD**	**CE**	**CF**	**CG**	**CH**	**CI**	**CJ**	**CK**	**CL**	**CM**	**CN**
Vitrinite macerals	Telinite	0.0	0.4	0.3	0.2	1.4	0.0	0.0	0.5	0.9	0.8	0.2	0.5	0.0	0.7
	Collotelinite	13.5	1.8	23.5	16.5	12.9	34.4	1.7	44.4	5.2	13.0	3.9	12.5	2.9	15.2
	Collodetrinite	10.1	3.1	20.1	10.8	15.8	18.8	7.2	10.3	5.5	10.7	6.7	16.3	3.6	12.8
	Corpogelinite	0.0	0.2	0.3	0.9	0.2	1.3	0.0	0.5	0.0	0.3	0.0	0.0	0.0	0.4
	Gelinite	0.2	2.2	0.0	0.0	0.0	0.0	1.1	0.7	0.0	1.0	1.7	1.6	1.4	1.1
	Pseudovitrinite	0.0	0.0	0.0	0.0	0.0	5.6	0.0	0.0	0.0	0.0	0.2	0.0	0.0	0.2
Total vitrinite		23.8	7.9	44.3	28.6	30.5	60.0	9.9	56.3	11.6	26.3	12.8	31.1	7.9	30.5
Inertinite macerals	Fusinite	2.5	2.5	12.1	7.0	9.1	4.4	5.9	7.5	1.2	5.5	4.1	5.7	8.1	6.4
	Reactive semifusinite	7.9	7.4	1.0	8.4	5.0	1.3	2.1	0.9	0.3	2.1	2.6	1.6	1.4	1.5
	Inert semifusinite	7.4	17.5	16.8	27.3	25.9	16.3	43.8	13.2	29.7	12.0	23.3	21.0	30.4	21.0
	Micrinite	0.7	0.4	0.0	0.9	0.0	0.0	1.7	0.0	0.0	1.0	0.2	1.1	1.4	0.9
	Macrinite	0.0	0.0	0.0	0.2	0.5	0.0	0.2	0.0	0.6	0.5	0.2	0.0	0.0	0.2
	Secretinite	2.2	6.7	2.3	5.1	2.6	5.0	3.2	1.4	2.0	3.9	3.0	1.1	5.9	1.5
	Inertodetrinite Reactive	5.7	5.2	1.3	0.9	0.7	0.0	0.4	0.0	0.0	0.8	0.7	0.5	0.9	0.9
	Inertodetrinite Inert	46.7	50.3	21.1	19.2	22.3	10.0	32.1	17.5	50.3	46.4	50.9	33.8	41.4	32.9
Total inertinite		73.0	90.1	54.7	68.9	66.2	36.9	89.4	40.5	84.0	72.1	85.0	64.9	89.4	65.3
Liptinite macerals	Sporinite	2.2	0.7	0.7	2.0	3.1	1.9	0.6	2.1	2.6	1.6	2.2	3.8	2.7	3.8
	Cutinite	0.2	0.4	0.3	0.4	0.2	1.3	0.0	0.0	1.8	0.0	0.0	0.3	0.0	0.0
	Alginite	0.7	0.9	0.0	0.0	0.0	0.0	0.0	1.1	0.0	0.0	0.0	0.0	0.0	0.4
Total liptinite		3.2	2.0	1.0	2.4	3.4	3.1	0.6	3.2	4.4	1.6	2.2	4.1	2.7	4.2
Total reactive maceral		40.5	22.5	47.7	40.3	39.6	64.4	13.1	60.4	16.3	30.7	18.3	37.3	12.8	37.1
Wits-Ehac Index		4.64	4.64	4.52	4.6	4.76	4.49	4.91	4.69	3.82	4.46	4.44	4.87	4.76	4.84
Wits-CT Index		6.29	6.96	5.31	6.8	5.42	3.97	7.53	7.51	4.05	6.61	9.1	9.59	7.27	7.91

Table 4.12 Vitrinite reflectance measurements for the coal

	Sample no:													
		CA	**CB**	**CC**	**CD**	**CE**	**CF**	**CG**	**CH**	**CI**	**CJ**	**CK**	**CL**	**CM**	**CN**
Vitrinite Reflectance (RoV%)	Random	0.7	0.8	0.8	0.7	0.7	0.6	0.8	0.8	0.8	0.8	0.8	0.8	0.8	0.8
	st. dev.	0.1	0.1	0.1	0.1	0.1	0.0	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
	Max.	1.0	1.0	0.9	0.9	0.9	0.7	1.0	0.9	1.0	0.9	1.0	1.0	0.9	0.9
	Min.	0.6	0.6	0.7	0.5	0.6	0.5	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6
	No. readings	102.0	49.0	100.0	100.0	102.0	101.0	35.0	100.0	43.0	100.0	71.0	100.0	26.0	100.0
	Rank Category	Med Rank C	Med Rank C	Med Rank C	Med Rank C	Med Rank C	Med Rank C	Med Rank C	Med Rank C	Med Rank C	Med Rank C	Med Rank C	Med Rank C	Med Rank C	Med Rank C

where Med Rank C is Medium Rank C bituminous coal

Table 4.13 Macerals analysis (vol.%) for the coal-shale

MACERAL Group	MACERAL	Sample no:													
		SA	SB	SC	SD	SE	SF	SG	SH	SI	SJ	SK	SL	SM	SN
Vitrinite macerals	Telinite	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.6	0.6
	Collotelinite	0.6	0.8	0.0	1.9	1.2	0.2	1.0	1.4	0.0	1.1	0.0	4.2	0.0	2.8
	Vitrodetrinite	0.0	0.8	0.0	0.0	0.0	0.0	0.2	0.0	0.0	0.0	0.0	0.2	0.0	0.0
	Collodetrinite	1.6	0.8	1.4	1.1	2.0	0.4	0.8	1.4	0.0	0.5	0.4	2.6	0.0	1.2
	Corpogelinite	0.8	0.0	0.0	0.0	0.0	0.0	0.4	0.7	0.0	0.5	0.0	0.0	0.2	0.2
	Gelinite	0.2	0.3	0.0	0.3	0.0	0.0	0.4	0.0	0.4	0.3	0.4	0.0	0.0	0.2
	Pseudovitrinite	0.0	0.3	0.0	0.0	0.0	0.0	0.4	0.0	0.0	0.0	0.0	1.4	0.0	0.0
Total vitrinite		3.2	2.9	1.4	3.3	3.2	0.6	3.2	3.6	0.4	2.5	0.8	8.4	0.8	5.0
Inertinite macerals	Fusinite	1.2	4.0	1.6	1.9	0.5	4.0	1.8	0.0	1.8	0.5	1.0	1.4	3.2	2.2
	Reactive semifusinite	0.0	1.1	0.2	0.3	0.0	0.2	1.0	0.0	0.0	0.0	0.0	0.4	0.2	0.2
	Inert semifusinite	3.4	4.0	4.6	1.4	4.5	1.2	2.4	8.0	2.4	1.1	0.2	1.6	1.8	6.6
	Secretinite	0.0	1.6	0.8	0.6	1.2	0.8	0.8	0.7	0.8	0.0	0.0	0.8	1.4	1.8
	Inertodetrinite Reactive	1.6	0.8	0.4	0.3	2.5	1.0	1.8	3.6	0.0	1.4	0.6	0.6	1.0	0.4
	Inertodetrinite Inert	5.7	17.4	17.9	8.8	17.4	13.5	7.3	2.9	10.2	1.9	11.8	4.8	21.3	34.8
Total inertinite		12.0	28.9	25.6	13.2	26.1	20.8	15.2	15.2	15.3	4.9	13.7	9.6	28.9	46.1
Liptinite macerals	Sporinite	0.4	4.5	4.4	4.4	5.5	2.0	3.4	0.7	1.2	2.5	1.0	3.4	1.8	2.0
	Cutinite	0.0	0.0	0.0	0.0	0.0	0.0	0.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Resinite	0.0	0.0	0.0	0.0	0.0	0.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Total liptinite		0.4	4.5	4.4	4.4	5.5	2.4	3.6	0.7	1.2	2.5	1.0	3.4	1.8	2.0
Total reactive maceral		5.3	9.4	6.4	8.3	11.2	4.2	9.7	8.0	1.6	6.3	2.4	12.8	3.8	7.6
Total mineral matter		84.4	63.6	68.6	79.1	65.2	76.2	78.0	80.4	83.1	90.1	84.5	78.6	68.5	46.9
Wits-Ehac Index		3.09	3.06	-	3.27	3.73	3.10	-	-	-	-	2.98	2.99	-	3.77
Wits-CT Index		1.33	1.30	0.91	0.70	1.60	1.36	0.67	0.27	0.95	0.42	1.18	1.34	1.44	3.99

Table 4.14 Macerals analysis (vol.%, mmf) for the coal-shale

MACERAL Group	MACERAL	Sample no:													
		SA	**SB**	**SC**	**SD**	**SE**	**SF**	**SG**	**SH**	**SI**	**SJ**	**SK**	**SL**	**SM**	**SN**
Vitrinite macerals	Telinite	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	1.9	1.1
	Collotelinite	3.9	2.2	0.0	9.2	3.6	0.8	4.6	7.4	0.0	11.1	0.0	19.6	0.0	5.3
	Vitrodetrinite	0.0	2.2	0.0	0.0	0.0	0.0	0.9	0.0	0.0	0.0	0.0	0.9	0.0	0.0
	Collodetrinite	10.4	2.2	4.5	5.3	5.7	1.7	3.7	7.4	0.0	5.6	2.6	12.1	0.0	2.3
	Corpogelinite	5.2	0.0	0.0	0.0	0.0	0.0	1.8	3.7	0.0	5.6	0.0	0.0	0.6	0.4
	Gelinite	1.3	0.7	0.0	1.3	0.0	0.0	1.8	0.0	2.4	2.8	2.6	0.0	0.0	0.4
	Pseudovitrinite	0.0	0.7	0.0	0.0	0.0	0.0	1.8	0.0	0.0	0.0	0.0	6.5	0.0	0.0
Total vitrinite		20.8	8.1	4.5	15.8	9.3	2.5	14.7	18.5	2.4	25.0	5.2	39.3	2.5	9.5
Inertinite macerals	Fusinite	7.8	11.0	5.1	9.2	1.4	16.9	8.3	0.0	10.8	5.6	6.5	6.5	10.2	4.2
	Reactive semifusinite	0.0	2.9	0.6	1.3	0.0	0.8	4.6	0.0	0.0	0.0	0.0	1.9	0.6	0.4
	Inert semifusinite	22.1	11.0	14.7	6.6	12.9	5.1	11.0	40.7	14.5	11.1	1.3	7.5	5.7	12.5
	Secretinite	0.0	4.4	2.6	2.6	3.6	3.4	3.7	3.7	4.8	0.0	0.0	3.7	4.5	3.4
	Inertodetrinite Reactive	10.4	2.2	1.3	1.3	7.1	4.2	8.3	18.5	0.0	13.9	3.9	2.8	3.2	0.8
	Inertodetrinite Inert	36.4	47.8	57.1	42.1	50.0	56.8	33.0	14.8	60.2	19.4	76.6	22.4	67.5	65.5
Total inertinite		76.6	79.4	81.4	63.2	75.0	87.3	68.8	77.8	90.4	50.0	88.3	44.9	91.7	86.7
Liptinite macerals	Sporinite	2.6	12.5	14.1	21.1	15.7	8.5	15.6	3.7	7.2	25.0	6.5	15.9	5.7	3.8
	Resinite	0.0	0.0	0.0	0.0	0.0	1.7	0.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Total liptinite		2.6	12.5	14.1	21.1	15.7	10.2	16.5	3.7	7.2	25.0	6.5	15.9	5.7	3.8
Total reactive macerals		33.8	25.7	20.5	39.4	32.1	17.8	44.0	40.7	9.6	63.9	15.6	59.8	12.1	14.4
Wits-Ehac Index		3.09	3.06	-	3.27	3.73	3.10	-	-	-	-	2.98	2.99	-	3.77
Wits-CT Index		1.33	1.30	0.91	0.70	1.60	1.36	0.67	0.27	0.95	0.42	1.18	1.34	1.44	3.99

Table 4.15 Percentage distribution of ash oxides (XRF) for the coal (wt.%)

Samples	**SiO_2**	**Al_2O_3**	**Fe_2O_3**	**MgO**	**CaO**	**K_2O**	**TiO_2**	**P_2O_5**	**MnO**	**Cr_2O_3**	**NiO**	**Na_2O**	**LOI**
CA	49.33	24.20	9.28	2.02	10.39	0.88	1.52	0.87	0.07	0.03	0.00	0.09	0.85
CB	34.52	30.08	7.67	3.22	18.70	0.28	0.99	1.41	0.13	0.02	0.01	0.07	1.91
CC	23.72	31.38	26.37	5.75	10.37	0.51	0.64	0.25	0.06	0.04	0.04	0.00	0.76
CD	52.55	28.05	2.82	0.94	11.96	0.61	1.63	0.24	0.05	0.03	0.00	0.05	0.45
CE	53.91	21.38	6.86	1.38	12.37	0.66	1.22	0.97	0.04	0.03	0.00	0.03	1.05
CF	61.90	22.22	10.20	0.65	0.57	1.68	0.95	0.06	0.04	0.03	0.01	0.02	0.14
CG	44.49	28.63	3.09	0.31	11.67	0.36	1.52	8.46	0.18	0.04	0.01	0.08	1.10
CH	39.97	27.82	13.89	0.41	10.41	0.68	1.12	5.27	0.01	0.02	0.01	0.13	1.66
CI	64.05	26.54	4.70	0.45	0.28	1.47	1.54	0.17	0.02	0.03	0.01	0.06	0.48
CJ	23.07	28.58	23.35	2.99	18.83	0.36	1.10	0.50	0.05	0.03	0.01	0.16	0.67
CK	42.05	28.19	20.35	0.22	2.54	0.37	1.96	1.77	0.02	0.04	0.02	0.00	2.40
CL	28.65	25.03	6.83	4.97	24.20	0.36	0.93	4.05	0.13	0.03	0.00	0.03	4.69
CM	26.99	23.76	13.83	4.12	25.77	0.20	0.97	2.99	0.32	0.06	0.02	0.00	0.64
CN	35.58	31.91	8.11	0.81	16.78	0.32	1.37	1.90	0.12	0.03	0.00	0.09	2.88
Average	41.48	26.98	11.24	2.02	12.49	0.62	1.25	2.07	0.09	0.03	0.01	0.06	

where **LOI** is loss on ignition

Table 4.16 Percentage distribution of ash oxides (XRF) for the coal-shale (wt.%)

Samples	**SiO_2**	**Al_2O3**	**Fe_2O_3**	**MgO**	**CaO**	**K_2O**	**TiO_2**	**P_2O_5**	**MnO**	**Cr_2O_3**	**NiO**	**Na_2O**	**LOI**
SA	64.47	23.69	4.95	0.95	0.33	3.50	1.09	0.15	0.06	0.03	0.01	0.21	0.55
SB	56.10	24.95	12.44	1.13	0.60	2.69	1.08	0.24	0.15	0.03	0.01	0.07	0.48
SC	60.78	31.21	2.47	0.76	0.38	1.93	1.26	0.36	0.03	0.04	0.01	0.07	0.66
SD	54.83	25.37	14.44	0.83	0.48	1.94	1.08	0.13	0.15	0.04	0.01	0.05	0.55
SE	60.05	23.51	9.33	1.05	1.27	2.73	1.02	0.16	0.11	0.04	0.01	0.14	0.31
SF	59.90	27.70	5.35	1.14	0.60	2.68	1.20	0.27	0.08	0.04	0.01	0.25	0.40
SG	56.22	38.10	1.01	0.35	0.21	0.68	2.31	0.08	0.02	0.04	0.01	0.03	0.99
SH	60.93	31.81	2.42	0.74	0.21	1.74	1.28	0.22	0.03	0.04	0.01	0.06	0.45
SI	50.74	43.06	1.16	0.45	0.78	0.63	2.02	0.09	0.01	0.07	0.01	0.08	0.27
SJ	60.96	19.12	10.44	2.30	1.10	2.97	1.02	0.27	0.18	0.03	0.01	0.28	0.40
SK	67.93	24.92	1.11	0.25	0.13	2.76	1.56	0.07	0.01	0.04	0.01	0.07	0.48
SL	59.74	30.92	3.39	1.07	0.45	1.94	1.24	0.23	0.03	0.04	0.01	0.09	0.64
SM	56.51	27.17	9.10	1.30	1.42	2.37	1.13	0.22	0.15	0.04	0.01	0.10	0.24
SN	59.60	24.90	9.02	1.18	0.56	2.85	1.00	0.14	0.10	0.04	0.01	0.25	0.22
Average	59.20	28.32	6.19	0.96	0.61	2.24	1.31	0.19	0.08	0.04	0.01	0.13	

Table 4.17 Mineralogical composition (wt.%) for the coal determined by X-ray diffraction

Samples	**Q**	**Sa**	**K**	**P**	**Mu**	**D**	**I**	**Ca**	**G**	**Mn**	**Mc**	**H**	**PC**	**OC (LOI)**
CA	14.21	0.00	25.28	0.75	0.00	1.82	0.00	3.38	0.00	0.00	0.00	0.00	0.00	54.57
CB	1.36	0.00	29.61	1.64	0.00	1.91	0.00	4.75	0.00	0.00	0.00	0.00	0.00	60.73
CC	5.34	0.04	24.22	10.23	0.00	8.74	0.00	0.00	3.66	0.27	0.00	0.00	0.00	47.5
CD	8.48	0.04	27.45	0.28	0.00	0.32	0.00	2.10	0.00	0.05	0.00	0.00	0.00	61.29
CE	15.59	0.01	25.53	0.37	0.00	0.85	0.00	4.56	0.00	0.05	0.00	0.00	0.00	53.03
CF	17.46	0.73	29.77	5.41	2.54	0.22	0.00	4.84	3.11	0.43	0.00	0.37	0.00	35.14
CG	5.11	3.13	23.94	0.26	0.66	0.00	0.00	0.00	0.00	0.03	0.00	0.64	0.00	66.23
CH	10.33	0.00	14.84	3.19	1.23	0.00	5.19	0.00	0.00	0.00	0.00	0.00	0.00	65.22
CI	22.58	0.00	32.62	0.22	4.28	0.00	0.00	0.00	1.17	0.14	2.94	0.00	0.00	36.06
CJ	1.13	0.05	24.19	8.67	4.49	2.56	0.00	5.51	0.00	0.13	0.00	0.00	0.28	52.99
CK	0.66	0.04	23.09	0.32	0.00	0.01	0.00	2.54	0.00	0.00	2.48	0.00	1.09	69.78
CL	0.73	0.70	21.69	4.03	0.00	5.27	0.00	8.28	0.00	0.13	0.00	0.00	0.75	58.42
CM	0.80	0.80	19.67	0.63	2.45	2.37	0.00	3.11	0.00	0.06	4.03	0.00	0.01	66.06
CN	4.02	0.18	23.26	3.15	0.00	0.00	0.00	0.00	0.00	0.00	2.51	0.00	1.19	65.7

Q is quartz (%), **Sa** is siderite (%), **K** is kaolinite (%), **P** is pyrite (%), **Mu** is Muscovite (%), **D** is dolomite (%), **I** is illite (%), **Cc** is calcite (%), **G** is gypsum (%), **Mn** is magnetite (%), **Mc** is microline (%), **H** is haematite (%), **Pc** is plagioclase (%), and **OC** is organic carbon (%).

Table 4.18 Mineralogical composition (wt.%) for the coal-shale determined by X-ray diffraction

Samples	**Q**	**S**	**K**	**P**	**M**	**D**	**I**	**C**	**G**	**M**	**MC**	**H**	**PC**	**OC (LOI)**
SA	29.15	2.37	43.60	0.24	8.75	0.00	0.00	0.00	0.00	0.00	2.80	0.00	2.01	11.06
SB	21.9	9.87	40.05	1.16	9.25	0.23	0.00	0.00	0.00	0.11	6.97	0.00	0.00	10.46
SC	21.14	0.87	52.73	0.29	7.96	0.05	0.00	0.14	0.00	0.06	4.31	0.00	0.00	12.45
SD	23.03	2.12	43.76	3.85	8.95	0.00	0.00	0.00	3.74	0.27	4.34	0.00	0.00	9.94
SE	12.90	1.57	43.13	9.68	8.31	0.00	0.00	0.00	2.30	0.18	5.96	0.00	0.00	15.98
SF	16.56	8.76	46.69	0.35	7.96	1.01	0.00	0.00	0.00	0.03	5.72	0.00	2.23	10.68
SG	26.72	4.20	47.27	0.14	9.16	0.00	0.00	0.00	0.88	0.00	4.32	0.00	1.70	5.55
SH	32.18	0.05	50.07	0.16	5.35	0.00	0.00	0.00	1.33	0.02	4.22	0.00	2.64	3.97
SI	22.12	1.07	48.24	0.07	7.79	0.00	0.00	0.00	0.78	0.00	8.25	0.00	1.71	9.96
SJ	28.11	12.4	32.89	0.19	8.43	0.00	4.13	0.00	0.88	0.00	6.15	0.00	3.82	3.00
SK	22.83	0.35	59.08	0.01	5.66	0.00	0.00	0.00	0.00	0.19	2.44	0.00	0.00	9.44
SL	1.21	0.33	74.74	0.29	5.08	0.29	0.00	0.72	1.08	0.06	0.00	0.00	5.29	10.92
SM	24.36	3.41	44.99	0.05	7.08	0.00	0.00	0.00	0.00	0.00	4.96	0.00	3.98	11.17
SN	8.26	0.00	53.93	0.16	3.64	0.00	0.00	0.04	0.00	0.09	0.00	0.00	0.00	33.88

4.3.1 Relationship between the spontaneous combustion tests and the proximate analysis for coal and coal-shale

The air-dried moisture content varies between 1.6% to 2.5% for coal and 0.8% to 1.7% for coal-shale samples respectively. All the samples have a low moisture content and are more liable to spontaneous combustion than one another. The low moisture content could be caused by the amount of water molecules absorbed on the external surface and internal open pore surface of the coal and coal-shale. The low moisture content of the physically and chemically absorbed water is one of the characteristics of higher rank coal (Stach *et al.,* 1975). It was found that most coal with a lower moisture content show high liability indices toward spontaneous combustion compared with the other coal samples. This is in-line with the studies reported by Beamish & Hamilton (2005) and McPherson (1993). Slight differences were observed in the moisture content of the samples which show good agreement with the study reported by Gurdal (2011) and Gurdal & Bozcu (2011). The differences in the moisture content of the samples between bands of coal seams may have noticeable influences on spontaneous combustion liability. The interaction between the coal and moisture of the investigated samples may be caused by two opposing processes (Wade *et al.* 1987). Firstly, the heat of evaporation occurs when the coal moisture is driven off at the early stage of self-heating. Secondly, the process involves adsorption of water vapour from the air which caused an increase in the coal temperature. The behaviour of the moisture content in one of the study areas as shown in Figure 4.8 indicates an increase in moisture on the coal surface during spontaneous combustion.

The moisture content of coal samples CG to CJ varies from 1.9% to 2.5%, and CA to CC ranges between 2.2% to 2.3%. Also, CD to CF varies between 2.3% to 2.5% and CK to CN have the same moisture content (1.6%) respectively. Coal samples CF and CG have the highest moisture contents, followed by CH and those from CA, CB, CD and CE respectively. The samples have roughly the same moisture contents and are more liable to spontaneous combustion except for samples CF and CI, with lower liability indices. Coal-shale samples have low moisture content similar to the tested coal samples. The ranges in moisture content of the coal-shale are in-line with the study reported by Restuccia *et al.* (2017). Coal-shale samples SE, SD and SN have roughly the same moisture contents among the coal-shale. Coal-shale sample SN is more liable to spontaneous combustion from the results of the liability indices compared to the other coal-shale samples. This may be due to the presence of various microscopic organic and inorganic constituents which accelerate the spontaneous combustion liability. Literature studies revealed

that the moisture content increases the spontaneous combustion liability of coal in addition to parameters such as oxygen, organic matter type, surface area exposed, mineral matter content (particularly pyrite) and rank (Kaymakci & Didari (2002) and Pone *et al.,* (2007)). This may likely be the same case with the studied coal-shale samples.

The air-dried volatile matter content varies between 16.7% to 26.9% for the coal and 6.5% to 16.6% for the coal-shale samples respectively. The ranges in volatile matter content of the coal-shale are in-line with the study reported by Restuccia *et al.* (2017). The volatile matter content for the coal is greater than 20%, except for sample CI (16.7%). Most of the coal samples with the average-high volatile matter have high spontaneous combustion liability indices and vice versa. This supports the studies reported by Banerjee (2000) and Eroglu (1992). Coal-shale sample SN and SE have the highest volatile matter contents and liability indices compared to the other coal-shale samples. It was found that the coal-shale samples with an average-high volatile matter are more liable to spontaneous combustion. This characteristic of a coal-shale with an average to high volatile matter corresponding to a high liability index is in agreement to coal with the average to high volatile matter content.

The air-dried ash content ranges between 13.7% to 48.4% for the coal samples and 51.5% to 88.7% for the coal-shale samples respectively. The ranges in the ash content are in-line with the characteristic of South African coal (Falcon & Ham (1988), Hagelskamp & Snyman (1988), Snyman & Botha (1993) and Van Niekerk *et al.,* (2008)). For the coal-shale, the ranges in the ash content are in-line with the studies reported by Alpern & Lemos de Sousa (2002) and Restuccia *et al.* (2017). The amounts of the ash content in some samples are similar to properties of some Indian coal samples with ash contents greater than 45% (Choudhury *et al.,* 2007). The cause of the high mineral content may be due to the peat depositional environment where the condition of flooding of the paleomire occurred periodically during deposition (Sia & Abdullah (2012), Siavalas *et al.,* (2009) and Zivotic *et al.,* (2013)). It is known that the physical and chemical properties of coal changes during coal oxidation (Taylor *et al.,* 1998). The differences in the ash content may also be caused by the variations in the rate, volume and direction of the influx of clastic or volcano-clastic material into the area during peat formation (Gurdal *et al.* 2015). It was found from the study that the ash content varies between selected bands of coal seams. Coal CK has a very low ash content compared to coal CL, CM and CN from the same coal seam. Furthermore, coal samples CG and CH have an ash content lower than coal samples CI and CJ from the seam same. Coal samples CI and CF have the highest

ash contents and the lowest liability indices. It was indicated that the spontaneous combustion liability of coal decreases with an increasing ash content and vice versa.

Coal-shale samples SN and SE have the lowest ash contents (corresponding to average-high volatile matter and carbon contents) and have the highest liability indices compared to the other coal-shale samples. The coal-shale samples SC, SG, SH, SI and, SJ have the highest ash contents and displayed very low liability indices with the Wits-CT Index but with no liability indices with the Wits-Ehac Index because of their low reactivity. The coal-shale sample with a low ash content indicates a high spontaneous combustion liability index compared to the other coal-shale samples with a high ash content. The variations in their spontaneous combustion liability indices could be due to the influence of mineral absorbing heat within the coal-shale samples (Humphreys *et al.*, (1981) and Smith *et al.*, (1988)). This characteristic is related to those exhibited by coal. Therefore, coal and coal-shale display similar ash behaviour with respect to spontaneous combustion liability. Some mineral matter of ash such as soda, lime and iron compounds may aid spontaneous combustion, while silica and alumina reduces the accelerating effect of spontaneous combustion liability. It is known that certain chemical compounds contribute to spontaneous combustion, while some impede the development of oxidation, thereby lowering the spontaneous combustion liability. Proximate analyses results obtained from the coal-shale may be used as a reference for a new study since limited research has been reported on spontaneous combustion liability of coal-shale.

Figure 4.8 Moisture occurrence on the surface of the coal at Khwezela Mine (Bokgoni Pit), Witbank, South Africa

4.3.2 Relationship between spontaneous combustion tests and ultimate analysis for coal and coal-shale samples.

The air-dried carbon content varies between 35.9% to 69.7% for coal and 2.66% to 33.7% for coal-shale samples respectively. Coal samples CF and CI have the lowest carbon contents and the lowest liability indices, while coal with high carbon contents have high liability indices. Coal-shale sample SN has the highest carbon content and the highest liability index compared to the other coal-shale samples. The high spontaneous combustion liability indices of the studied coal samples could be due to the high carbon content (corresponding to low ash content) compared to the coal-shale samples with considerably low carbon contents (corresponding to high ash content). The capacity to self-heat appears to be directly related to the amount of carbon and ash content present in the samples. Most of the coal samples with high carbon and low ash contents indicate high liability indices, while coal-shale samples with a carbon content of 33.7% and less than 55% ash content are more liable to spontaneous combustion.

The air-dried hydrogen, nitrogen, and calculated oxygen content varies from 2.55% to 4.21%, 0.85% to 1.63%, and 5.65% to 10.35% for coal and 0.75% to 2.87%, 0.08% to 0.96%, and 5.01% to 11.85% for the coal-shale samples. Coal samples CI and CF have the lowest hydrogen and nitrogen contents and the lowest spontaneous combustion liability indices, while most coal samples with high hydrogen and nitrogen contents have high spontaneous combustion liability indices. Coal-shale SN has the highest hydrogen and nitrogen content and the highest spontaneous combustion liability index compared to the other coal-shale samples. Most coal-shale samples with low hydrogen and nitrogen contents have low spontaneous combustion liability indices. Coal sample CH has the lowest oxygen content and high spontaneous combustion liability index, while the highest oxygen content was found in coal sample CB which indicates a high liability index. Coal-shale sample SE has the lowest oxygen content, while coal-shale sample SL has the highest oxygen content. The influence of oxygen content from the obtained results for both coal and coal-shale samples does not clearly show any effects on the spontaneous combustion liability. Hence, the percentage oxygen content does not seem to indicate the liability of the samples toward spontaneous combustion. The study identifies that coal and coal-shale samples with high carbon, hydrogen and nitrogen contents are liable to spontaneous combustion. Therefore, the influence of these factors might play a significant role in evaluating the incidents of spontaneous combustion. The data obtained from the contents

of carbon, hydrogen and nitrogen for the coal-shale samples may be used as a reference since limited research have been reported on spontaneous combustion liability of coal-shale.

The air-dried total sulphur content varies from 0.59% to 5.30% for the coal and 0.12% to 6.90% for the coal-shale samples. Six of the coal samples have sulphur content more than 2%, CC (3.96%), CF (3.42%), CH (2.19%), CJ (5.30%), CL (3.88%), and CN (2.92%) respectively. The presence of a high sulphur content can be related to the peat depositional environment and conditions, and alkaline depositional environments with concentrated sulphide mineralization. Coal deposited in fresh-water and a calcium-rich environment is usually sulphur-rich and this may cause the presence of a high sulphur content in some of the coal (Teichmuller & Teichmuller, 1982). Coal-shale samples SE (6.90%) and SD (2.53%) have high sulphur contents and high liability indices compared to the other coal-shale samples. The ranges in the value of total sulphur of the coal-shale are in-line with the study reported by Rumball *et al.* (1986). The presence of high total sulphur in some of the coal and coal-shale samples may be related to the peat depositional environment as the organic sulphur and pyritic sulphur are high, or due to epigenetic mineralization as the sulphate pyrite and pyritic values are all high. Coal-shale SL has the lowest sulphur content at 0.12% and a low liability index. Hence, the influence of sulphur might play a significant role in evaluating the incidents of spontaneous combustion.

The concentration of the total sulphur and its forms varied from one sample to another. The sulphur forms analyses show that the pyritic, sulphate and organic sulphur contents vary between 0.13% to 4.13%, 0.003% to 0.422%, 0.28% to 1.09% for coal, and 0.038% to 4.26%, 0.003% to 0.450% and 0.05% to 2.15% for the coal-shale samples respectively. The results obtained are in-line with the studies reported on coal by Boudou *et al.* (1987), Gryglewicz *et al.* (2002), Hsieh & Wert (1985), Marinov *et al.* (2005), Olivella *et al.* (2002) and William (1994). The high organic sulphur could be due to the bacterial reduction of sulphate and subsequent absorption of reduced sulphur within the coal matrix (Casangrade & Nug, 1979). Coal and coal-shale rich in organic sulphur may indicate a syngenetic influence of fluids with a high sulphate content. Coal and coal-shale samples (CC, CF, CJ, CL, CN, SB, SD, and SE) with average to high amounts of pyritic and organic sulphur are more liable to spontaneous combustion as shown in Table 4.8 and Table 4.9.

The pyrite content in coal is known to be one of the main factors accelerating spontaneous combustion liability. High amounts of pyrite were found in coal samples CJ, CL, CN and CC, while coal samples CG and CI had the lowest values. The high liability indices found in coal-

shale samples SN and SE compared to the other coal-shale samples might be related to the presence of pyrite reacting with oxygen and moisture (Rumball *et al.*,1986). The reason for the high liability index of coal-shale SE compared to the other coal-shale samples except for coal-shale sample SN could be due to the high pyrite of 4.26%. This supports the studies reported by Stach *et al.* (1975) and Rumball *et al.* (1986). The results obtained illustrate the advance effects of pyrite oxidation on spontaneous combustion liability of coal and coal-shale as shown in Figure 4.9 and Figure 4.10. The burning areas indicate that the areas of spontaneous combsution are causes of consistent ingress of oxygen within the coal seam. The distribution of different minerals may be formed where gas streams appear on the coal surface. These conditions allow sulphur deposits in the form of crystals and their aggregate and other forms of native sulphur and ammonium chloride to be found. This study indicates a similar experience in the burning areas as shown in Figure 4.9.

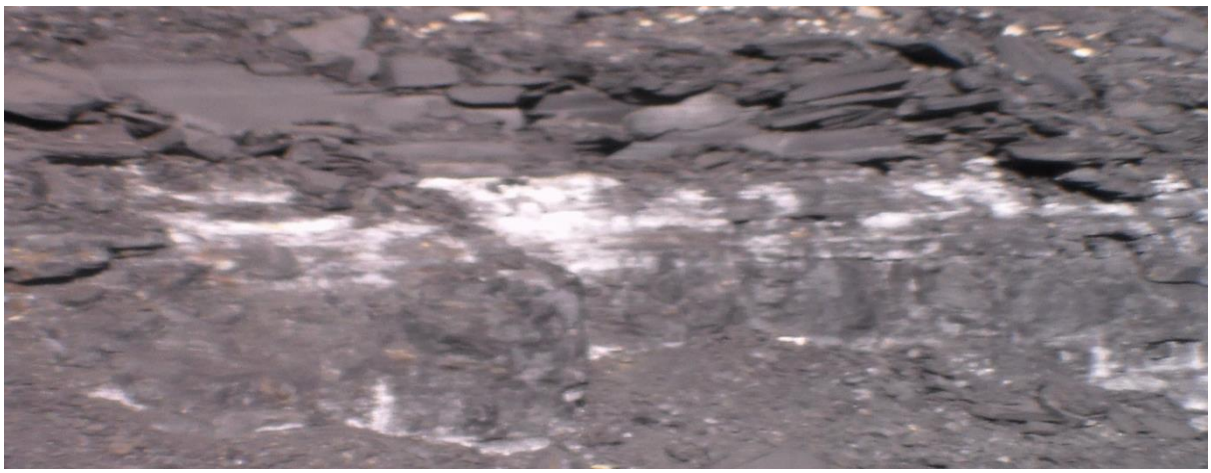

Figure 4.9 Forms of crystals and ammonium chloride present on the surface of coal-shale at iMpunzi Mine, Witbank, South Africa

Figure 4.10 Blooms of sulphur and ammonium chloride on the surface of coal-shale at Tweefontein Mine, Witbank, South Africa

Figure 4.10 shows that a light-yellow to straw coloured sulphur deposits and white powdeish ammonium chloride appeared on the coal-shale surface in one of the studied areas. The presence of sulphur deposits on the coal-shale surface indicated that coal fire may be located beneath the coal seams. Figure 4.9 and Figure 4.10 shows that pyrite reacts exothermally with oxygen to liberate heat and oxidizes to form sulphuric acid which increases the oxidation potential. Figure 4.11 and Figure 4.12 shows crystals of hydrated sulphates on the exposed surface of the coal and highwall. The formation of the hydrated iron crystal sulphates is exothermic and may promote consistent spontaneous combustion.

Figure 4.11 Hydrated iron sulphates on exposed coal surfaces, iMpunzi Mine, Witbank, South Africa

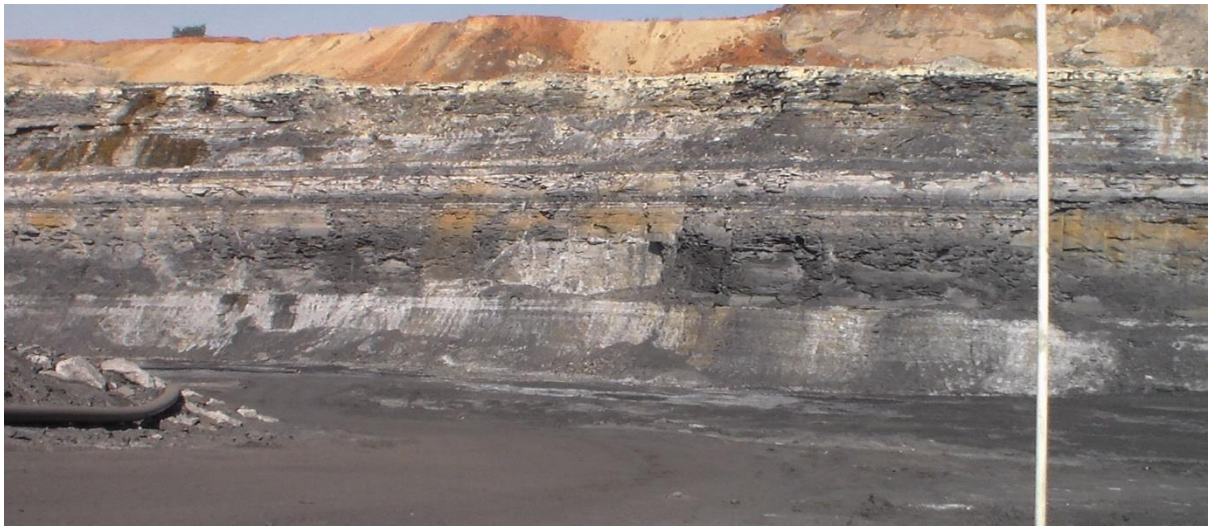

Figure 4.12 Crystals of hydrated iron sulphates on highwall, iMpunzi Mine, Witbank, South Africa

If the self-heating rate of coal and coal-shale is associated with the heat generated and heat dissipated from a single process, it is expected that the temperature differences would basically increase the reaction rate of that process. The carbon contents found in the coal and coal-shale samples varied from one sample to the next, hence, this indicates that different reactions with different activation energies might take place in the spontaneous combustion process. When the carbon contents for the coal-shale samples were compared with each other, samples SN and SE are more liable under the two spontaneous combustion tests than the other coal-shale samples. The event of spontaneous combustion in coal mines may be caused by various organic and inorganic constituents of coal-shale between (above and below) coal seams. Coal-shale can initiate self-heat when it absorbs sufficient oxygen and moisture when exposed to oxygen in the air. The heat generated due to the influence of oxygen may exceed the heat dissipated to the surrounding area via conduction, convection and radiation and accumulates to cause spontaneous combustion of coal-shale.

4.3.3 Relationship between petrographic analyses and spontaneous combustion of coal

Microscopic examinations of the coal samples show that they are characterized by varying amounts of organic and inorganic constituents which may cause the difference in spontaneous combustion liability of each sample. It was found that the relationship between the three maceral groups, petrographically observable mineral matter and the spontaneous combustion tests are not clearly indicated because each sample consists of varying microscopic organic constituents (vitrinite, inertinite and liptinite) and inorganic constituents. The difference in the

spontaneous combustion liability indices could be caused by the variation in the dominant macerals of each sample.

The inertinite group was dominated generally by inertodetrinite and semifusinite. The presence of a high amount of inertodetrinite appears to be certainly very typical of the Witbank and Highveld Coalfields. The sample CH has the lowest value of total inertodetrinite among the high liable coal samples. The high liability of this sample toward spontaneous combustion could be due to its average-high volatile matter content as shown in Table 4.8. Inertinite maceral show that samples with a high fusinite content are more liable to spontaneous combustion than those with a low fusinite content as shown in Table 4.10 and Table 4.11. Samples CF and CI have low fusinite contents and are moderately liable to spontaneous combustion. Therefore, samples with an average-high fusinite content may be liable to spontaneous combustion. It was found that coal with certain amounts (above 20%) of inertodeterinite is more liable to spontaneous combustion when compared with coal containing small amounts of inertodetrinite content as shown in Table 4.10 and Table 4.11. Samples CF and CI contained no amount of reactive inertodetrinite and were moderately liable to spontaneous combustion. Coal samples with high inert inertodetrinite has a high liability index except for sample CH. The high liability of sample CH toward spontaneous combustion may be due to its average-high volatile matter and organic sulphur content as shown in Table 4.8. Sample CI is moderately liable to spontaneous combustion and has a high inert inertodetrinite content as shown in Table 4.10 and Table 4.11. The moderate liability index could be caused by its low volatile matter and high ash content as shown in Table 4.8. Among the coal samples identified to be more liable to spontaneous combustion, only coal sample CH has a high total reactive maceral, while the other samples liable to spontaneous combustion have average total reactive macerals as shown in Table 4.10 and Table 4.11. The cause of the very high total reactive maceral may be due to its average-high volatile matter content as shown in Table 4.8. Coal sample CF has the lowest total maceral and moderate liability index, while coal sample CH with the highest value of total maceral has a high liability index. Therefore, coal with a high amount of total maceral may be more liable to spontaneous combustion than those with a low total maceral.

The spontaneous combustion test results show that most of the studied coal samples are liable to spontaneous combustion. The study indicated that coal with a high amount of inertinite macerals may be considered as reactive constituents of spontaneous combustion than the

vitrinite and liptinite macerals. This is because the inertinite maceral (inertodetrinite and semifusinite) are less compact and naturally have high microscopic and sub-microscopic porosity such as structured inertinites (semifusinite and fusinite) and bands of inertodetrinite which provides pathways for ingress of air within the coal seam more than the vitrinite (Didari (1988) and Falcon (1987)). The temperature of the inertinite constituents might likely increase very fast with shorter ignition rates under oxidation reaction, thereby causing low-temperature oxidation (Misra & Singh, 1994). It was found that coal confirmed to be more liable by the spontaneous combustion tests have high amounts of inertinite macerals as shown in Table 4.10 and Table 4.11. This relationship could be based on the fact that oxygen is transported through the coal surface more quickly due to moisture, volatile matter and porosity. It could be because of this reason that the inertinite-rich South African coal are more liable to spontaneous combustion. Therefore, coal more liable to spontaneous combustion seem to have different petrographic properties. Sample CL has the highest spontaneous combustion liability index despite the high mineral matter as shown in Table 4.10. The high liability index may be due to its average pyrite content as shown in Table 4.8. Sample CC has an average pyrite content but not as liable to spontaneous combustion as sample CL, this may be due to its average-high ash content as shown in Table 4.8. Therefore, the pyrite content present in the coal is more likely to promote spontaneous combustion.

The petrographically observable mineral matter found in the coal may be caused by additional constituents derived by partings, bands and other concentrations of non-coal material between coal seams. It may also be associated with peat accumulation and rank advance, as well as changes in subsurface fluids and other aspects of sediment diagenesis (Wust *et al.,* 2008). Sample CG has the lowest total mineral matter and a high liability index, while sample CF has the highest mineral matter and a low liability index. The values of petrographically observable mineral matter found in the analysed coal are in-line with the study reported by Van Niekerk *et al.* (2008). The cause of petrographically observable mineral matter may be caused by inorganic elements incorporated within the organic constituents of the coal macerals and dissolved salts, and other inorganic substances (Finkelman, 1994). The results show that the spontaneous combustion liability of coal decreases with increasing petrographically observable mineral matter and vice versa. Therefore, coal with low petrographically observable mineral matter content is more liable to spontaneous combustion than those with an average-high petrographically observable mineral matter. This agrees to the fact that coal with high a carbon

and low ash content have a high liability index. Coal with an average to high petrographically observable mineral matter and average to high pyrite content may be more liable toward spontaneous combustion (samples CC, CJ and CL). Hence, most of the fires caused by spontaneous combustion in the affected areas of this study are more likely to have occurred in the areas that have been identified as having low petrographically observable mineral matter content. The occurrence of macerals and petrographically observable mineral matter in the coal samples are shown in Figure 4.13.

4.2.4 Relationship between petrographic analyses and spontaneous combustion for coal-shale.

The maceral groups and petrographically observable mineral matter data were related to the spontaneous combustion liability index. Microscopic observations show that these samples are characterized by varying amounts of macerals and inorganic constituents as shown in Table 4.13 and Table 4.14. The relationships between the three macerals, petrographically observable mineral matter and the spontaneous combustion liability index are not clearly indicated because each sample consists of varying microscopic organic constituents (vitrinite, inertinite and liptinite) and inorganic constituents. The difference in the spontaneous combustion liability index could be caused by variation in the dominant macerals in each sample. Most of the coal-shale samples have average to high inertinite macerals and hence, may be considered to be reactive constituents of spontaneous combustion. Coal-shale confirmed to be more liable to spontaneous combustion contains a certain amount of inert semifusinite and inert inertodetrinite. Inert semifusinite and the inert inertodetrinite among the macerals shows a relationship with the spontaneous combustion liability index. It was found that coal-shale with a high amount of inertinite macerals are more liable to spontaneous combustion than those with low inertinite macerals. This is similar to the characteristics of the studied coal (as explained above) with a high amount of inert semifusinite and inert inertodetrinite. The results imply that the inertinite macerals may be controlled by a number of factors such as temperature and method of oxidation (Scott & Glasspool, 2007). The high total petrographically observable mineral matter content found in the coal-shale samples could be associated with the high contribution of clastic material and conditions unfavourable for the preservation of botanical material. The influence of the high petrographically observable mineral matter may act as heat absorbing mineral within the coal-shale and suppress their liability toward spontaneous combustion (Humphreys *et al.,* (1981) and Smith *et al.,* (1988)). Most of the coal-shale samples

with a high petrographically observable mineral matter have low liability indices and vice versa. This characteristic is similar to the spontaneous combustion liability of the studied coal, although coal-shale contain a higher mineral matter than the coal. Sample SN has the lowest observable mineral matter and the highest liability index. The occurrence of macerals and petrographically observable mineral matter in the coal-shale is shown in Figure 4.14.

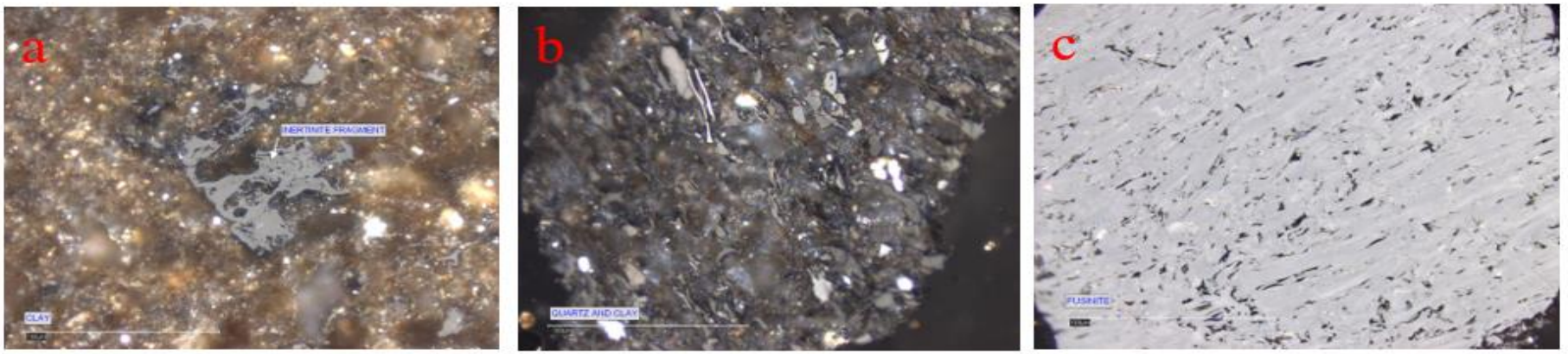

Figure 4.13 Coal CL showing the occurrence of inertinite, quartz and clay and fusinte (Reflected light X 500, Oil immersion, scale=100µm)

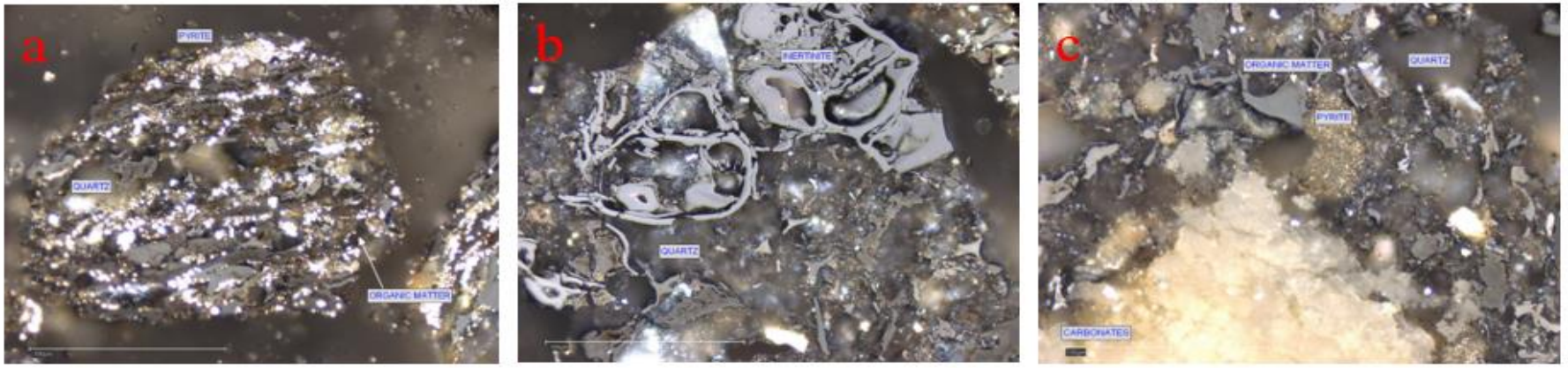

Figure 4.14 Coal-shale SE showing the occurrence of pyrite, quartz, organic matter, inertinite and carbonates (Reflected light X 500, Oil immersion, scale=100µm)

4.3.4 Relationship between ash oxides/inferred mineral matter and spontaneous combustion liability of coal and coal-shale.

Table 4.15 and Table 4.16 presents the results of ash oxides/inferred mineral matter in coal and coal-shale. The identification of different chemical compositions between coal and the coal-shale samples may be attributed to the differences in their ash contents. Coal-shale contains relatively higher amounts of Si, Al, K, Ti, Mg and lower loss on ignition (LOI) than coal. The differences in ash oxides/inferred mineral matter between the coal and coal-shale samples may be due to the amount of inorganic matter and the differences in chemical composition. The SiO_2 content in the coal-shale sample being higher than that of the coal is likely because coal-shale contains a higher quartz content, also Al_2O_3 is slightly higher in the coal-shale sample than the coal as shown in Table 4.15 and Table 4.16. The contents of kaolinite, muscovite and plagioclase may be high in both the coal and coal-shale samples, indicating the content of Al_2O_3 been associated to the mineral content. The values of LOI for the coal-shale sample is lower than the coal, indicating higher amounts of organic contents or unburned carbon in the coal ashes as shown in Table 4.15 and Table 4.16. The coal ash contains higher amounts of CaO than coal-shale, which may occur as dolomite, calcite and gypsum as documented by Koukouzas *et al.* (2006) and Medina *et al.* (2010). The Ca concentrations of the coal-shale is relatively low from that of coal, which indicate possible dissolution of the Ca-containing phases in the coal-shale. The K_2O concentration is also slightly different in the coal and coal-shale samples. The concentration of K_2O in the samples may be associated to the presence of muscovite and microcline in the ashes transformed from those minerals by combustion, which is in-line with the XRD.

The relative elemental abundance (ash oxides) in the coal samples was found in the order of SiO_2> Al_2O_3> CaO>Fe_2O_3>P_2O_5>MgO>TiO_2>K_2O>MnO>Na_2O>Cr_2O_3>NiO. The order of elemental concentration is similar to the order in which these elements are observed in the crustal samples. The high amount of these elements may accumulate in combustion residue (Bhatt, 2003). The coal-shale samples were found to have the relative elemental abundance in the order of SiO_2> Al_2O_3> Fe_2O_3> K_2O> TiO_2> MgO> CaO> P_2O_5>Na_2O>MnO>Cr_2O_3>NiO. Most of the elements were found to be well enriched in both the coal and coal-shale samples. Elements such as Fe, Ca, Mg and P have a higher concentration in the coal than in the coal-shale samples, while Si, Al, K and Ti have a higher concentration in the coal-shale samples than coal. Silica (SiO_2) was found most abundant in both samples. It ranges from 23.07% to

64.05% for coal and 67.93% to 50.74% for the coal-shale respectively. Coal sample CI had the highest value of silica (64.04%) and coal sample CJ had the lowest value (23.07%). Coal-shale sample SK had the highest value of silica (67.93%), while coal-shale sample SI had the lowest value (50.74%). Alumina (Al_2O_3) was found to be the second most abundant constituent, followed by iron oxide (Fe_2O_3) for coal-shale and CaO for coal respectively.

The concentration of alumina was found to be higher in the coal-shale sample SI (43.06%) compared to coal sample (CN) with the highest value of 31.91%. The concentration of NiO was found to be the lowest in both coal and coal-shale samples (0.01%). Both coal and coal-shale samples contains lesser amounts of MnO, Na_2O, Cr_2O_3 and NiO. It was found that the presence of high ash oxides may have influence on spontaneous combustion in coal mines. The ash analysis for both coal and coal-shale samples containing predominantly of SiO_2 and Al_2O_3, followed by CaO and Fe_2O_3 with the relatively small amount of other ash oxides. Coal-shale samples have higher concentration of SiO_2, Al_2O_3, K_2O, TiO_2 and MgO than coal samples. The results obtained from the ash analysis are in-line with the study reported by Pinheiro (1999) on washed South African export coal. Ash oxides such as SiO_2 and Al_2O_3 account for a high amount of the composition in the studied coal and coal-shale samples. The main elemental composition present in all the samples is qualitatively similar to that of natural earthly materials (soils). Figure 4.15 and Figure 4.16 show the percentage distributions of various inferred minerals found in the coal and coal-shale samples.

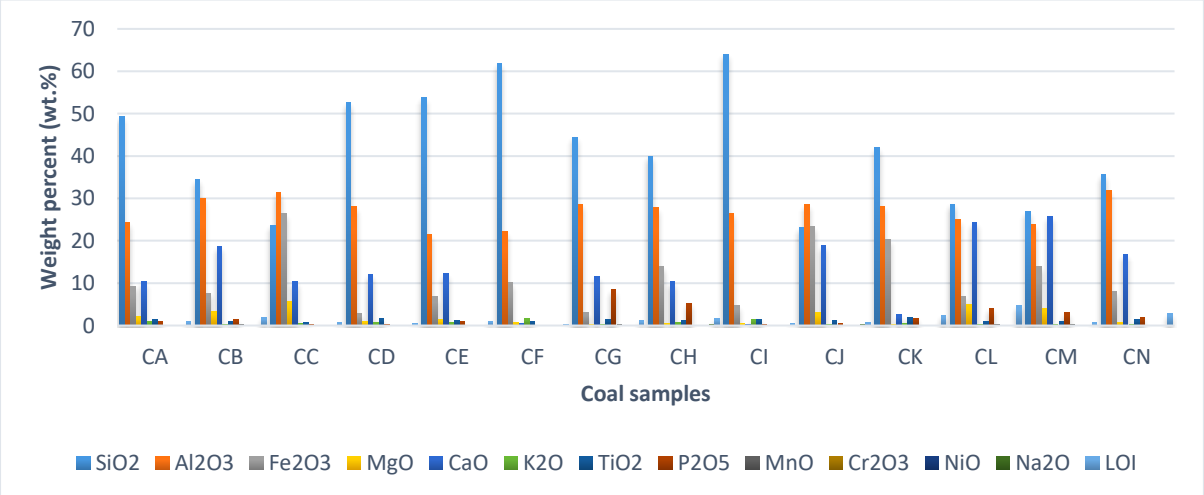

Figure 4.15 Percentage distributions of ash oxides for the coal samples

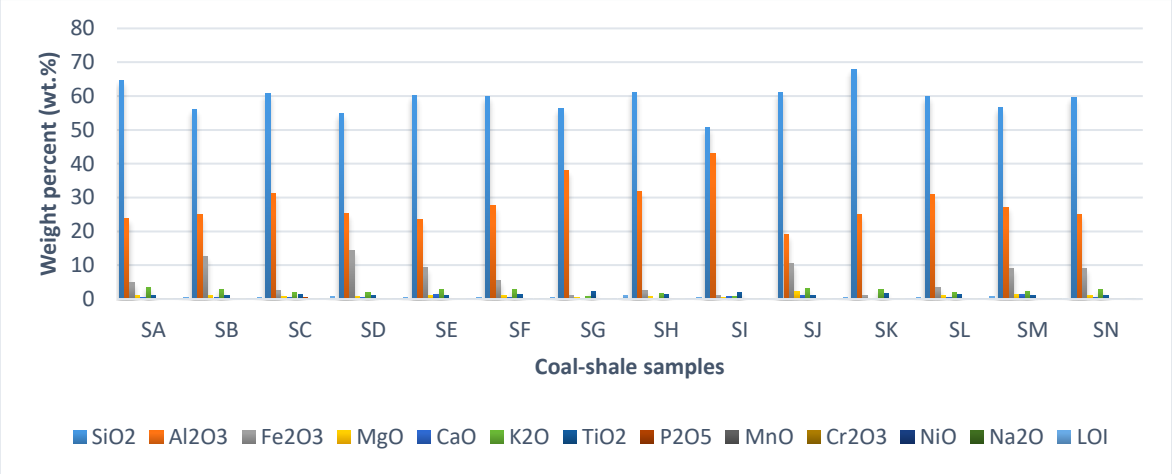

Figure 4.16 Percentage distributions of ash oxides for the coal-shale samples

4.3.5 Relationship between mineral composition and spontaneous combustion liability of coal and coal-shale samples

The mineralogical composition of the coal and coal-shale samples is characterised mainly by kaolinite, quartz, pyrite, siderite, muscovite, dolomite, illite, calcite, gypsum, magnetite, microcline, hematite, plagioclase and organic carbon as shown in Table 4.17 and Table 4.18. The mineral composition of the samples varies from one another, generally dominated by kaolinite and quartz. Slight differences were found in the mineral phase of the samples, which is in agreement with the difference in ash compositions obtained by the XRF. The XRF analysis indicated that most of the samples were rich in ash oxides and inferred minerals such as kaolinite and quartz. The same result is confirmed by the XRD analysis. The study shows that the XRF results are consistent with the XRD data.

The content of quartz in coal-shale is higher than that of coal as shown in Table 4.17 and Table 4.18. Petrographically, the quartz is observed as discrete particles in most cases. This may be caused by the precipitation from Si-rich solutions formed later by the underlying rock formation that had flowed continuously within the coal seams (Ward, 2016). K-feldspar transformed through early coalification or peat accumulation may also be another cause for the silica (Dai *et al.* 2005). Therefore, the high amount of infillings silica reveals an epigenetic cause for quartz. All the samples are generally dominated by kaolinite as shown in Figure 4.18 and Figure 4.19. Coal-shale has higher concentration of kaolinite than coal. The high concentration of kaolinite could be because kaolinite was seemingly obtained from the

variation of the clastic inputs within the coal seams during peat accumulation (Dai *et al.*, 2017). The dominant minerals are relatively high and vary from each sample, which may cause each sample to have a different spontaneous combustion liability index. Furthermore, the high content of minerals such as quartz and kaoloinite may affect spontaneous combustion liability. It was found that mineral matter act as inorganic constituents that modify spontaneous combustion characteristics and is closely related to the ash content in both coal and coal-shale samples. Coal and coal-shale samples with a high mineral matter have a low spontaneous combustion liability index. Pyrite crystals generally occurred as individual and clustered bodies, and disseminated within minerals such as quartz and organic matter as shown in Figure 4.16 and Figure 4.17. The differences in pyrite crystalline accumulations may indicate multistage pyrite formation. The disseminated and massive pyrites present in some of the samples may be associated to the syngenetic and early diagenetic authigenic region, while the replacement pyrite and cleat-infilling pyrite may be formed during the epigenetic periods (Chou (2012) and Querol *et al.*, (1989)). The later might be a measure of the effects of sulphur-rich solutions either in the epigenetic or late diagenetic periods (Dai *et al.,* (2005) and Zhang *et al.,* 2004)).

Coal-shale samples are generally dominated by siderite and plagioclase than coal samples. Coal-shale samples have a higher concentration of muscovite than coal. Furthermore, plagioclase occurred more in coal-shale samples than in coal as shown in Table 4.17 and Table 4.18. Plagioclase and muscovite may be caused by clastic inputs from the erosion of volcanic rocks (basalts) or metamorphic rocks (schists) from the basin margins which could have easily changed into clay minerals (Kostova & Zdravkov, 2007). Illite was found in only two samples (CH and SJ) by the XRD. The origin of this mineral is more likely caused as a weathering product from the samples. Hematite was found in only two of the coal samples (CF and CG). Magnetite and gypsum were found in both the coal and coal-shale samples. Coal samples has a higher amount of calcite occurring as cleat fragments than coal-shale, while coal-shale samples has a higher amount of quartz and microcline which may be attributed to higher amounts of coarse grained quartz, kaolinite, muscovite, microcline and plagioclase than the coal samples. Microcline is generally dominated more in the coal-shale samples than coal. Gypsum was identified in a few samples, generally more in the coal-shale than coal. Gypsum was found in some of the pyrite-bearing samples and may have been formed as a by-product of sulphide oxidation, among others by pyrite oxidation, when the sulphuric acid produced

chemically reacts with calcium carbonate. The presence of high proportions of gypsum within coal seams may likely be a cause of oxidation as reported by Dawson *et al.* (2012).

The relative mineral compositions in the coal samples was found in the order of kaolinite> quartz>pyrite>calcite>dolomite>muscovite>microcline>gypsum>siderite>illite>plagioclase> magnetite>hematite.

The coal-shale samples were found to have the relative mineral compositions in the order of kaolinite>quartz>muscovite>microcline>siderite>plagioclase>pyrite>gypsum>illite>dolomit e>magnetite>clacite>hematite.

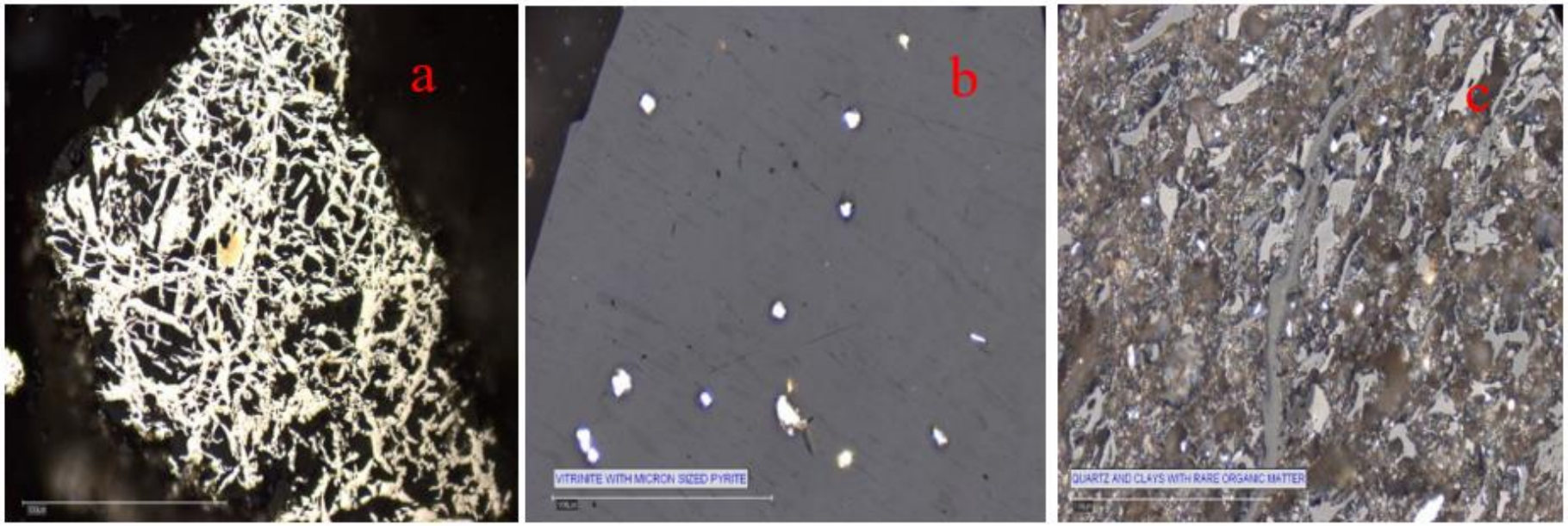

Figure 4.17 Coal-shale SE showing the occurrence of pyrite (b) Coal CJ showing the occurrence of vitrinite with micron size pyrite (c) Coal-shale SI showing the occurrence of quartz and clays with rare organic matter (Reflected light x500, oil immersion, scale=100µm)

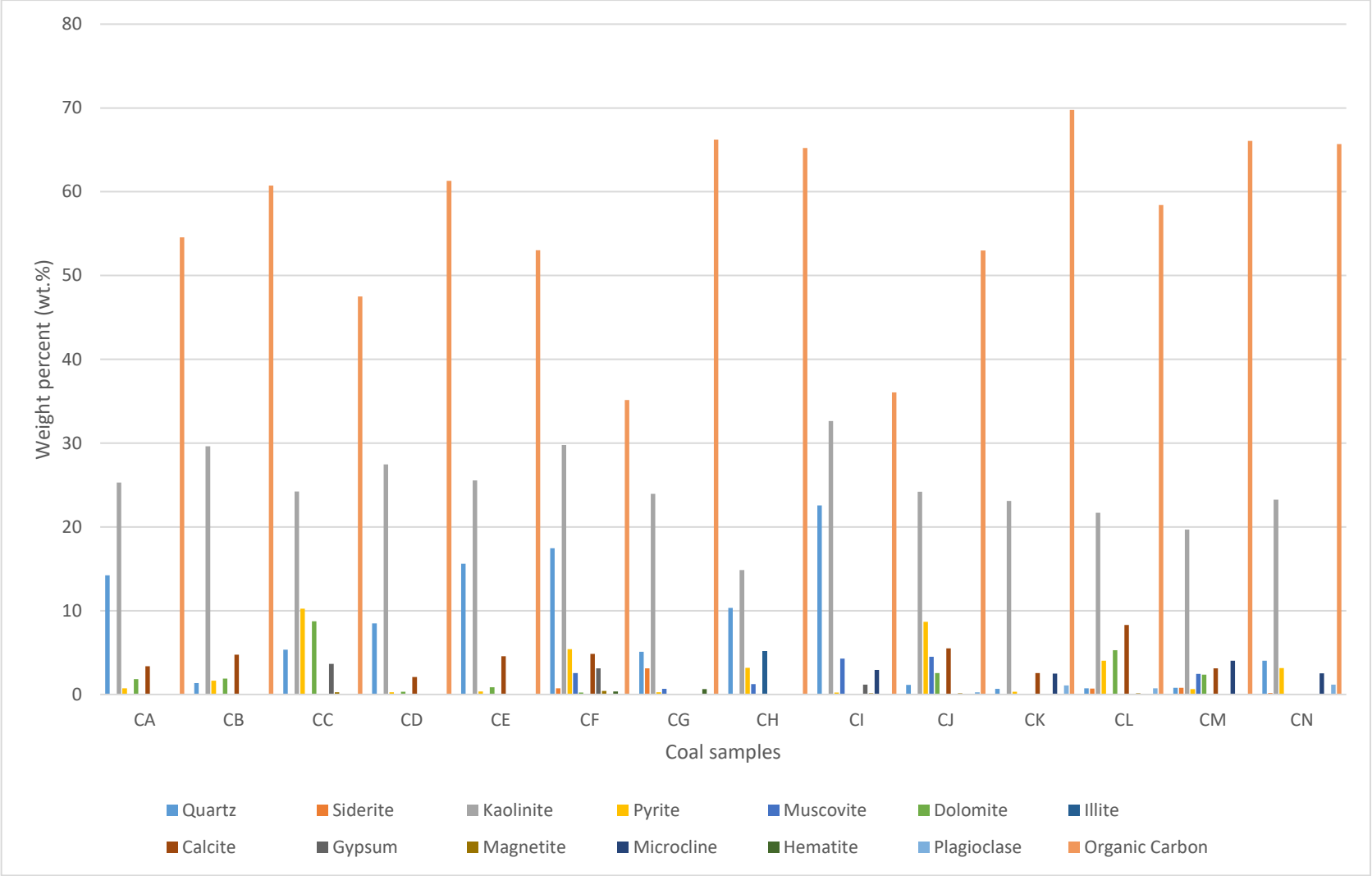

Figure 4.18 Percentage distributions of mineral composition in coal samples

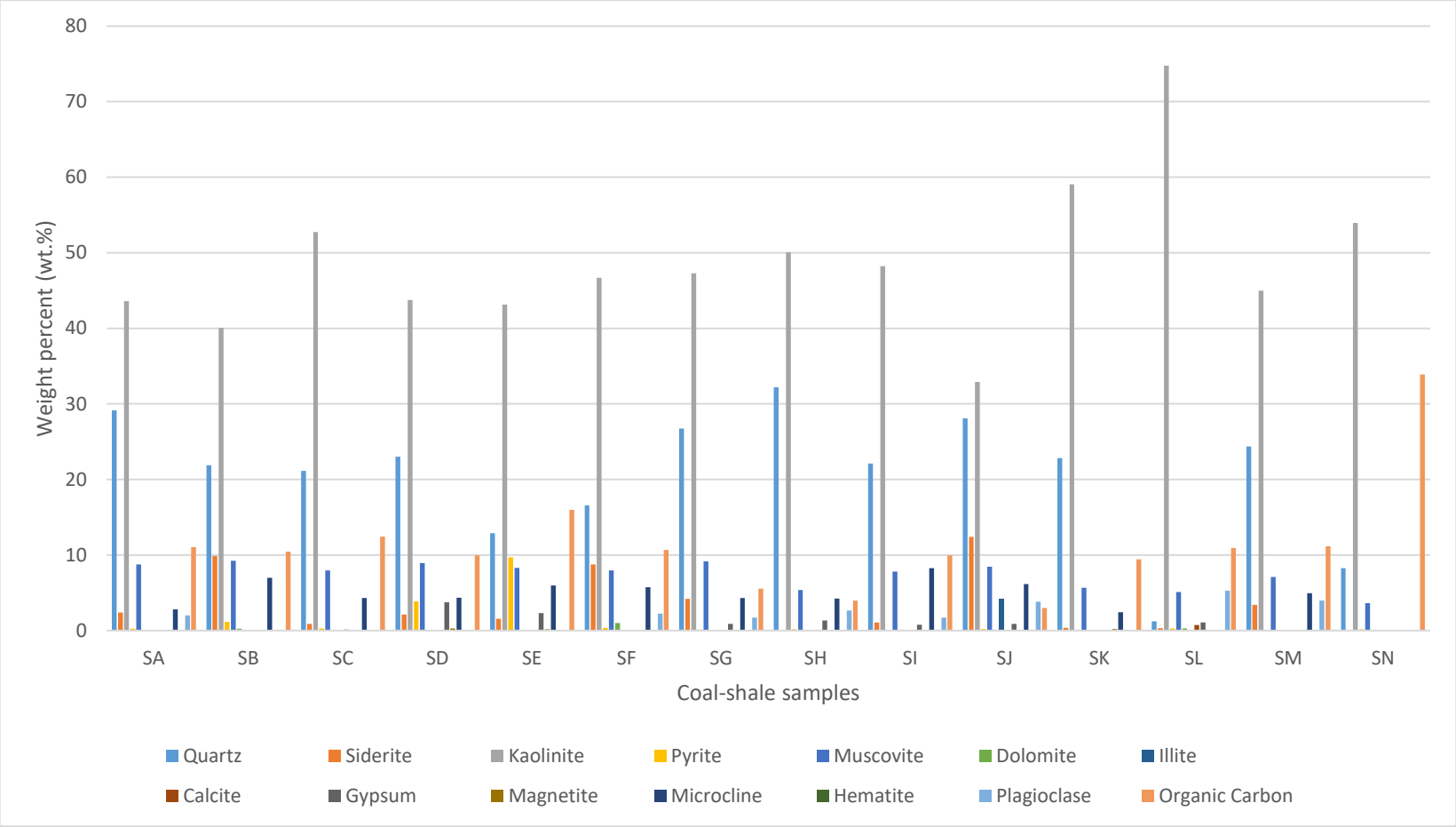

Figure 4.19 Percentage distributions of mineral composition in coal-shale samples

4.4 Summary

A general characterisation which included chemical (proximate and ultimate analysis, total sulphur and forms of sulphur), petrographic, XRF and XRD analysis and as well as spontaneous combustion tests were conducted on fourteen (14) coal and fourteen (14) coal-shale samples. This study has identified a detailed experimental methodology to investigate and evaluate factors affecting spontaneous combustion liability of coal and coal-shale. A detailed report on the equations, calculations and graphical representation of the XPT, Stage II slope and the spontaneous combustion liability indices (Wits-Ehac Index and Wits-CT Index) were discussed. A summary of the results obtained from the geochemical and spontaneous combustion liability tests follows:

This study developed and employed a new spontaneous combustion liability index (Wits-CT Index) based on temperature variations due to chemical reactions between oxygen and coal and coal-shale samples. The Wits-CT Index established has the aim of measuring the spontaneous combustion liability of coal, coal-shale and other carbonaceous materials. The index is found to be a reliable tool to predict the spontaneous combustion liability of different coal and coal-shale. The Wits-CT and Wits-Ehac Index uses different methodologies to measure the liability of a material towards spontaneous combustion. Hence, comparison between the results of the two methods were made with respect to the incident in the mines. The Wits-CT Index evaluates the spontaneous combustion liability index of coal-shale that could not be determined by the Wits-Ehac Index. A new classification for spontaneous combustion liability index of coal and coal-shale samples was established.

The spontaneous combustion tests show that for the coal samples, sample CN (4.84), CM (4.79), CL (4.87), CG (4.91) and CE (4.76) with higher Wits-Ehac Indices will undergo spontaneous combustion faster than the other coal samples due to their high liability indices. However, for the coal-shale, samples SN (3.77) and SE (3.73) will undergo spontaneous combustion faster than the other coal-shale samples based on their higher Wits-Ehac Indices. The results obtained from the Wits-CT Index varies between 4.05 to 9.59 for the coal and 0.27 to 3.77 for the coal-shale respectively. Coal samples CL (9.59), CM (7.27), CN (7.91), CG (7.53) and CK (9.1) with higher Wits-CT Indices will undergo spontaneous combustion faster than the other coal samples with lower Wits-CT Indices. For the coal-shale, samples SN (3.99) and SE (1.66) with higher Wits-CT indices will undergo spontaneous combustion faster than coal-shale samples with lower liability indices.

The study indicated that coal samples have higher intrinsic properties and spontaneous combustion liability index than the coal-shale samples. Both materials show that an increase in carbon, moisture, hydrogen, volatile matter, nitrogen and decrease in ash content can complement spontaneous combustion liability. It was found that the chemical analysis can be used as a tool to measure the liability of coal and coal-shale based on their relationship with the spontaneous combustion test results. A majority of the macerals of South African coal and coal-shale are rich in inertinite fraction than vitrinite and liptinite macerals. The relationships between the three macerals, petrographically observable mineral matter and the spontaneous combustion liability were not clearly indicated because each sample consists of a number of various microscopic organic constituents and inorganic constituents. Furthermore, it was found that dominated macerals in each sample differs, which is more likely to cause each sample to have a different spontaneous combustion liability index.

The XRF results provides reliable information on the concentration of various elements present in the coal and coal-shale samples and indicated that SiO_2 and Al_2O_3 are the dominant oxides. The XRD analysis shows that coal and coal-shale differ in mineralogical composition and are generally dominated by kaolinite and quartz.

This study found that coal and coal-shale tend to undergo spontaneous combustion when exposed to oxygen in the air. Their proneness to spontaneous combustion and intrinsic properties differ between bands of coal seams. This section provides the combined effects of the intrinsic properties on spontaneous combustion liability for predictive purposes. However, the relationship between various intrinsic properties and the spontaneous combustion liability index cannot be established. Therefore, further studies on selected intrinsic properties were conducted to establish relationships with the spontaneous combustion liability indices and develop models using statistical analysis as discussed in the next Chapter.

5 MODELLING SPONTANEOUS COMBUSTION LIABILITY OF COAL AND COAL-SHALE

5.1 Introduction

This section evaluates selected intrinsic properties (proximate, ultimate, total sulphur and forms of sulphur, and macerals) affecting the spontaneous combustion liability of coal and coal-shale using statistical tools (regression). The results of the intrinsic properties were correlated with the spontaneous combustion liability indices (Wits-Ehac Index and Wits-CT Index) using linear and multiple regression analysis. A statistical interpretation of coal and coal-shale analysis data and the combined effects of the major intrinsic factors affecting spontaneous combustion liability for predictive purposes are examined. The best significant parameters along with the most appropriate models were then selected and used for the prediction of spontaneous combustion liability. A study of these factors is important in understanding the spontaneous combustion liability of coal and coal-shale in mines. Furthermore, this will be useful to establish significant relationships between different coal and coal-shale in terms of spontaneous combustion.

5.2 Statistical analysis

In statistics, the correlation coefficient measures the strength and direction of a linear relationship between two variables (dependent and independent variables) on a scatter plot (Deborah (2016) and Stigler (1989)). Its value ranges between -1.0 and 1.0 and is a representation of the linear interdependence of two variables or data set. A correlation coefficient of 1.0 indicates a perfect positive relationship between two variables, i.e. for a positive increase in one variable, there is also a positive increase in the second. A correlation coefficient of -1.0 indicates a perfect negative relationship between the two variables, i.e. the variables move in opposite directions, for a positive increase in one variable, there is a decrease in the second variable. A zero correlation means that for every increase, there is no positive or negative increase (i.e. the two are not related). The R-squared values and the correlation coefficients were used to measure the trends and any significant relationships between the intrinsic properties and the Wits-Ehac Index and Wits-CT Index.

5.2.1 Linear regression

Regression analysis is a type of statistical evaluation that enables three things (Cook, (1979) Fahrmeir *et al.*, (2013), Kuhn & Johnson, (2013) and Rawlings, (1988)):

- Description: Relationships among the dependent variables and the independent variables can be statistically described by means of regression analysis;
- Estimation: The values of the dependent variables can be estimated from the observed values of the independent variables; and
- Prediction: Risk factors that influence the outcome can be identified, and individual prognoses can be determined.

Regression analysis employs a model that describes the relationships between the dependent variables and the independent variables in a simplified mathematical form (Fahrmeir *et al.*, (2013), James *et al.*, (2013), Herrell, (2015) and Heumann *et al.*, (2016)). Linear regression is used to study the linear relationship between a dependent variable Y (spontaneous combustion liability index)) and one or more independent variables X (intrinsic properties). The dependent variable Y must be continuous, while the independent variables may be either continuous or discontinuous. The initial judgment of a possible relationship between two continuous variables should always be made on the basis of a scatter plot (Aneiros *et al.*, (2017), Fahrmeir *et al.*, (2013), Rawlings, (1988) and Zelteman, (2015)).

i. Univariable linear regression

Univariable linear regression studies the linear relationship between the dependent variable Y and a single independent variable X (Fahrmeir *et al.*, (2013), James *et al.*, (2013), Herrell, (2015) and Zelteman, (2015)). The linear regression model describes the dependent variable with a straight line that is defined in Equation 5.1:

$$Y = a + bX \tag{5.1}$$

where **a** is the y-intersect of the line and **b** is its slope.

First, the parameters a and b of the regression line are estimated from the values of the dependent variable Y and the independent variable X with the aid of statistical methods. The regression line enables one to predict the value of the dependent variable Y from that of the

independent variable X (Fahrmeir *et al.,* (2013), James *et al.,* (2013), Herrell, (2015) and Zelteman, (2015)).

The slope b of the regression line is called the regression coefficient. It provides a measure of the contribution of the independent variable X toward explaining the dependent variable Y (Fahrmeir *et al.,* (2013), James *et al.,* (2013), Herrell, (2015) and Zelteman, (2015)). If the independent variable is continuous, then the regression coefficient represents the change in the dependent variable per unit of change in the independent variable. The proper interpretation of the regression coefficient thus requires attention to the units of measurement (Fahrmeir *et al.,* (2013) and Rawlings, (1988)).

ii. Multivariable linear regression

In many cases, the contribution of a single independent variable is not alone sufficient to describe the dependent variable Y. If this is so, a multivariable linear regression to study the effect of multiple variables on the dependent variable can be performed (Fahrmeir *et al.,* (2013), Heumann *et al.,* (2016), Kuhn & Johnson, (2013) and Zelteman, (2015)). In the multivariable regression model, the dependent variable is described as a linear function of the independent variables Xi (Herrell, (2015), Heumann *et al.,* (2016), Kuhn & Johnson (2013) and Zelteman (2015)), as follows in Equation 5.2:

$$Y = a + b_i X_i + b_2 X_2 + \ldots b_n X_n \quad (5.2)$$

The model permits the computation of a regression coefficient bi for each independent variable Xi.

Regression line for a multivariable regression is shown in Equation 5.3:

$$Y = a + b_1 X_1 + b_2 X_2 + \ldots b_n X_n \quad (5.3)$$

where **Y** is the dependent variable, $\mathbf{X_i}$ are the independent variables, **a** is the constant (y-intersect) and **bi** is the regression coefficient of the variable X_i.

Just as in univariable regression, the coefficient of determination describes the overall relationship between the independent variables X_i (intrinsic properties) and the dependent variable Y (spontaneous combustion liability index). It corresponds to the square of the multiple correlation coefficient, which is the correlation between Y and $b_1 X_1 + \ldots b_n X_n$. However, the corrected coefficient is defined above.

Each of the coefficients bi reflects the effect of the corresponding individual independent variable X_i on Y, where the potential influences of the remaining independent variables on X_i have been taken into account, i.e., eliminated by an additional computation (Herrell, (2015), Heumann *et al.,* (2016), Kuhn & Johnson, (2013) and Zelteman, (2015)). Thus, in a multiple regression analysis with intrinsic properties as independent variables and the spontaneous combustion liability index as the dependent variable, the adjusted regression coefficient for intrinsic properties represents the amount of variation in the spontaneous combustion liability index. This is due to one intrinsic property alone, after other intrinsic properties have been taken into account. This is done by a computation that adjusts for other intrinsic properties, so that the effect of one intrinsic property is not confounded simultaneously by the influence of other intrinsic properties.

In this way, multivariable regression analysis permits the evaluation and analysis of multiple independent variables at the same time, with adjustment of their regression coefficients for possible confounding effects between variables (Fahrmeir *et al.,* (2013), Heumann *et al.,* (2016); Kuhn & Johnson, (2013) and Zelteman, (2015)). Multivariable analysis does more than describe a statistical relationship, it also permits individual prediction and the evaluation of the effect of each variable. For the regression model to be robust and to explain Y as well as possible, it should include only independent variables that explain a large portion of the variance in Y. Variable selection can be performed so that only such independent variables are included (Fahrmeir *et al.,* (2013), Heumann *et al.,* (2016); Kuhn & Johnson, (2013) and Zelteman, (2015)).

5.2.2 R-squared value equations and calculations

R-squared is a statistical measure that is used to measure how fit a regression model is. It simply describes how good a model is when compared to the baseline model.

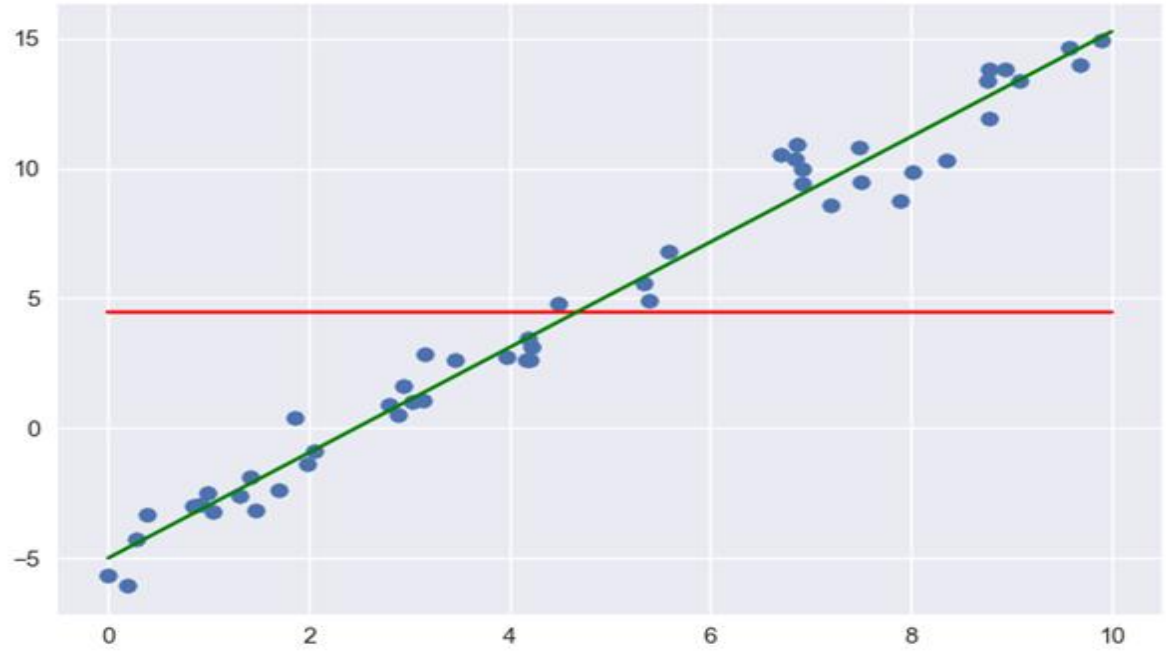

Figure 5.1 (Fahrmeir *et al.*, (2013), Heumann *et al.*, (2016), Kuhn & Johnson, (2013) and Zelteman, (2015))

The red line in Figure 5.1 is the baseline model that usually predicts the mean of observed response of the dependent variable Y as the value of Y irrespective of the value of the independent variables. The green line is the fitted model which makes use of independent variables to predict the value of the dependent variable Y.

Mathematical representation of R-squared is shown as follows:

$$r^2 = 1 - \frac{SS_{Err}}{SS_{Tot}}\pi \tag{5.4}$$

The SS_{Err} or the sum of squares residuals or simply the square of the value of the residuals is defined as follows:

$$\Sigma y_i^2 - B_0\Sigma y_i - B_1\Sigma x_i y_i \tag{5.5}$$

The residual value is the difference between the obtained y-value and the expected y-value. The expected y-value is the calculated value from the equation of line. For example, for a system with 1 unknown parameter/variable x, the calculated y-value would be the sum of B_o and B_1X as shown in Equation 5.6:

$$(\text{i.e. } Y = B_o + B_1X) \tag{5.6}$$

For a system with 2 unknown parameters/variables, X_1 and X_2, the calculated y-value would be the sum of B_o, B_1X_1 , and B_2X_2 as shown in Equation 5.7:

(i.e. $Y = B_o + B_1X_1 + B_2X_2$) (5.7)

And in general

$$Y = B_o + B_1X_1 + B_2X_2 + B_3X_3 + B_4X_4 + \cdots + B_nX_n \quad (5.8)$$

Furthermore, the

$$SS_{Tot} = \Sigma y_i^2 - \frac{(\Sigma y_i)^2}{n} \quad (5.9)$$

where, **n** is the number of observations.

5.2.3 Correlation coefficient equation and calculation

A correlation coefficient is a statistical measure of the degree to which changes to the value of one variable predict changes to the value of another. The formula is shown in Equation (5.10) and all the variables defined.

$$r = \frac{n(\Sigma xy) - (\Sigma x)(\Sigma y)}{\sqrt{\{n(x^2 - (\Sigma x)^2\}\{n\Sigma y^2 - (\Sigma y)^2\}}} \quad (5.10)$$

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

5.2.4 Standard error of estimate

The standard error of estimate is the measure of variation of an observation made around the computed regression line (Fahrmeir *et al.*, (2013), Heumann *et al.*, (2016), Kuhn & Johnson, (2013) and Zelteman, (2015)). It is used to check the accuracy of predictions made with the regression line. Just as the standard deviation measures the variation in the set of data from its mean, the standard error of estimate also measures the variation in the actual values of Y from the computed values of Y (predicted) on the regression line. It is computed as a standard deviation, and here the deviations are the vertical distance of every dot from the line of average relationship. The regression line seeks to minimize the sum of the squared errors of prediction. The square root of the average squared error of prediction is used as a measure of the accuracy of prediction. This measure is called the standard error of the estimate and is designated as σ_{est}. The deviation of each dot from the regression line is expressed as $Y\text{-}Y_e$. The formula for the standard error of the estimate can be expressed as follows:

$$\sigma_{est} = \sqrt{\frac{\Sigma(Y-Y_e)^2}{N}} \quad (5.11)$$

where, **Y** is the actual values, **Ye** is the estimated values and **N** is the number of pairs of (X, Y) points and σ_{est} is the standard error of estimate.

An alternate formula for the standard error of the estimate can be expressed as follows:

$$\sigma_{est} = \sigma_y\sqrt{1-r^2} \quad (5.12)$$

where is σy is the population standard deviation of Y and r is the population correlation between X and Y.

The smaller the value of a standard error of estimate the closer are the dots to the regression line and better is the estimate based on the equation of the line. If the standard error is zero, then there is no variation corresponding to the computed line and the correlation will be perfect. Thus, the standard error of estimate measures the accuracy of the estimated figures, i.e. it is possible to ascertain the goodness and representativeness of the regression line as a description of the average relationship between the two series.

5.3 Discussion

The statistical equations and calculations discussed in section 5.2 were used to establish the relationship between the geochemical (proximate, elemental and various petrographic properties) and spontaneous combustion tests (the Wits-Ehac Index and the Wits-CT Index) results. The initial evaluation of coal and coal-shale intrinsic factors affecting spontaneous combustion liability was based on data obtained from the spontaneous combustion liability indices (Wits-Ehac Index and the Wits-CT Index) and the intrinsic properties (moisture, ash, volatile matter, carbon, hydrogen, nitrogen, total sulphur, pyritic sulphur, sulphate sulphur, organic sulphur and macerals) relating to the samples as determined in the laboratory. It is appropriate to group the data into dependent and independent variables in order to facilitate the analyses. The observed temperature values of spontaneous combustion tests are dependent on the coal and coal-shale properties, coal-forming elements (major and trace elements forming the mineral constituents) and the coal structure. Hence, the statistical analysis was carried out by correlating coal and coal-shale intrinsic properties as the independent variables with the values of the Wits-Ehac Index and the Wits-CT Index as the dependent variables. The range

value is the value of the independent variables from the lowest to the highest. The R-squared value is a statistical tool that is well-known in the context of the statistical model. It is used either to test the hypothesis or predict future outcome on the basis of other related information. It provides a measure of how well observed outcomes are reproduced by a model on the basis of the proportion of total variation of outcomes described by the model (Draper & Smith (1998) and Glantz & Slinker (1990)). The higher the R-squared value, the better the fit is. In this study, the R-squared value was used to measure how close the data are to the fitted regression line and to measure the strength of the relationship between the model and dependent variable. The study interpreted the linear relationship between the intrinsic properties and spontaneous combustion liability index based on the set criterion as shown in Table 5.1.

Table 5.1 Criterion for predicting factors affecting spontaneous combustion liability for the coal and coal-shale

Category	Criterion	Remarks
1	Correlation coefficient/R-squared value between 0.95 to 1 or -0.95 to -1	Variable indicate a perfect positive or negative linear relationship
2	Correlation coefficient/ R-squared value between 0.51 to 0.94 or -0.51 to -0.94	Variable indicate a strong positive or negative linear relationship
3	Correlation coefficient/ R-squared value between 0.25 to 0.50 or -0.25 to -0.50	Variable indicate a moderate positive or negative linear relationship
4	Correlation coefficient/ R-squared value between 0.1 to 0.24 or -0.1 to -2.24	Variable indicate a weak positive or negative linear relationship
5	Correlation coefficient/ R-squared value less than 0.1 but not zero	Variable indicate a very weak positive or negative linear relationship
6	Correlation coefficient of zero	Variable indicate no linear relationship at all

The criterion is established based on the strength and directions of a linear relationship between the intrinsic properties and spontaneous combustion liability index on a scatter plot. Statistically, correlations of +1 or -1 are expected, however, this is not possible due to the use of real dataset from experimental tests, and real data are never perfect to get a perfect correlation coefficient (Deborah, 2016). Since the results of the real data is expected to determine how close is close enough to indicate a strong, moderate or weak linear relationship, this necessitated the need to interpret the correlation coefficient and R-squared values, and classify and find how close they are using the set criterion as shown in Table 5.1. Given that

the correlation coefficient measures the degree to which how two variables vary together, establishing a criterion means whether each intrinsic property and spontaneous combustion liability index vary strongly, moderately or weakly and if it is a positive or negative relationship. The higher the correlation coefficient, the stronger the relationship is (higher correlation coefficients means a better prediction). The results of the geochemical tests and the spontaneous combustion liability indices (Wits-Ehac Index and the Wits-CT Index) were correlated statistically and the linear regression graphs are shown in Figures 5.2 to 5.23. The linear regression graphs were obtained using the intrinsic properties on the x-axis (independent variable) and the spontaneous combustion liability index on the y-axis (dependent variable). A summary of the R-squared and correlation coefficient values are in Appendix 9.3. The overall database involved the spontaneous combustion test results and intrinsic properties of fourteen (14) coal and 14 coal-shale samples in order to determine the factors affecting spontaneous combustion.

As a preliminary step, the experimental data was analysed using statistical methods to ascertain whether any of the parameters were linearly related to the oxidation potential of the coal and coal-shale. Each factor has an influence towards spontaneous combustion liability based on their relationships. The higher correlation coefficients obtained from the data sets suggest that this may be the best way to minimize the overriding effect of the various coal and coal-shale on analysis of the general oxidation trends. Thus, obtaining a result which may be an indication of spontaneous combustion liability of certain coal and coal-shale. Coal and coal-shale behaves differently when exposed to oxygen in the air, due to differences in their intrinsic properties. The analysis of variable pairs (moisture/Wits-Ehac Index; moisture/Wits-CT Index; volatile matter/Wits-Ehac Index; volatile matter/Wits-CT Index; ash/Wits-Ehac Index; ash/Wits-CT Index and etc.) demonstrated uniform trends, i.e. the spontaneous combustion liability increases with increasing volatile matter, carbon, hydrogen, etc. and decreases with increasing ash and mineral matter, etc. for the coal and coal-shale. A study reported by Eroglu (1992) indicated that working with coal having similar rank makes it difficult to establish significant relationships between the petrographic composition and spontaneous combustion liability index.

Based on the criterion set as provided in Table 5.1 and the linear regression graphs illustrated in Figures 5.2 to 5.23, it was found that contents of moisture, ash, volatile matter, carbon, hydrogen, nitrogen, sulphate sulphur, inertinite maceral (fusinite), total maceral and petrographically observable mineral matter with strong linear relationships are factors affecting

spontaneous combustion liability of coal. For the coal-shale, contents of moisture, ash, volatile matter, carbon, hydrogen, nitrogen, sulphur and its group (pyrite, sulphate and organic sulphur), total inertinite and its group (fusinite, total semifucinite, total inertodetrinite), total maceral and petrographically observable mineral matter with strong linear relationships are the main factors affecting spontaneous combustion liability of coal-shale. It was found that each intrinsic property has an influence on the spontaneous combustion liability and their strength varies from one another. Studies reported by Falcon (2004), Gouws & Wade (1989a), Kaymakci & Didari (2002), Nimaje & Tripathy (2016) and Sahu *et al.* (2005) indicated that intrinsic properties are a measure of spontaneous combustion liability. Weak linear relationships were found between the spontaneous combustion liability index and major macerals (total vitrinite, total inertinite and total liptinite) which necessitated the need to investigate the relationships between various constituents in each maceral and the spontaneous combustion liability index. The unsatisfactory (very weak) correlation coefficients and R-squared values obtained from the petrographic analysis and spontaneous combustion liability index may be due to the samples having similar rank values. This is in agreement with the study reported by Eroglu (1992).

5.3.1 Effects of intrinsic properties on spontaneous combustion of coal and coal-shale

The linear regression illustrating the effects of coal and coal-shale intrinsic properties on spontaneous combustion liability are shown in the Figures 5.2 to 5.23.

Figure 5.3l and Figure 5.5a shows the effect of moisture content on the spontaneous combustion liability of the coal and coal-shale obtained from the linear regression analysis. The results indicated a negative correlation for the coal and a positive correlation for the coal-shale. As the correlation displays an R-squared value of 0.3418 for coal, it shows that the linear model has a moderate strength. It is different for the coal-shale. Here the R-squared value is higher, 0.5952, than the coal. This means the coal-shale has a strong effect size.

The R-squared value for volatile matter is 0.2666 for coal and 0.4763 for coal-shale as shown in Figure 5.2b and Figure 5.6m**.** This is an indication that both coal and coal-shale have a moderate effect size. There is a positive relationship between volatile matter and spontaneous combustion liability index for both cases. This means that as the volatile matter increases, the self-heating capacity of coal and coal-shales is more likely to increase in general. This is in-line with the study reported on coal by Singh & Demirbilek (1987).

There is a negative relationship between the ash content and spontaneous combustion liability index for both coal and coal-shale. This means that as ash content increases, the spontaneous combustion liability decreases. This is in-line with the study reported on coal by Beamish & Blazak (2005) and Singh & Demirbilek (1987). The relationship between the ash content and spontaneous combustion liability index seems to be strong. The R-squared value is 0.75 for coal and 0.8926 for coal-shale as shown in Figure 5.3n and Figure 5.6n. The value for coal and coal-shale have a strong effect size. The decreasing ash content affects the self-heating potential by causing a relatively high spontaneous combustion liability for both the coal and coal-shale analysed. This is in agreement with the studies reported on coal by Beamish & Arisoy (2008a), Humphreys *et al.* (1981), Panigrahi & Sahu (2004) and Smith *et al.* (1988).

The R-squared value for the carbon content is 0.7336 for coal and 0.9273 for coal-shale as shown in Figure 5.3o and Figure 5.6o. The relationship between the carbon content and spontaneous combustion liability index for coal and coal-shale seems to be strong. The value for coal and coal-shale have a strong effect size. There is a positive relationship between the carbon content and spontaneous combustion liability index for both coal and coal-shale. This means that as the carbon content increases, the self-heating potential of coal and coal-shale are more likely to increase in general. The increasing carbon content affects the self-heating potential of both coal and coal-shale by causing a relatively high spontaneous combustion liability.

There is a positive relationship between the hydrogen content and spontaneous combustion liability index for both the coal and coal-shale. This means that as the hydrogen content increases, the self-heating potential of coal and coal-shale is more likely to increase in general. The increasing hydrogen content affects the self-heating potential of both coal and coal-shale by causing a relatively high spontaneous combustion liability. The R-squared value for the hydrogen content is 0.5703 for coal and 0.7742 for coal-shale as shown Figure 5.3p and Figure 5.6p. The relationship between the hydrogen content and spontaneous combustion liability index for coal and coal-shale are a strong positive.

The R-squared value for the nitrogen content is 0.7026 for coal and 0.8953 for coal-shale as shown in Figure 5.3q and Figure 5.6q. The relationship between the nitrogen content and spontaneous combustion liability index for coal and coal-shale are a strong positive. This means that as the nitrogen content increases, the self-heating potential of coal and coal-shale is more likely to increase in general.

Figure 5.3r and Figure 5.6r show the effect of calculated oxygen on the spontaneous combustion liability of the coal and coal-shale obtained from the linear regression analysis. The evaluation of the calculated oxygen indicated a negative correlation for the coal and a positive correlation for the coal-shale. As the correlation in Figure 5.3r displays an R-squared value of 0.0103 for coal, it shows that the linear model does not fit well. It is different for the coal-shale. Here, the R-squared is a bit higher, 0.1052, than the coal. The coal and coal-shale have a very weak effect size.

The evaluation of the total sulphur indicated a negative correlation for the coal and a positive correlation for the coal-shale as shown in Figure 5.4 and Figure 5.5h**.** This is in-line with the study reported on coal by Singh & Demirbilek (1987). As the correlation in Figure 5.4s displays an R-squared value of 0.0047 for coal, it shows that the linear model does not fit well. It is different for the coal-shale. Here, the R-squared value is a bit higher, 0.3353, than the coal. The value of coal has a very weak effect size, while the value for coal-shale has a moderate effect size.

The R-squared value for pyritic sulphur is 0.0076 for coal and 0.3254 for coal-shale as shown in Figure 5.2i and Figure 5.5i. The evaluation of the pyritic sulphur indicated a positive correlation coefficient for both the coal and coal-shale. This means that as the pyritic sulphur increases, the self-heating potential of coal and coal-shales is more likely to increase in general. This is in-line with the study reported on coal by Singh & Demirbilek (1987). The scatter plots show that the corresponding spontaneous combustion liability values display a wide variation. The value for coal has a very weak effect size, while coal-shale have a moderate effect size. The linear regression analysis was quite similar to those obtained from the total sulphur content data.

The R-squared value for sulphate sulphur is 0.5806 for coal and 0.2878 for coal-shale as shown in Figure 5.4u and Figure 5.6j. The value for coal have a strong effect size, while the value for coal-shale have a moderate effect size. The evaluation of the sulphate sulphur indicated a negative correlation for the coal and a positive correlation for the coal-shale. This means that as sulphate sulphur increases for the coal, the self-heating potential decreases, while for the coal-shale as the pyritic sulphur increases, the self-heating potential is more likely to increase in general. This is in-line with the study reported on coal by Singh & Demirbilek (1987). The scatter plots show that the corresponding spontaneous combustion liability values display a

wide variation which is quite similar to that obtained from the total sulphur and pyritic sulphur data.

The evaluation of the organic sulphur indicated a negative correlation for the coal and a positive correlation for the coal-shale. This means that as organic sulphur increases for the coal, the self-heating potential decreases, while for the coal-shale as the organic sulphur increases, the self-heating potential is more likely to increase in general. This is in-line with the study reported on coal by Singh & Demirbilek (1987). The R-squared value for organic sulphur is 0.0849 for coal and 0.3519 for coal-shale as shown in Figure 5.4v and Figure 5.6k. The value for coal have a very weak effect size, while the value for coal-shale have a moderate effect size.

The graphs of the major macerals (vitrinite, inertinite and liptinite) and spontaneous combustion liability index indicates that there may be a relationship even though the R-squared values and correlation coefficients are very weak in most cases. A similar study is reported by Eroglu (1992). Although satisfactory correlations were determined for some of the intrinsic properties (such as proximate, ultimate and forms of sulphur) but petrographic properties such as vitrinite and liptinite macerals showed very weak correlation coefficients and R-squared values except for some inertinite macerals which showed a weak-moderate relationship with the spontaneous combustion liability index. The correlation coefficients were unsatisfactory for most of the macerals (majorly the vitinite and liptinite macerals) which motivated the need to analyse further the various macerals in each group (such as collotelinite, collodetrinite, fusinite, inert semifusinite, reactive semifusinite, inertodetrinite, sporinite etc.). The analysis showed that the relationships between the various macerals and self-heating potential may be significant when the maceral constituents were unpacked either in the mineral matter free basis (mmf) or inclusive of mineral matter. It was found that the majority of the vitrinite macerals (collotelinite and collodetrinite) and liptinite macerals (sporinite) in both coal and coal-shale have a very weak size effect as shown in Figures 5.8 to 5.23**.** Inertinite macerals (total inertodetrinite, total semifusinite, fusinite and secretinite) shows a better linear relationship to the self-heating potential, more than the vitrinite and liptinite macerals. The study indicated that the self-heating potential of coal and coal-shale may be influenced by the amount of each maceral composition. This is in-line with the studies reported on coal by Eroglu (1992), Falcon (1987) and Scott & Glasspool (2007). Petrographically observable mineral matter in the coal-shale show a better linear relationship to self-heating potential than the coal as shown in Figure 5.21e and Figure 5.23e. The results of the linear regression analysis obtained indicated that the various macerals have either a positive or a negative effect size as shown in Figures 5.8 to 5.23.

The study showed that there is a tendency of an increase in intrinsic properties such as moisture, volatile matter, ash, carbon, total sulphur, calculated oxygen, pyritic sulphur, organic sulphur, inertinite macerals and petrographically observable mineral matter in both coal and coal-shale but this seems to be more pronounced for coal-shale than for the coal, while coal seem to be more pronounced in terms of hydrogen, nitrogen and sulphate sulphur content than coal-shale. The inconsistencies (i.e. an independent variable with a positive correlation coefficient with the Wits-Ehac Index may have a negative correlation with the Wits-CT Index) in the correlation coefficients between the dependent (Wits-Ehac Index and Wits-CT Index) and independent (some of the intrinsic properties) variables cannot allow accurate determination of the major intrinsic properties affecting the spontaneous combustion liability. Eroglu (1992), Kaymakci & Didari (2002) and Singh & Demirbilek (1987) reported different correlation coefficients between some intrinsic factors (moisture, volatile matter, carbon, nitrogen, inertinite, vitrinite, exinite, total sulphur, pyritic sulphur, organic sulphur and calorific value) and spontaneous combustion liability indices. The inconsistencies in the trend of correlation coefficients necessitated the developed models as shown in Table 5.2. It is known that different methods have been used by previous studies to measure the spontaneous combustion liability of coal but no standard method was considered the best in predicting spontaneous combustion liability (Beamish *et al.,* (2001), Cliff *et al.,* (1998), Gouws *et al.,* (1991), Stott (1980) and Wade *et al.,* (1987)). This study measures the spontaneous combustion liability of coal and coal-shale using two different spontaneous combustion test methods (the Wits-Ehac Index and the Wits-CT Index) and evaluates the factor affecting these materials using statistical analysis. The two liability indices have been used to predict the spontaneous combustion liability of these materials and the results are in-line with the incidents of spontaneous combustion in the mines. Limited studies have been conducted to establish relationships between two or more different spontaneous combustion liability indices and selected intrinsic properties (Eroglu (1992), Kaymakci & Didari (2002) and Singh & Demirbilek (1987)). This study established that some intrinsic properties show different linear trends (i.e. positive or negative correlation coefficients) with spontaneous combustion liability indices due to the method of calculating the value of each index.

The R-squared value was used in this study to measure the effect of each intrinsic property on spontaneous combustion liability indices and also to establish criterion to classify the effect of each property as either strong, moderate or weak linear relationship.

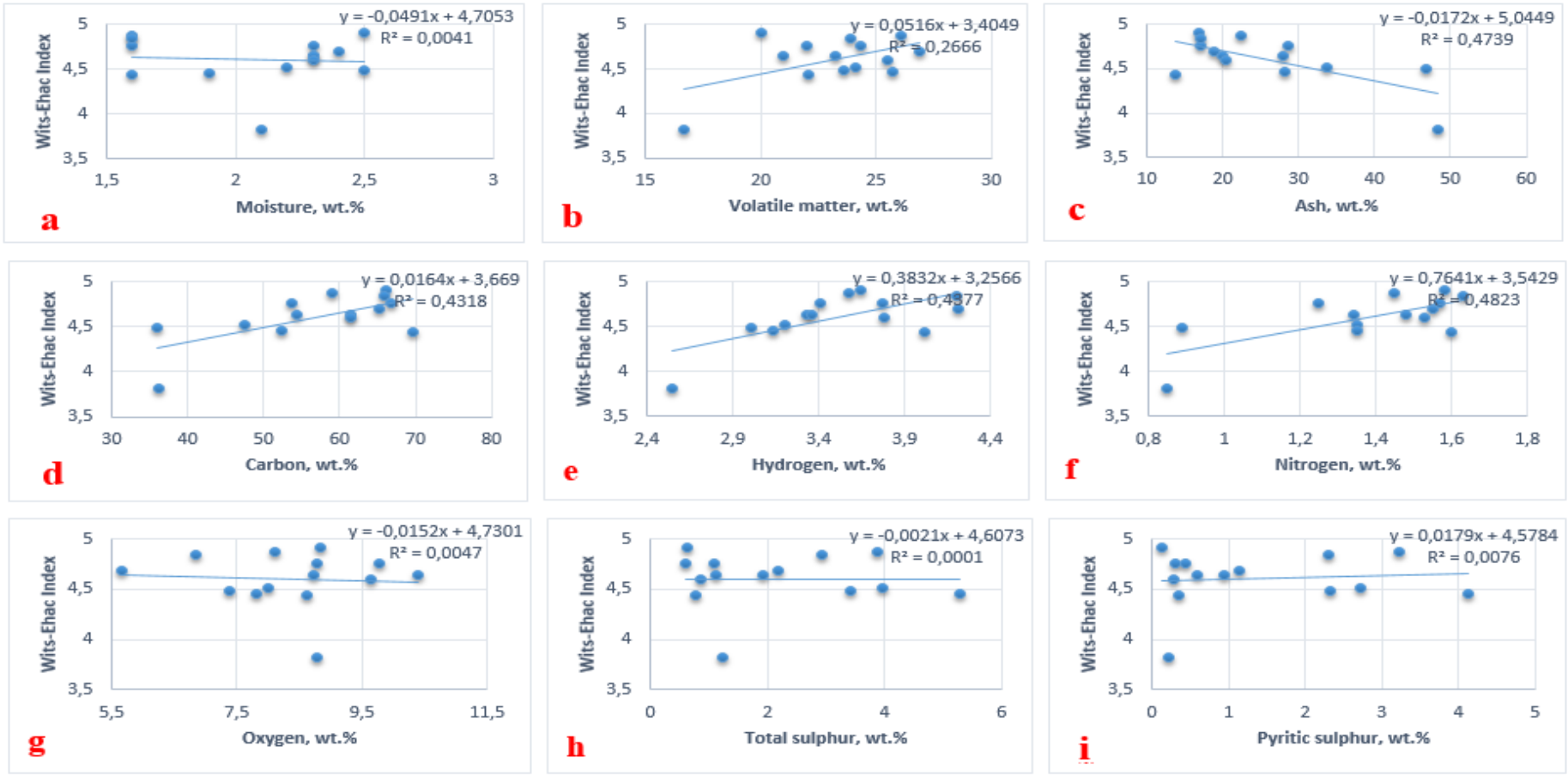

Figure 5.2 Influence of proximate and ultimate properties on spontaneous combustion liabilty for the coal (a to i)

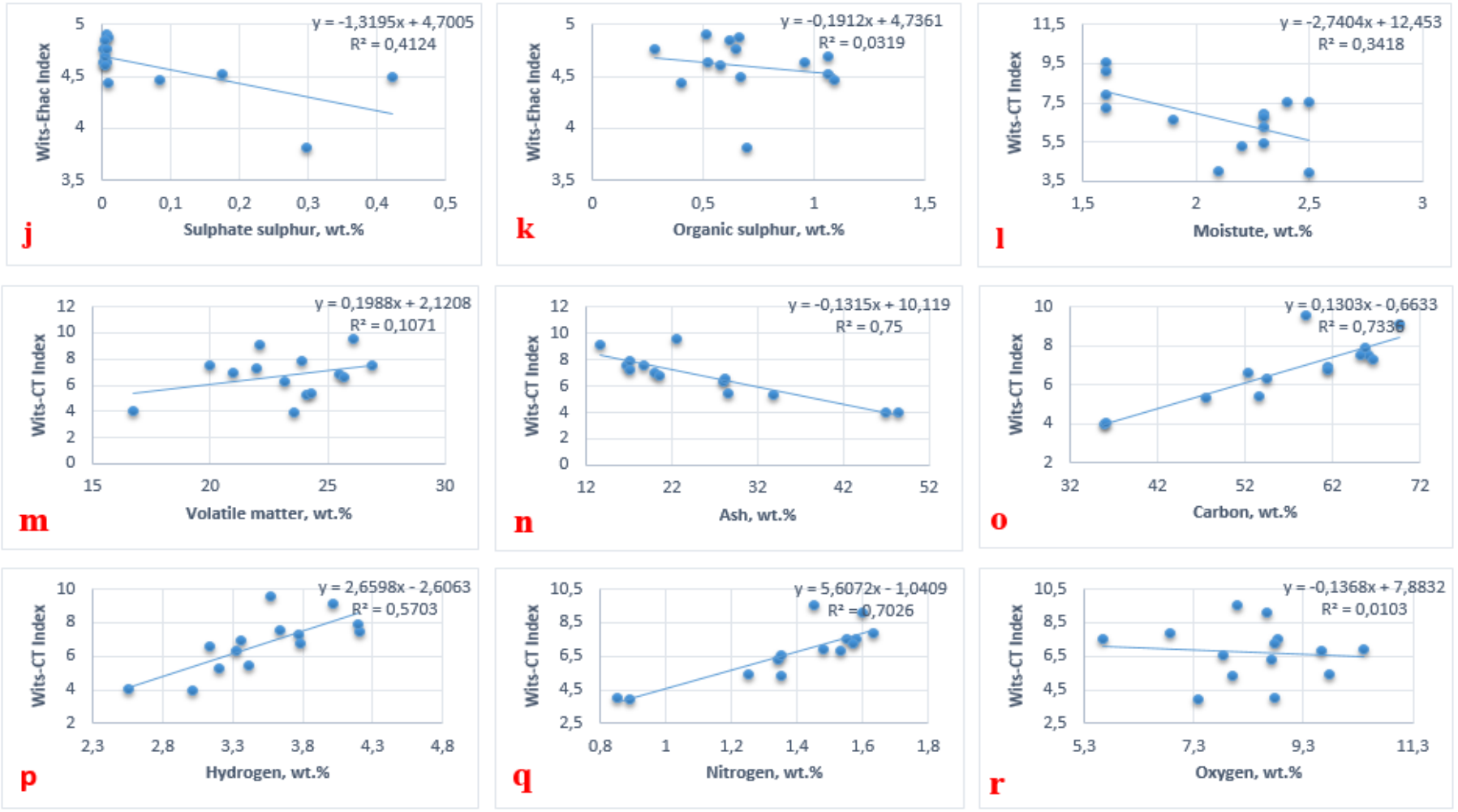

Figure 5.3 Influence of proximate and ultimate properties on spontaneous combustion liability for the coal (j to r)

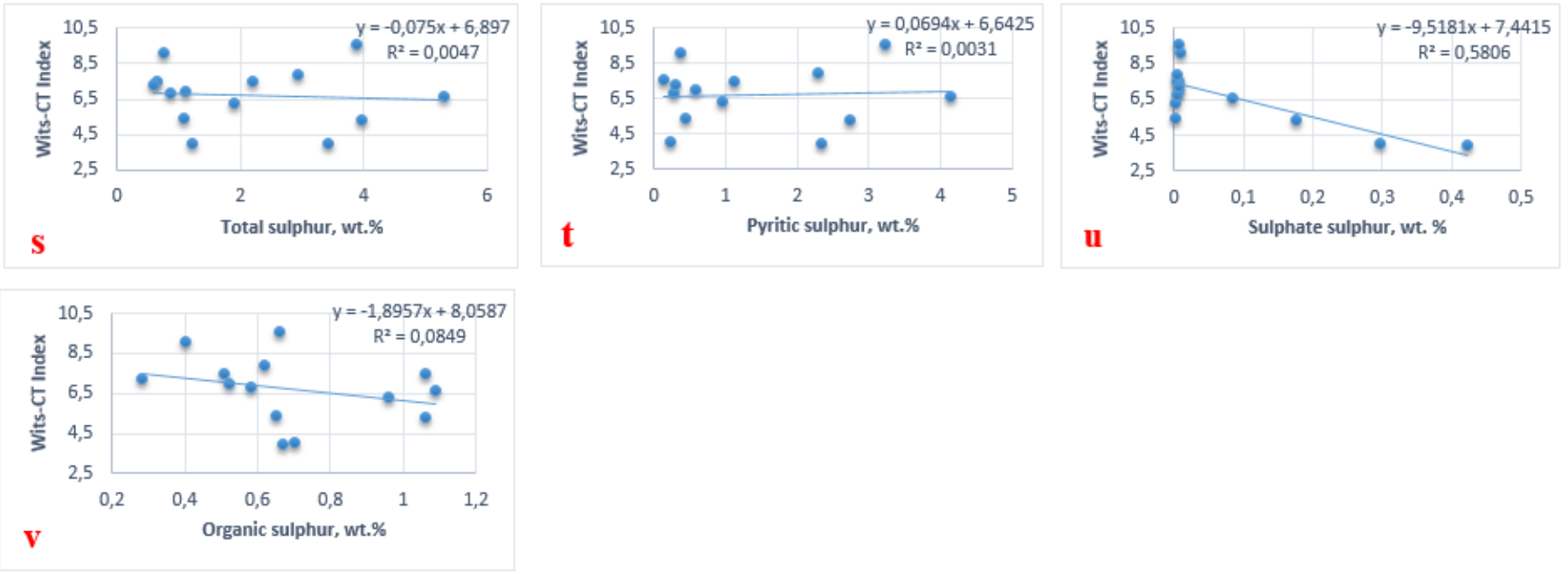

Figure 5.4 Influence of proximate and ultimate properties on spontaneous combustion liability for the coal (s to v)

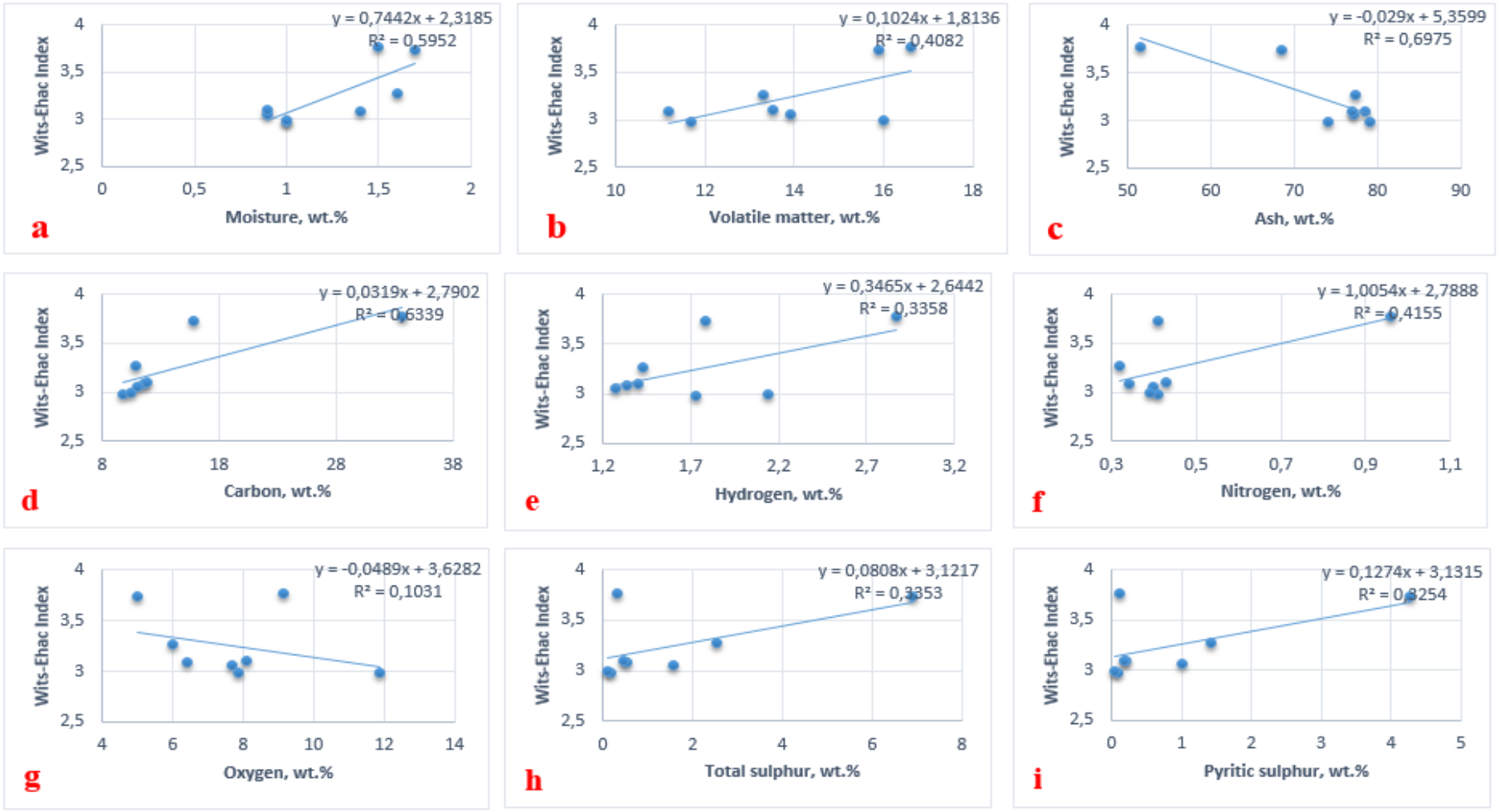

Figure 5.5 Influence of proximate and ultimate properties on sponatneous combustion liability for the coal-shale (a to i)

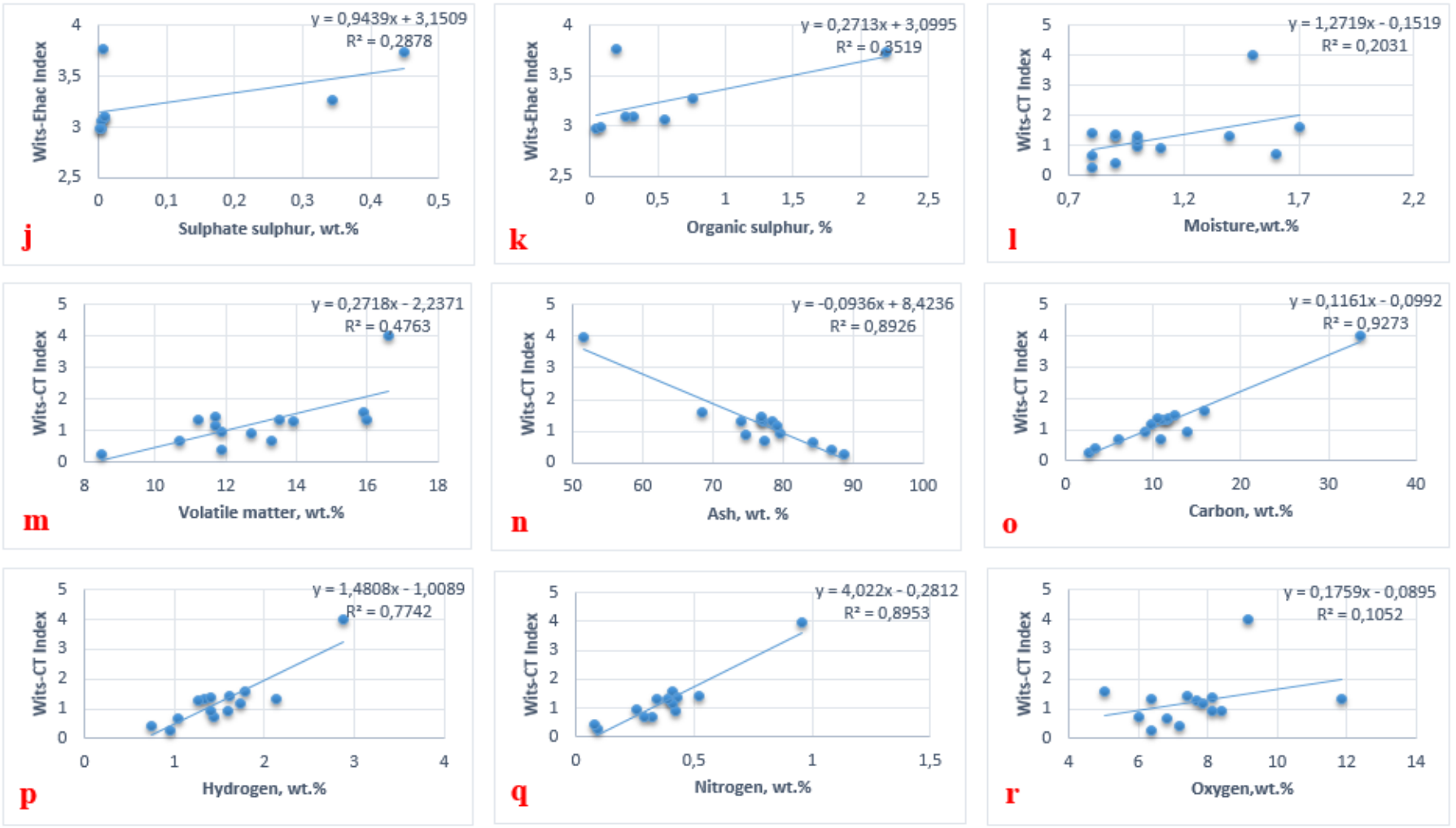

Figure 5.6 Influence of proximate and ultimate properties on sponatneous combustion liability for the coal-shale (j to r)

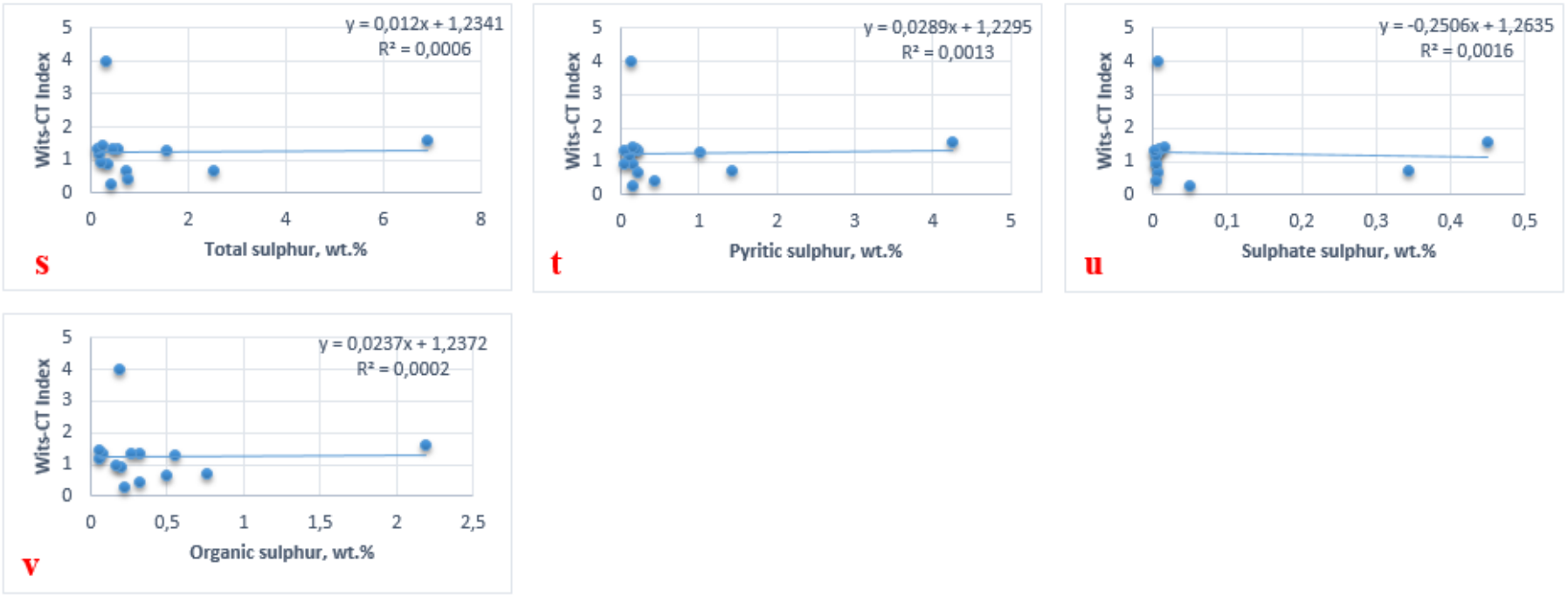

Figure 5.7 Influence of proximate and ultimate properties on sponatneous combustion liability for the coal-shale (s to v)

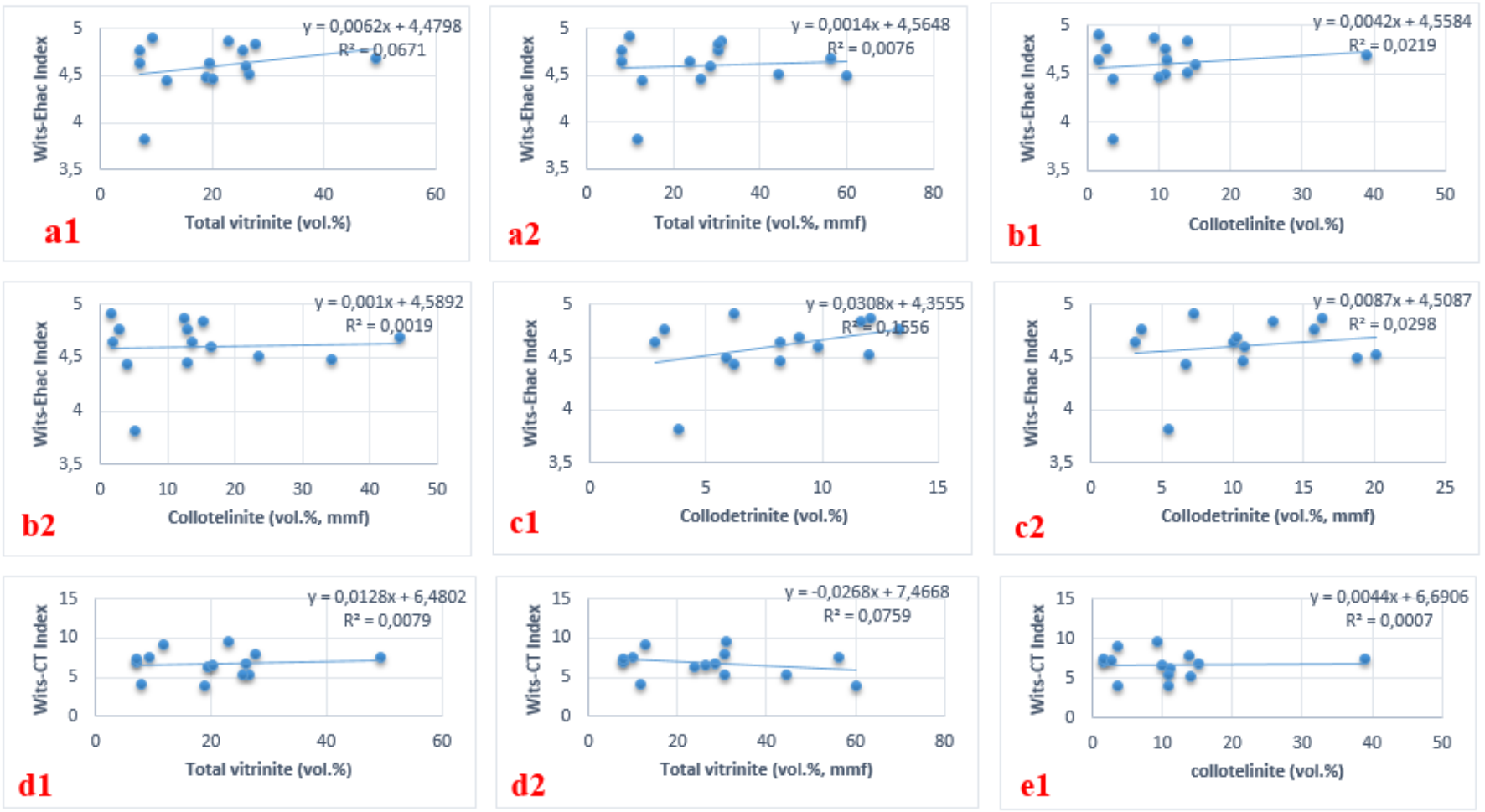

Figure 5.8 Influence of vitrinite maceral groups (vol% inclusing and exluding mienral matter) on spontaneous combustion liability for the coal (a1 to e1)

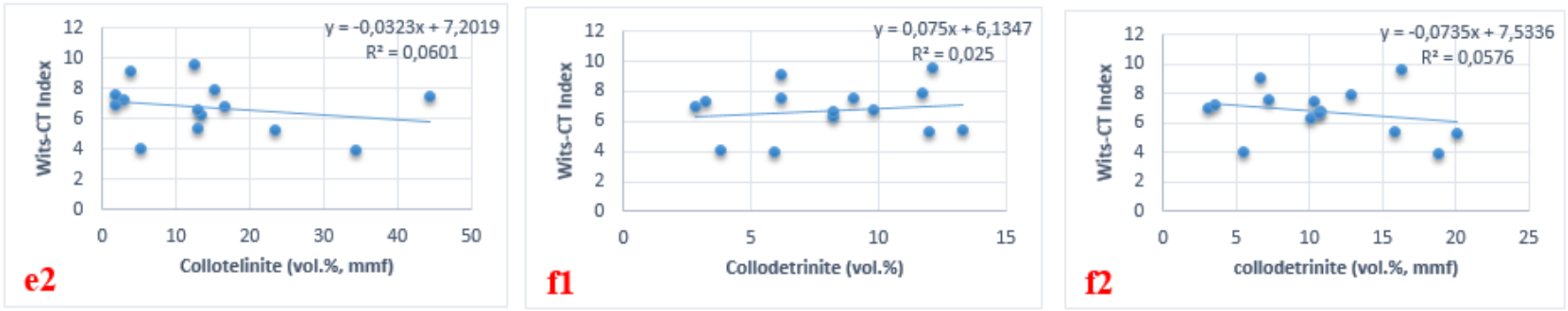

Figure 5.9 Influence of vitrinite maceral groupd (vol% inclusing and exluding mienral matter) on spontaneous combustion liability for the coal (e2 to f2)

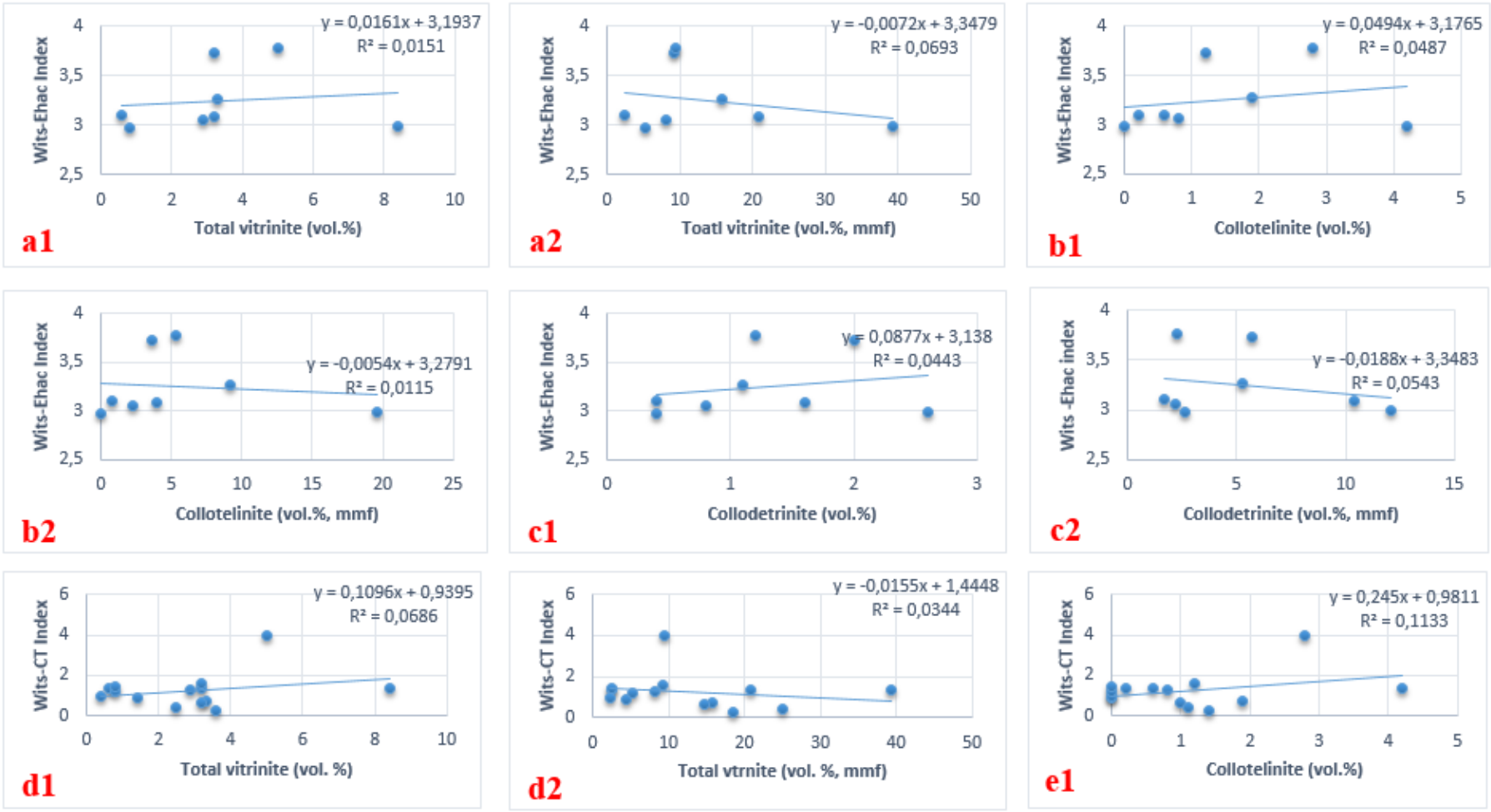

Figure 5.10 Influence of vitrinite maceral groups (vol% inclusing and exluding mienral matter) on spontaneous combustion liability for the coal-shale (a1 to e1)

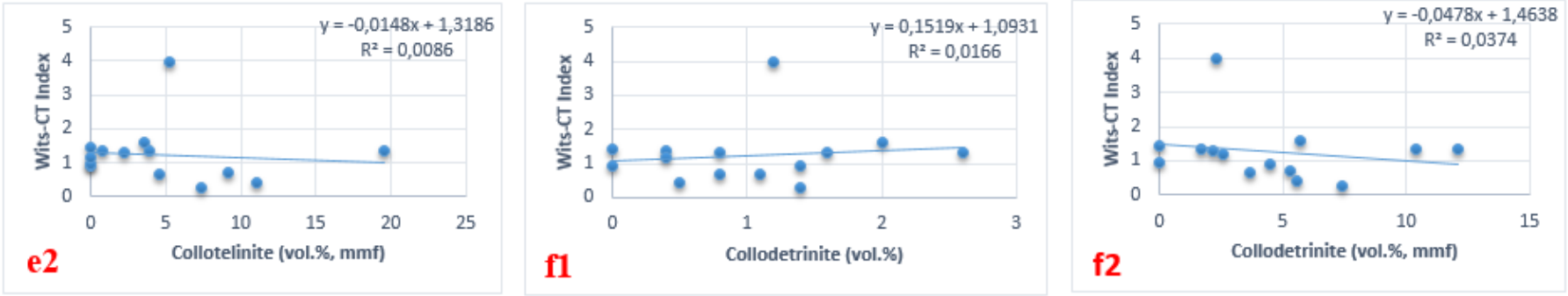

Figure 5.11 Influence of vitrinite maceral groups (vol% inclusing and exluding mienral matter) on spontaneous combustion for the coal-shale (e2 to f2)

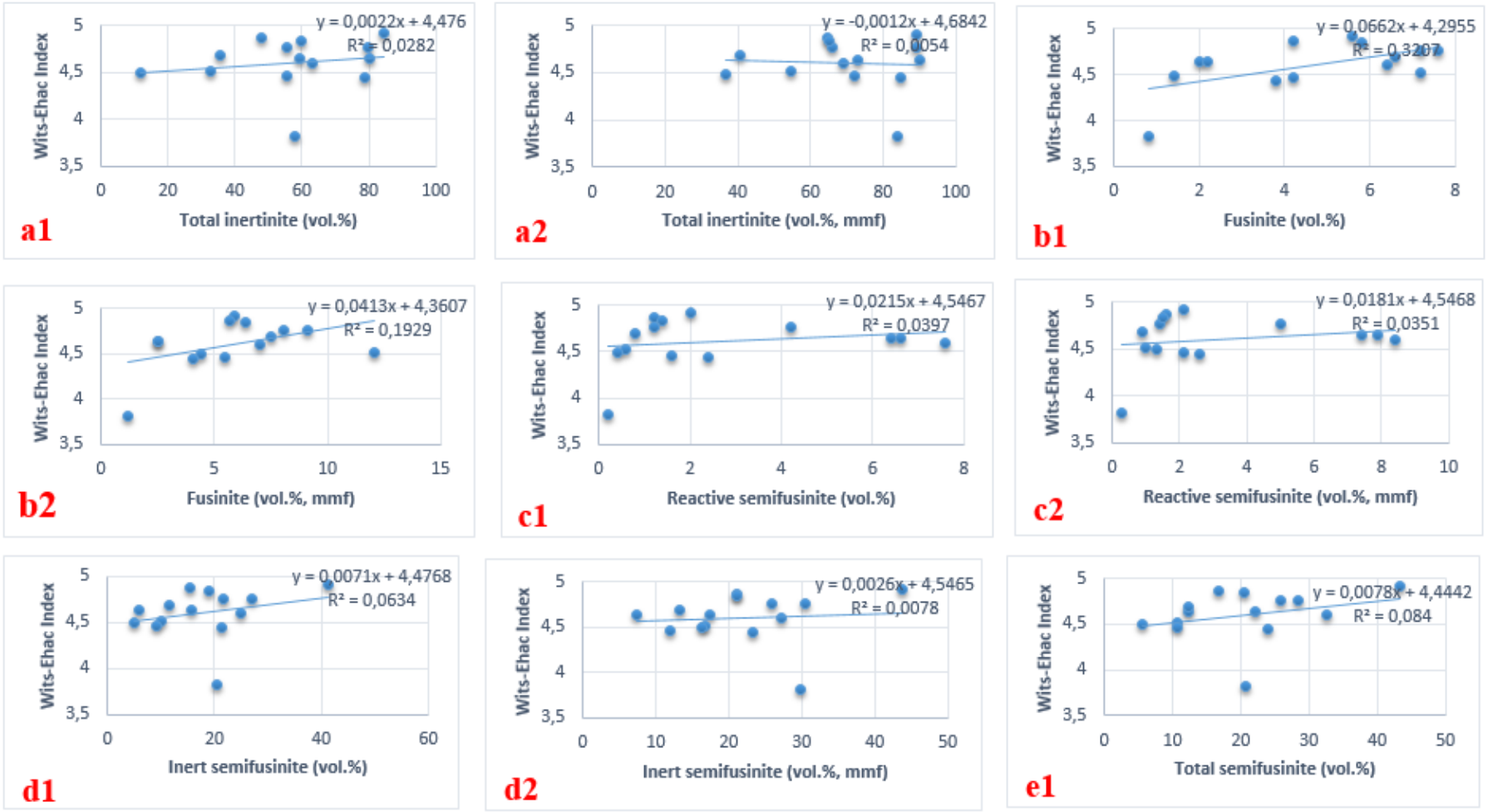

Figure 5.12 Influence of inertinite maceral groups(vol% inclusing and exluding mienral matter) on spontaneous combustion liability for the coal (a1 to e1)

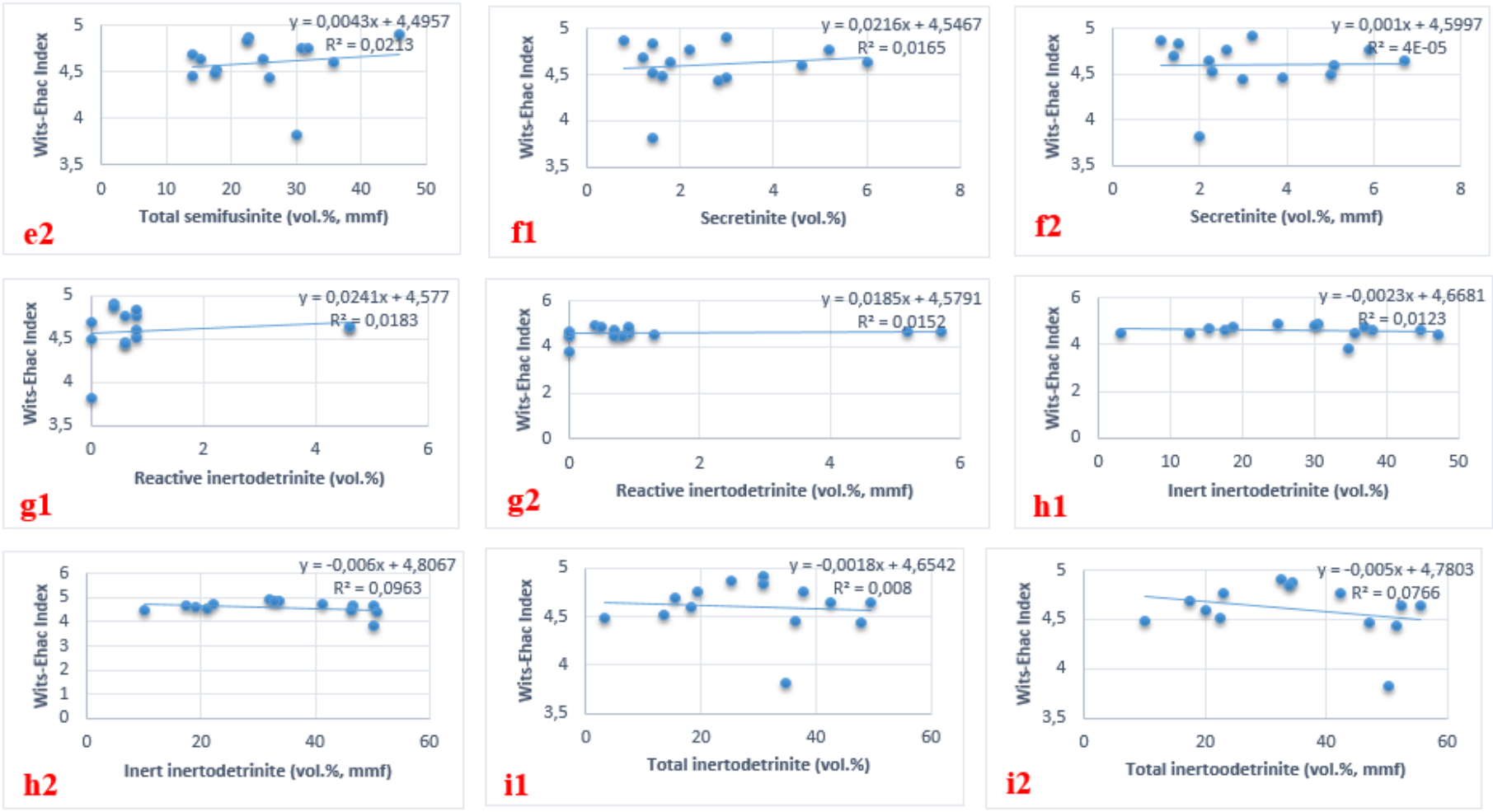

Figure 5.13 Influence of inertinite maceral groups (vol% inclusing and exluding mienral matter) on spontaneous combustion liability for the coal (e2 to i2)

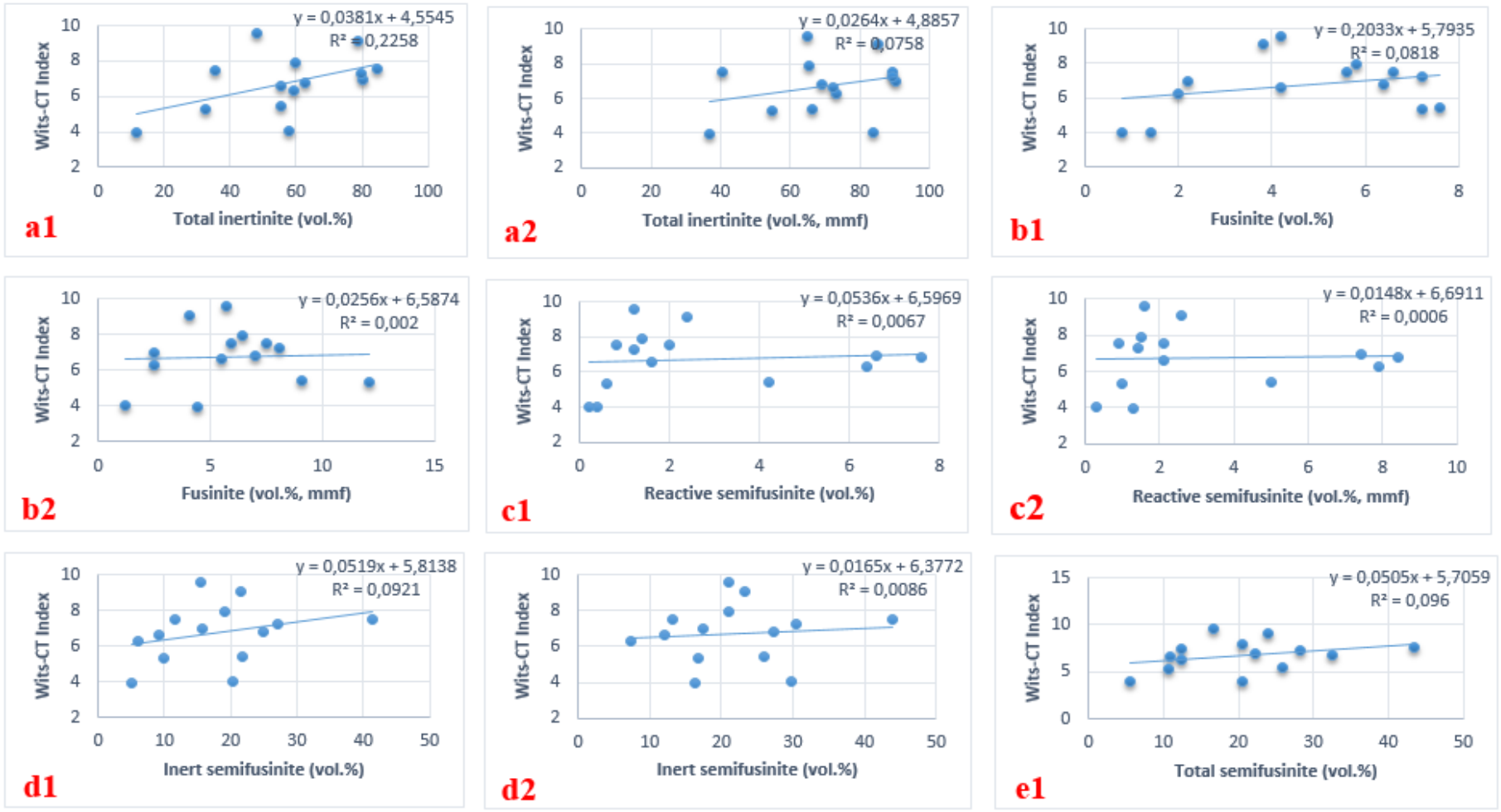

Figure 5.14 Influence of inertinite maceral groups (vol% inclusing and exluding mienral matter) on spontaneous combustion liability for the coal (a1 to e1)

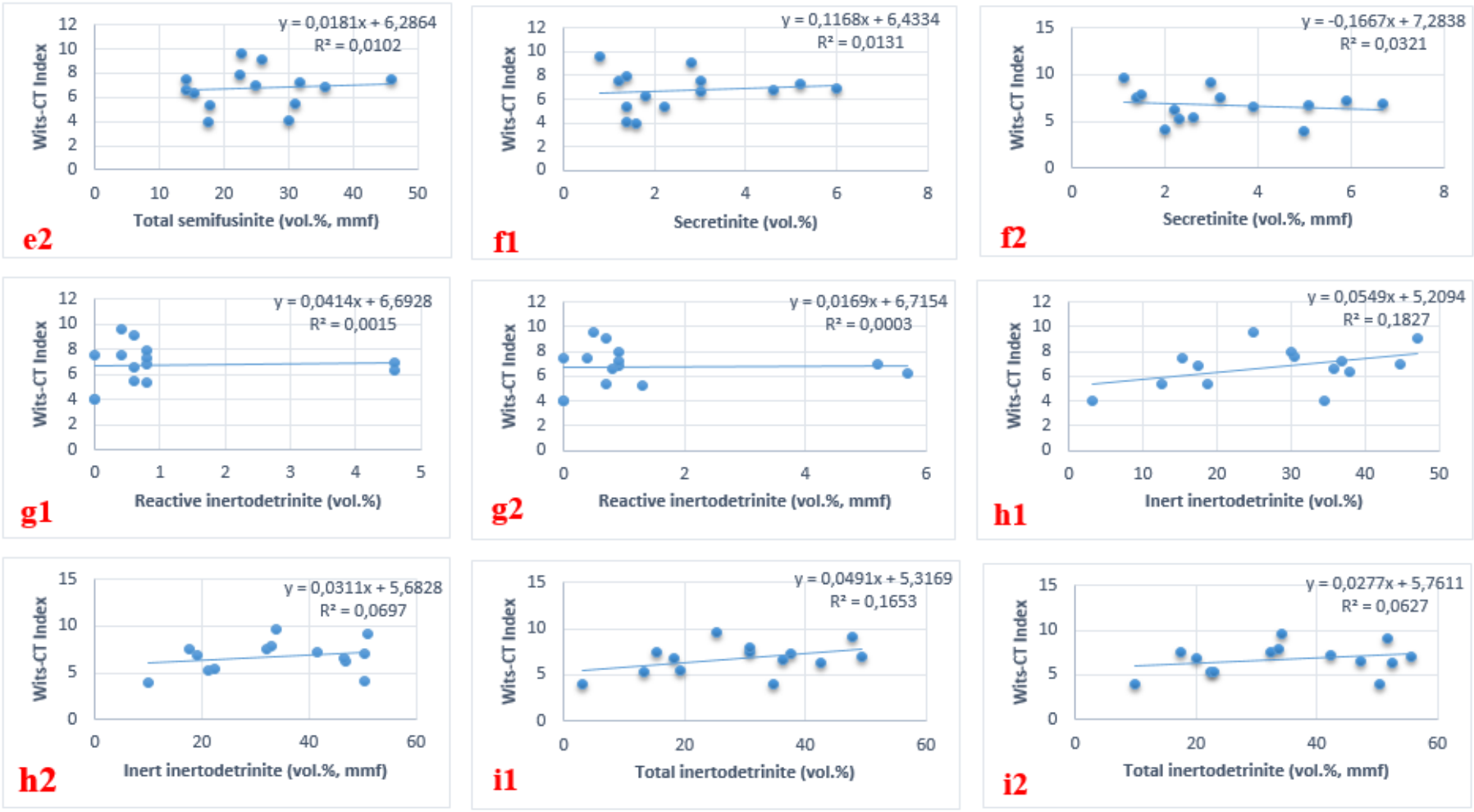

Figure 5.15 Influence of inertinite maceral groups (vol% inclusing and exluding mienral matter) on spontaneous combustion liability for the coal (e2 to i2)

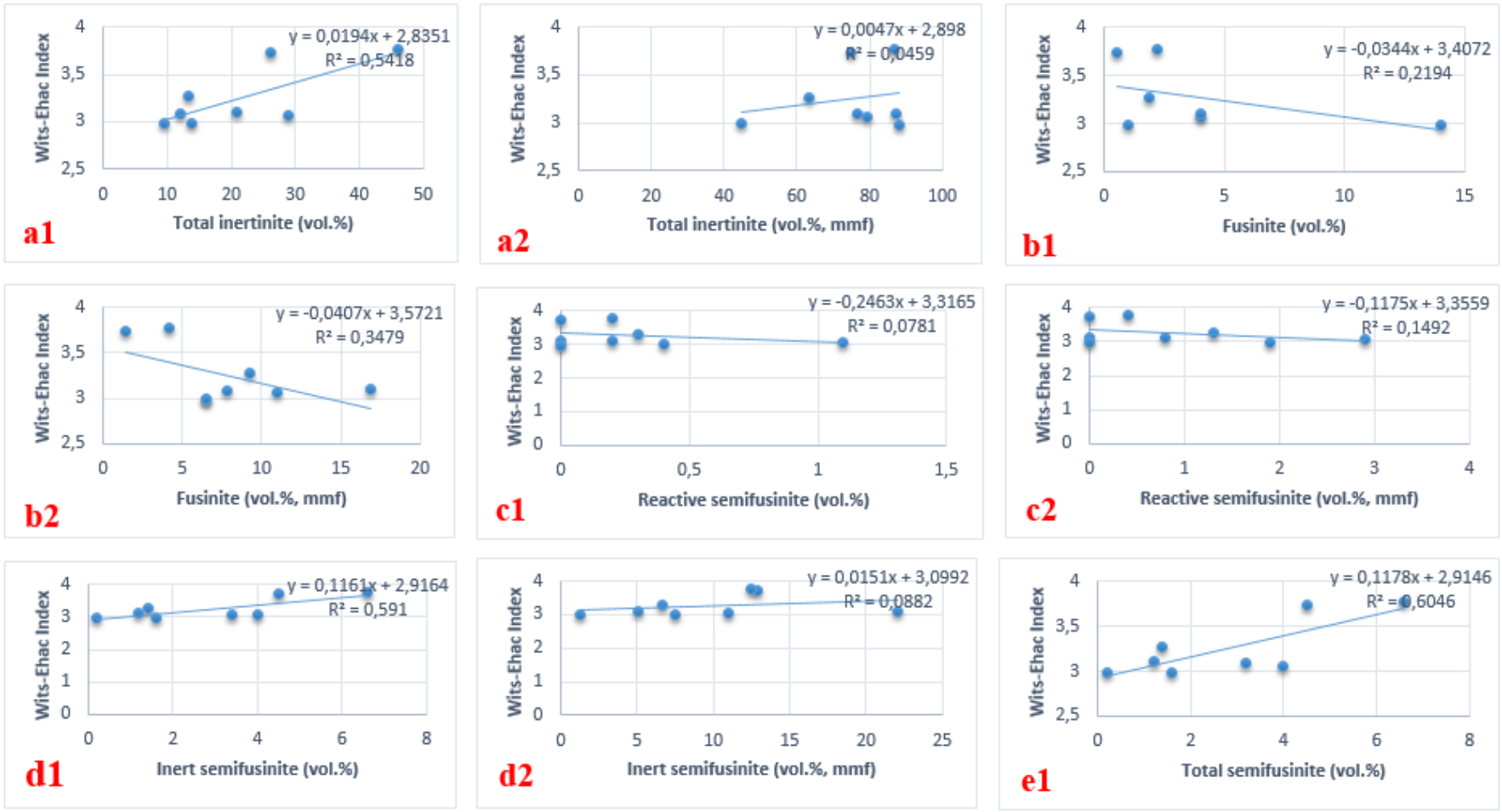

Figure 5.16 Influence of inertinite maceral groups (vol% inclusing and exluding mienral matter) on spontaneous combustion liability for the coal-shale (a1 to e1)

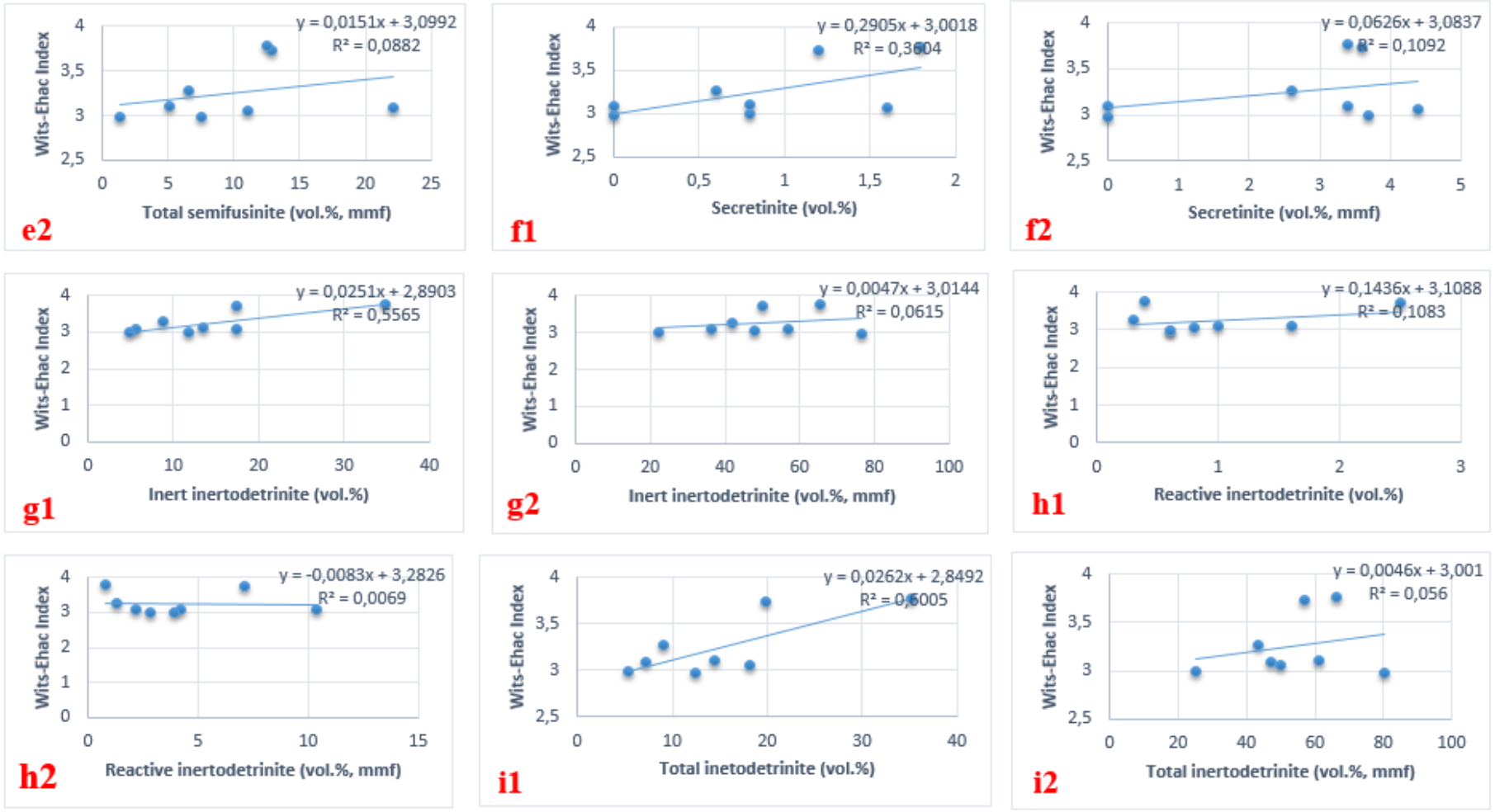

Figure 5.17 Influence of inertinite maceral groups (vol% inclusing and exluding mienral matter) on spontaneous combustion liability for the coal-shale (e2 to i2)

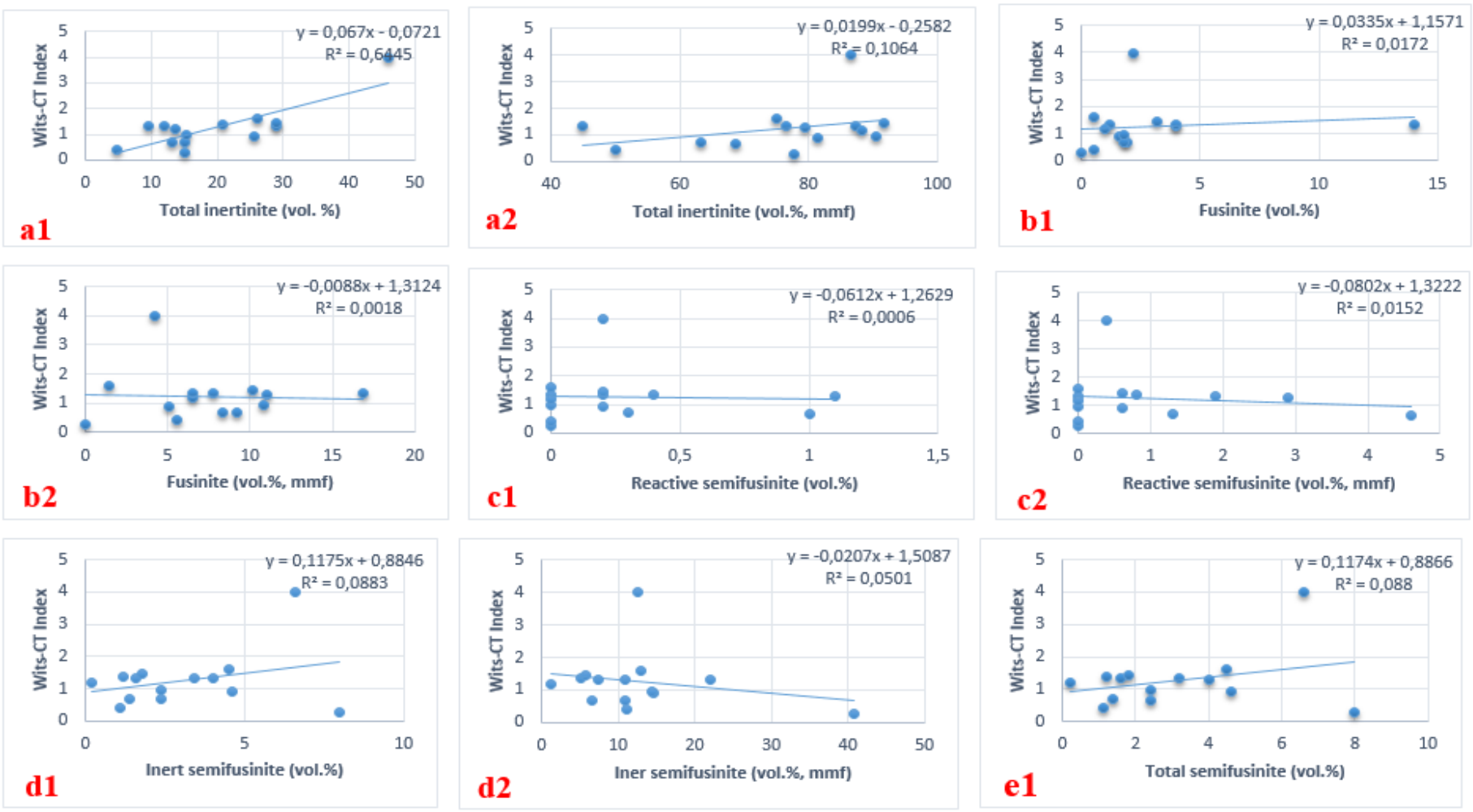

Figure 5.18 Influence of inertinite maceral groups (vol% inclusing and exluding mienral matter) on spontaneous combustion liability for the coal-shale (a1 to e1)

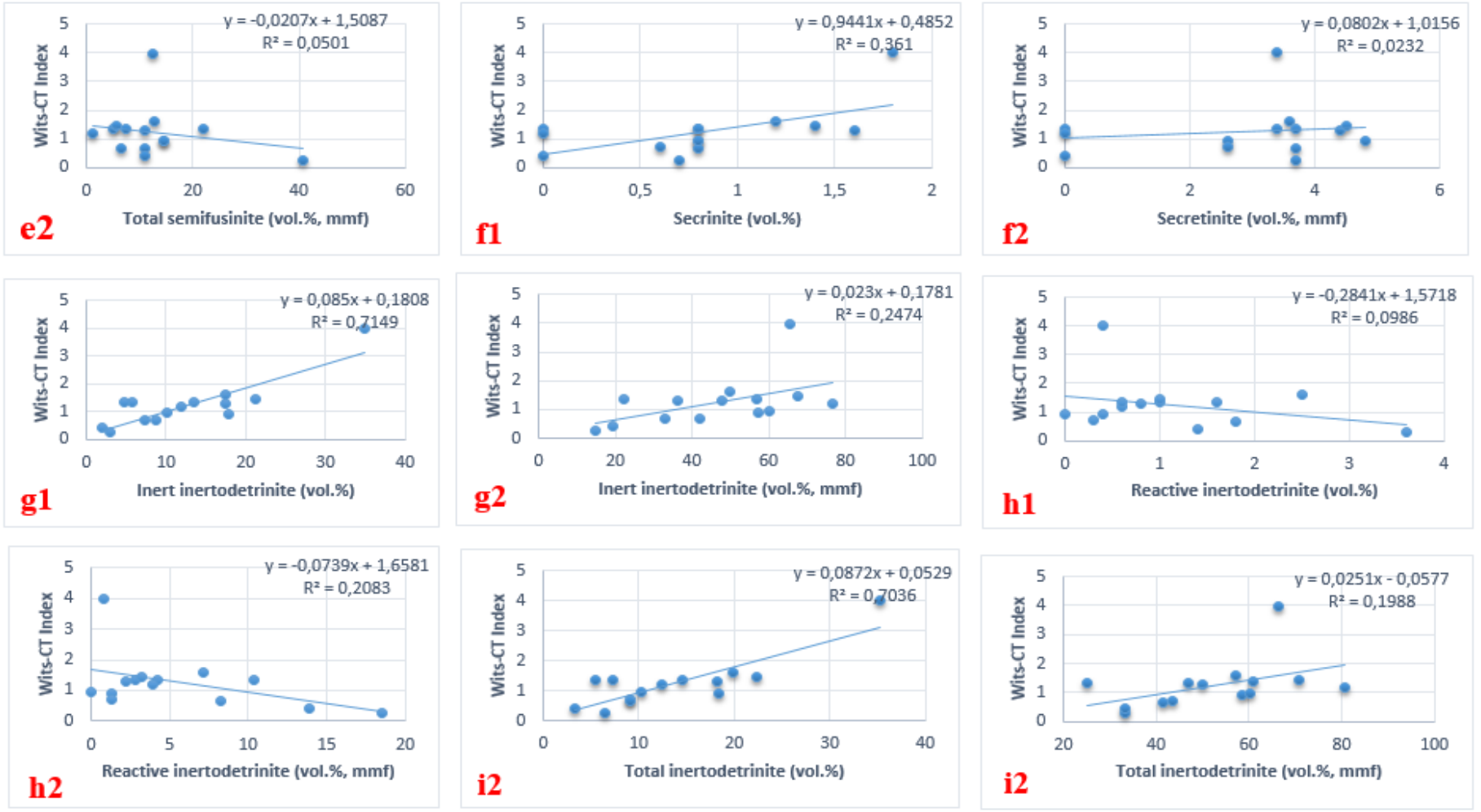

Figure 5.19 Influence of inertinite maceral groups (vol% inclusing and exluding mienral matter) on spontaneous combustion liability for the coal-shale (e2 to i2)

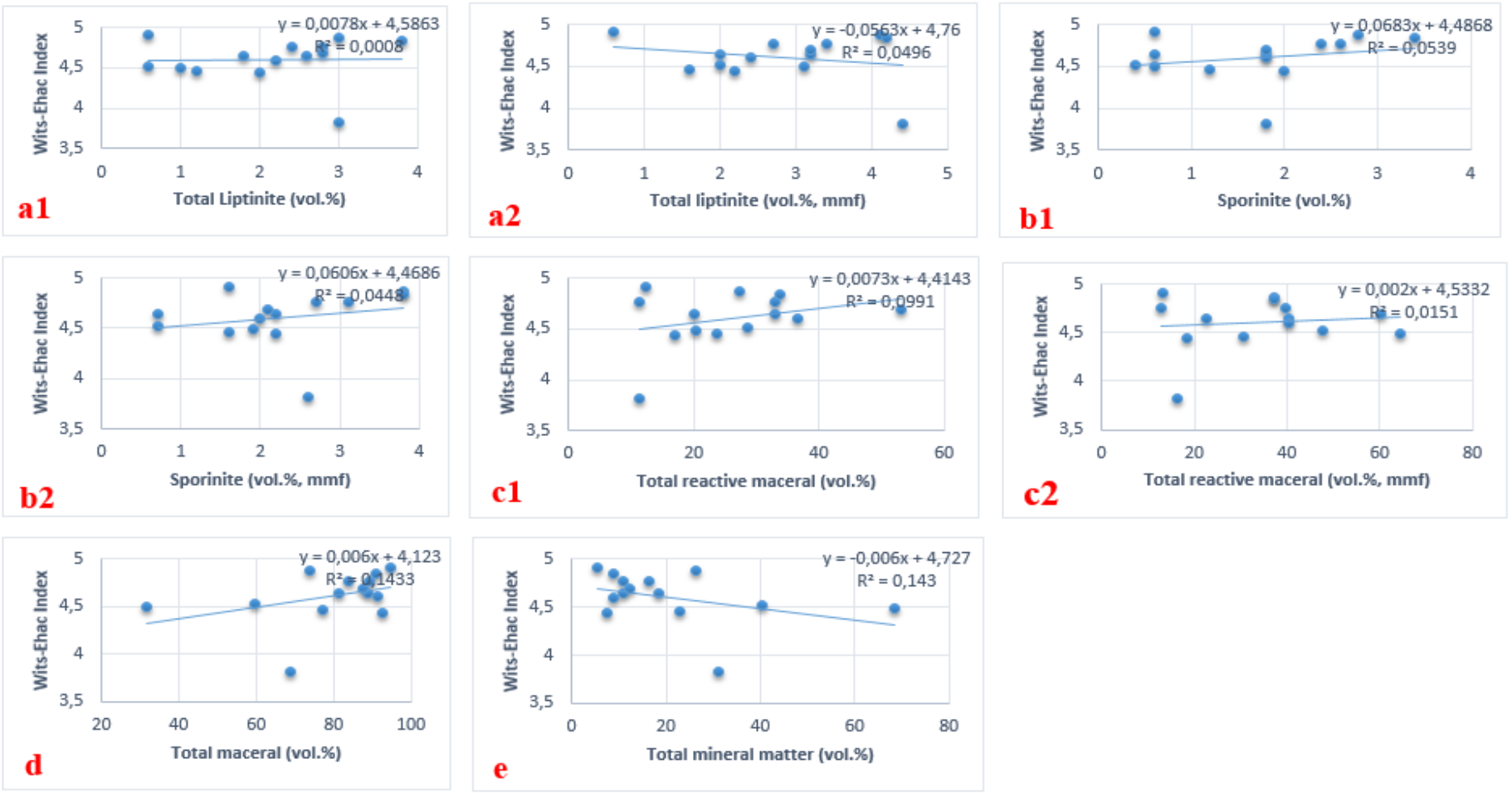

Figure 5.20 Influence of liptinite maceral groups and various petrographic properties (vol% inclusing and exluding mienral matter) on spontaneous combustion liability for the coal (a1 to e)

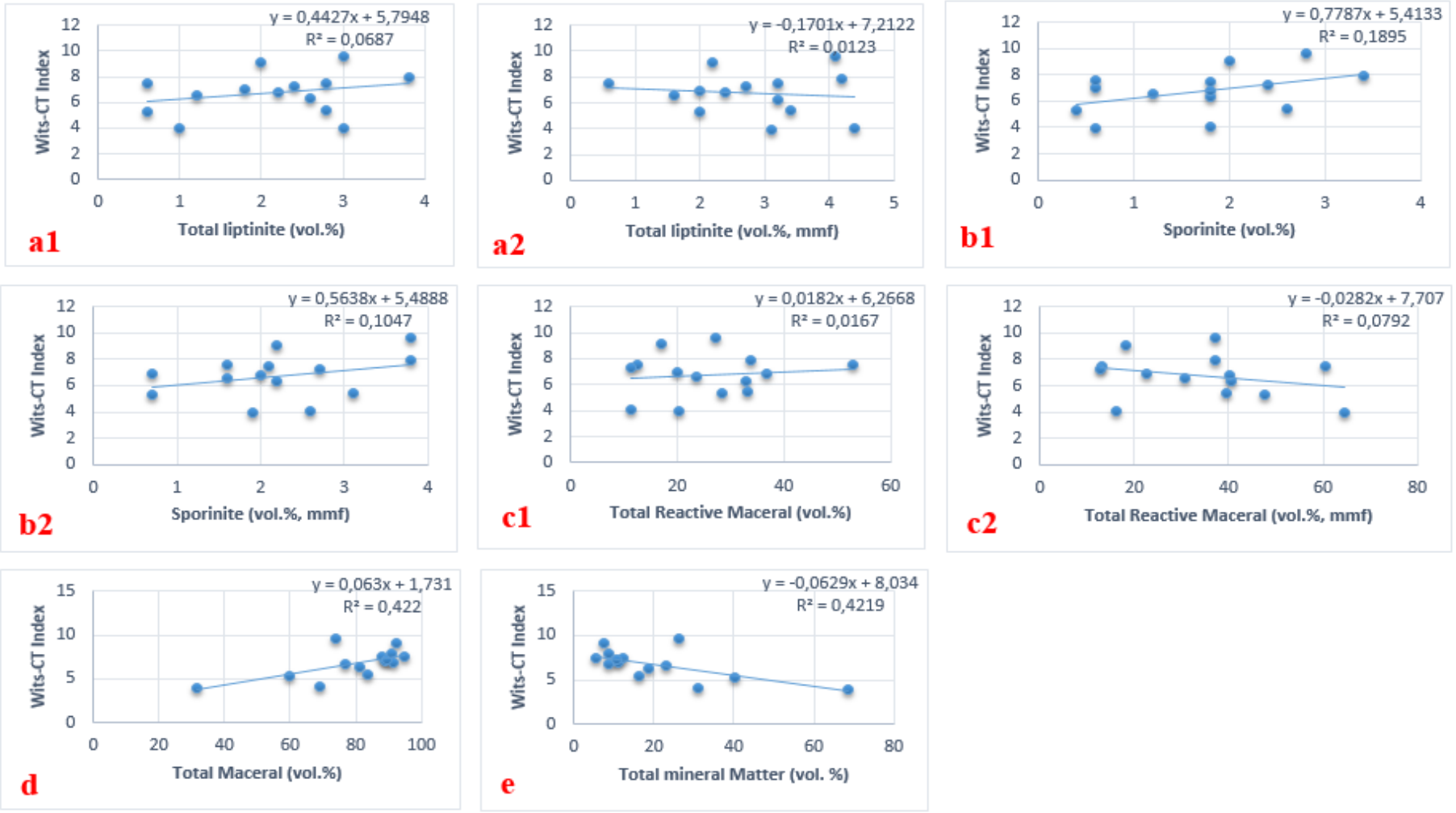

Figure 5.21 Influence of liptinite maceral groups and various petrographic properties (vol% inclusing and exluding mienral matter) on spontaneous combustion liability for the coal (a1 to e)

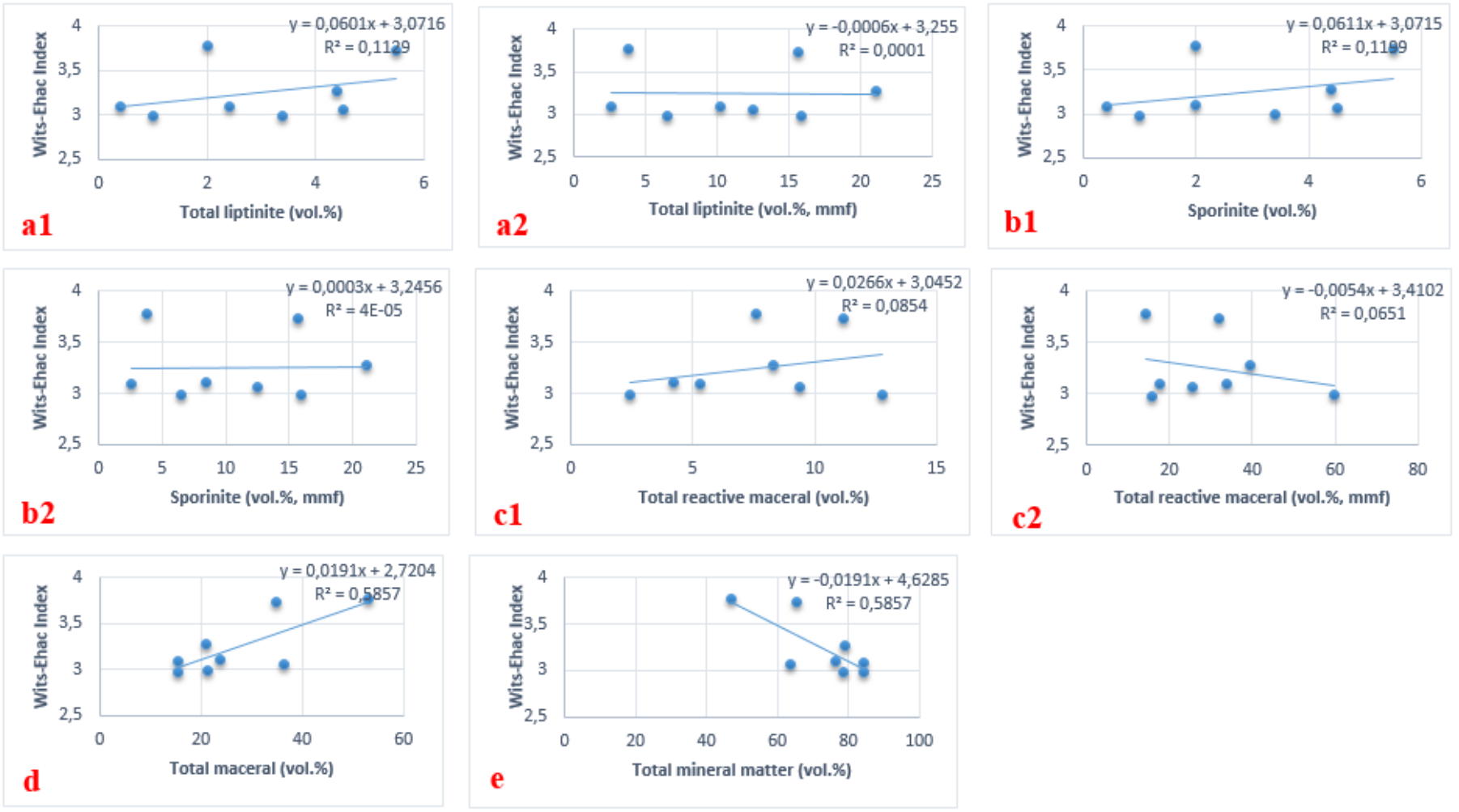

Figure 5.22 Influence of liptinite maceral groups and various petrographic properties (vol% inclusing and exluding mienral matter) on spontaneous combustion liability for the coal-shale (a1 to e)

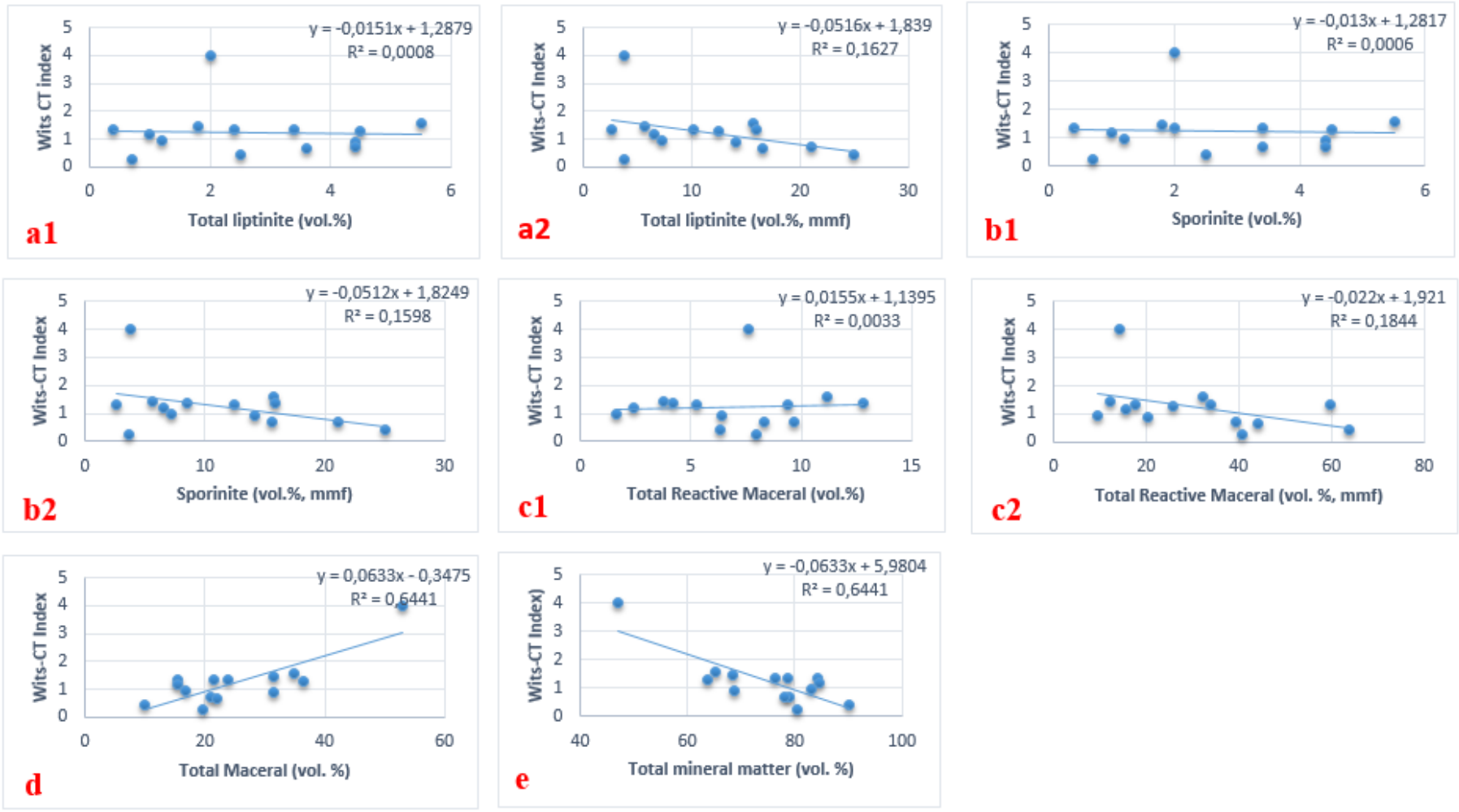

Figure 5.23 Influence of liptinite maceral groups and various petrographic properties (vol% inclusing and exluding mienral matter) on spontaneous combustion liability for the coal-shale (a1 to e)

5.3.2 Multiple regression analysis

The equations and calculations for the multiple regression analysis are discussed in Section 5.2.1 to 5.2.4 of this Chapter. The low correlation coefficients and R-squared values obtained from the Wits-Ehac Index and Wits-CT Index against some of the selected intrinsic properties necessitated the research towards multiple regression. Since it is well-known that spontaneous combustion occurs as a result of the cumulative effect of various factors, it was then decided to develop predictive models that comprised of various intrinsic properties to measure the spontaneous combustion liability of coal and coal-shale.

The study indicated that the use of a single variable equation as shown in Equation 5.1 obtained from the linear regression analysis to predict the spontaneous combustion liability of coal and coal-shale is unreliable as shown in Figures 5.2 to 5.23. This is because some properties show different trends with the spontaneous combustion liability indices. The lack of significant relationships between all the dependent and independent variables cannot allow separation of the main intrinsic properties influencing spontaneous combustion liability. It is more likely that the intrinsic properties have an influence on one another to some extent during low-temperature oxidation and this dependence of intrinsic properties may be used to determine the spontaneous combustion liability. This implies that the interrelationships of intrinsic properties could be one of the most favourable conditions for spontaneous combustion to occur. This necessitated the use of multiple regression to establish interrelationships between the intrinsic properties and spontaneous combustion liability results. The multiple regression analysis was used to derive the predictive models for spontaneous combustion liability of coal and coal-shale. The predictive models were derived by using twelve (12) independent variables (moisture, volatile matter, ash, carbon, hydrogen, nitrogen, sulphur, oxygen, vitrinite, liptinite, inertinite and mineral matter content). It was found that some of the variables were highly correlated with other independent variables. The best model which provides the highest R-squared values and correlation coefficient and lowest standard error of estimate was found through some multiple regression calculations as shown in Table 5.2.

Table 5.2 Predictive formulas derived by multiple regression analysis

Model No	Equations derived by multiple regression	RSV	R	SEE
All coal (A)	WE= 557.4939-6.6733M-0.0256VM-7.3322A-7.2405C-7.5779H-8.4405N-7.1169S-7.1643O+1.7441V+1.8823L+1.7419I+1.7577MM	0.882	0.939	0.336
All coal (B)	WC= 5780.205-43.1771M-0.3858VM-44.1651A-43.7848C -47.8671H-35.8312N-43.9474S-43.79O-13.7768V-12.3551L-13.8127I-13.6081MM	0.983	0.992	0.772
All coal-shale (C)	WE= -6159.64+61.9497M+1.0644VM+64.5252A+64.5374C +68.2129H+55.0262N+63.5985S+63.0291O-2.9441V-2.9167L-2.907I-2.9037MM	0.997	0.998	0.094
All coal-shale (D)	WC= 4302.218-37.9739M-0.5712VM-38.1227A-37.9071C -40.9502H-29.4017N-37.2958S-37.2423O-4.8458V-5.0309L-4.9672I-4.888MM	0.999	0.999	0.123

where: **M** is moisture (wt.%), **VM** is volatile matter (wt.%), **A** is ash (wt.%), **C** is carbon (wt.%), **H** is hydrogen (wt.%), **N** is nitrogen (wt.%), **S** is total sulphur (wt.%), **O** is calculated oxygen (wt.%), **V** is total vitrinite (vol%), **L** is total liptinite (vol.%), **I** is total inertinite (vol.%), MM is mineral matter (vol.%), **WE** is Wits-Ehac Index, **WC** is Wits-CT Index, **RSV** is R-squared values, **R** is correlation coefficient, and **SEE** is standard error of estimate.

A statistical standard form from the multiple regression results was selected to analyse and evaluate the influence of selected intrinsic properties on the spontaneous combustion liability of coal and coal-shale. This statistical approach indicated that the changes in spontaneous combustion liability index of coal and coal-shale may be due to an intrinsic property, while others are considered. The best prediction for spontaneous combustion liability of fourteen (14) coal and 14 coal-shale samples has been identified in the four models as shown in Table 5.2. The developed models show that high correlation coefficients and the standard error of estimates values have been reduced to acceptable limits. The multiple regression analysis indicates that the Wits-Ehac Index and intrinsic properties show significant correlation

coefficients of 0.939 and 0.998, R-squared values of 0.882 and 0.997, and low standard error of estimates of 0.336 and 0.094 for coal and coal-shale respectively. The Wits-CT Index with intrinsic properties shows significant correlation coefficients of 0.992 and 0.999, R-squared values of 0.983 and 0.999 and higher standard error of estimate of 0.772 and 0.123 for coal and coal-shale respectively. It was found that the models developed for the Wits-Ehac Index gives lower R-squared values, coefficient of correlations and standard error of estimates, while the Wits-CT Index shows higher R-squared values, correlation coefficients and standard error of estimates than the Wits-Ehac Index in both cases. However, considering the index with higher R-squared values and correlation coefficients, it was found that the Wits-CT Index is more suitable to predict the spontaneous combustion liability, while considering the index with a low standard error of estimates, the Wits-Ehac Index is also suitable. Hence, multiple regression analysis on the experimental data indicates that the two liability indices can be used to obtain reliable results for spontaneous combustion prediction. Establishing a correlation between the liability index and intrinsic properties for coal and coal-shale is essential for a better understanding of the role of coal-shale in spontaneous combustion. The models have shown that spontaneous combustion takes place due to the combined effect of different intrinsic factors.

5.3.3 Validation of predicted model results

The results of the actual and predicted Wits-Ehac and Wits-CT Indices for coal and coal-shale are presented in Table 5.3 and Table 5.4. The predicted spontaneous combustion liability indices (Wits-Ehac Index and Wits-CT Index) have been validated with the actual Wits-Ehac Index and the Wits-CT Index and the results are in-line with each other as shown in Table 5.9 and Table 5.10. Twenty-eight samples were evaluated in this thesis with an additional five (5) samples (4 coal samples- KCA, KCB, KCC and KCD; and one coal-shale-KSA) have confirmed the reliability of the developed models. The Wits-Ehac Index of coal-shale samples SC, SG, SH, SI, SJ and SM that could not be obtained by the actual Wits-Ehac Index due to their low reactivity were successfully obtained with the predicted models. The models present a high level of confidence as they produced results similar to the actual liability indices.

Table 5.3 Actual and predicted Wits-Ehac and Wits-CT Index for coal

Samples	Wits-Ehac Index		Wits-CT Index	
	Actual	Predicted	Actual	Predicted
CA	4.64	4.82	6.29	5.88
CB	4.64	4.67	6.96	6.90
CC	4.52	4.52	5.31	5.32
CD	4.60	4.64	6.80	6.71
CE	4.76	4.63	5.42	5.73
CF	4.49	4.49	3.97	3.98
CG	4.91	4.82	7.53	7.74
CH	4.69	4.70	7.51	7.49
CI	3.82	3.83	4.05	4.03
CJ	4.46	4.45	6.61	6.63
CK	4.44	4.62	9.10	8.98
CL	4.87	4.85	9.59	9.64
CM	4.76	4.63	7.27	7.47
CN	4.84	4.79	7.91	8.03
KCA	6.07	6.03	8.87	8.57
KCB	5.41	5.25	8.55	8.78
KCC	5.67	5.67	10.51	10.83
KCD	6.44	6.41	9.01	9.26

Table 5.4 Actual and predicted Wits-Ehac and Wits-CT Index for coal-shale

Samples	**Wits-Ehac Index**		**Wits-CT Index**	
	Actual	**Predicted**	**Actual**	**Predicted**
SA	3.09	3.07	1.33	1.37
SB	3.08	3.08	1.30	1.28
SC	-	2.56	0.91	0.97
SD	3.27	3.31	0.70	0.67
SE	3.73	3.72	1.60	1.61
SF	3.10	3.07	1.36	1.40
SG	-	2.49	0.67	0.63
SH	-	2.35	0.27	0.28
SI	-	2.58	0.95	0.88
SJ	-	2.63	0.42	0.42
SK	2.98	2.98	1.18	1.18
SL	2.99	2.99	1.34	1.35
SM	-	2.54	1.44	1.43
SN	3.77	3.79	3.99	3.97
KSA	3.49	3.50	4.40	4.38

5.3 Summary

The influence of the intrinsic properties of coal-shale in relation to coal could be a tool to predict spontaneous combustion risk in coal mines. This thesis analysed and established relationships between spontaneous combustion liability and intrinsic properties of coal and coal-shale. The results provide quantitative descriptions and showed the relationships between the dependence of the spontaneous combustion liability index and the intrinsic properties of coal and coal-shale. It was found that contents of moisture, ash, volatile matter, carbon, hydrogen, nitrogen, sulphate sulphur, inertinite maceral (fusinite), total maceral and petrographically observable mineral matter with strong linear relationships are the factors affecting spontaneous combustion liability of coal. For the coal-shale, contents of moisture,

ash, volatile matter, carbon, hydrogen, nitrogen, sulphur and its group (pyrite, sulphate and organic sulphur), total inertinite and its group (fusinite, total semifusinite, total inertodetrinite), total maceral and petrographically observable mineral matter with strong linear relationships are the main factors affecting spontaneous combustion liability of coal-shale. The study indicated that each intrinsic property has an influence on spontaneous combustion liability and their strength varies from one another based on the linear regression analysis. Furthermore, the two spontaneous combustion liability indices indicated linear relationships with some of the intrinsic factors based on the set criterion and thus, identifies the major intrinsic factors affecting spontaneous combustion liability. A definite positive or negative correlation coefficient exists between the intrinsic factors and spontaneous combustion liability index.

The liability indices and the intrinsic properties show significant correlation coefficients and less standard error for both coal and coal-shale using the multiple regression equations and calculations provided in Equation 5.2 to 5.12. The models developed shows that the Wits-Ehac Index gives a lower standard error of estimate, R-squared values and correlation coefficients than the Wits-CT Index for both coal and coal-shale. Multiple regression analysis on the experimental data indicated that the two liability indices can be used to obtain reliable results for spontaneous combustion prediction. Although, considering the index with a low standard error of estimates, the Wits-Ehac Index is suitable, while considering the index with higher correlation coefficients and R-squared values, the Wits-CT Index may provide more reliable results for spontaneous combustion prediction. The study indicated that statistical evaluation of data describes relationships between two variables or among several variables (spontaneous combustion liability indices and intrinsic properties). If the dependent and independent variables are continuous, then a correlation coefficient can be calculated as a measure of the strength of the relationship between them.

The study has developed acceptable and accurate predictive models as indicated by R-squared values, the coefficient of correlations and standard error of estimates. The models derived from the different samples produced strong correlation coefficients and a low standard error of estimates. The predicted spontaneous combustion liability indices (Wits-Ehac and Wits-CT Index) have been validated with the actual Wits-Ehac Index and the Wits-CT Index and the results are compared with each other. The use of the models may provide more accurate predictions for the spontaneous combustion liability of coal and coal-shale. The influence of intrinsic properties affecting spontaneous combustion liability of coal and coal-shale materials

may be evaluated more clearly by these models than by the linear regression. The results obtained from this study may be used as a reliable tool to support previous works on the role of intrinsic properties on the spontaneous combustion liability in coal mines.

The multiple regression analysis indicates that the Wits-Ehac Index and intrinsic properties show significant correlation coefficients of 0.939 and 0.998, R-squared values of 0.882 and 0.997, and low standard error of estimates of 0.336 and 0.094 for coal and coal-shale respectively. The Wits-CT Index with intrinsic properties shows significant correlation coefficients of 0.992 and 0.999, R-squared values of 0.983 and 0.999 and higher standard error of estimate of 0.772 and 0.123 for coal and coal-shale respectively. This section provides reliable information and better understanding on comparative analysis of individual intrinsic factors affecting spontaneous combustion liability of coal and coal-shale by using the statistical equations and calculations provided in this study. Multivariable linear regression was used to establish relationships between intrinsic properties and spontaneous combustion liabilities and the development of predictive models for the evaluation and analysis of the influence of intrinsic properties on spontaneous combustion liability of coal and coal-shale. The study indicated that the proper performance of the regression analysis requires a number of important factors that should be considered and tested in order to develop a predictive model. The effects of four (4) selected chemical inhibitors on spontaneous combustion liability of coal and coal-shale are discussed in the next Chapter.

6 EFFECTS OF CHEMICAL INHIBITORS ON SPONTANEOUS COMBUSTION LIABILITY

6.1 Introduction

The spontaneous combustion affects valuable and non-renewal resources, increases mining operating costs, and creates both health and environmental hazards due to the release of toxic gases. Therefore, minimizing the event of spontaneous combustion is desirable. This section describes the effects of four (4) chemical inhibitors such as calcium carbonate ($CaCO_3$), magnesium chloride ($MgCl_2$), ammonium dihydrogen phosphate ($NH_4H_2PO_4$) and sodium metasilicate pentahydrate ($Na_2SiO_3.5H_2O$) on spontaneous combustion liability of coal and coal-shale. These chemical inhibitors were chosen to prove the concept that inhibitors can be used to minimize spontaneous combustion liability. Hence, only four inhibitors were tested. By considering the logistics and the cost of the inhibitors, the Wits-Ehac Index instead of the Wits-CT Index was used to evaluate the effectiveness of the inhibitors, as it uses small amount of coal samples. A cost-performance evaluation of the inhibitor application is described which would be of much benefit to the industry. The performance evaluation (PE) and cost-performance evaluation (CPE) of the inhibitors were evaluated on a laboratory-scale test. The optimal concentration of various chemical inhibitors was determined and the self-heating potential of coal and coal-shale blended with the additive based on the spontaneous combustion liability data was evaluated to confirm the inhibiting effect. Thirty-five (35) tests were conducted on both untreated and treated samples.

6.2 Results

The results of the XPT, Stage II slope, Wits-Ehac Index and PE of seven (7) untreated and treated samples are provided in Table 6.1. The results and the discussions in this section are based on the experimental tests conducted to evaluate the effects of inhibitors on the spontaneous combustion liability of coal and coal-shale. The graphs of the XPT for the treated and untreated coal and coal-shale samples are shown in Figures 6.1 to 6.3, while the graphs of the liability index are shown in Figures 6.4 to 6.8. The PE is used to compute the percentage of reduction or increment in the spontaneous combustion liability index as a result of using a particular inhibitor and the PE for each inhibitor can be computed as follows:

$$PE = \frac{UWE - TWE}{UWE} * 100\% \quad (6.1)$$

where, **UWE** is Wits-Ehac Index for untreated sample and **TWE** is Wits-Ehac Index for treated sample.

Table 6.1 Performance Evaluation of selected inhibitors based on the results obtained from XPT, Stage II slope and Wits-Ehac Index

Samples	XPT	Stage II slope	WE	PE (%)
CL Untreated	148.6	1.4487	4.87	
CL + $MgCl_2$	181.1	1.188	3.28	32.5
CL + $CaCO_3$	131.8	1.202	4.56	6.37
CL + $NH_4H_2PO_4$	140.9	1.2005	4.26	12.53
CL + $Na_2SiO_3.5H_2O$	178.9	1.5636	4.37	10.27
CN Untreated	139	1.346	4.84	
CN + $MgCl_2$	183.1	1.2487	3.41	29.5
CN + $CaCO_3$	126.5	1.1587	4.58	5.37
CN + $NH_4H_2PO_4$	129.3	1.1172	4.32	10.74
CN + $Na_2SiO_3.5H_2O$	176.6	1.5859	4.49	7.23
CG Untreated	140.7	1.3815	4.91	
CG + $MgCl_2$	183.1	1.0437	2.85	41.95
CG + $CaCO_3$	148.5	1.3009	4.38	10.8
CG + $NH_4H_2PO_4$	150.3	1.3226	4.4	10.39
CG + $Na_2SiO_3.5H_2O$	180.9	1.5594	4.31	12.21
SN Untreated	161.8	1.2245	3.77	
SN + $MgCl_2$	163.7	0.789	2.41	36.1
SN + $CaCO_3$	135.2	0.8599	3.18	15
SN + $NH_4H_2PO_4$	134.6	0.7538	2.8	25.73
SN + $Na_2SiO_3.5H_2O$	174.2	1.3936	4	-6.1

Table 6.2 Performance Evaluation of selected inhibitors based on the results obtained from XPT, Stage II slope and Wits-Ehac Index

Samples	XPT	Stage II slope	WE	PE (%)
KCA Untreated	136.4	1.6559	6.07	
KCA + $MgCl_2$	176.6	1.0313	2.92	51.9
KCA + $CaCO_3$	149.1	1.4522	4.87	19.76
KCA + $NH_4H_2PO_4$	143.2	1.2716	4.44	26.85
KCA + $Na_2SiO_3.5H_2O$	179.2	1.7884	4.99	17.79
KCC Untreated	137.1	1.5547	5.67	
KCC + $MgCl_2$	160.1	1.1015	3.44	39.3
KCC + $CaCO_3$	132.5	1.3886	5.24	7.58
KCC + $NH_4H_2PO_4$	131.1	1.1851	4.58	19.22
KCC + $Na_2SiO_3.5H_2O$	177.3	2.3829	6.72	-18.52
KCD Untreated	135.6	1.7465	6.44	
KCD + $MgCl_2$	186.7	1.3122	3.52	45.3
KCD + $CaCO_3$	134.7	1.3712	5.09	20.96
KCD + $NH_4H_2PO_4$	129.6	1.2131	4.68	27.33
KCD + $Na_2SiO_3.5H_2O$	168.5	2.0961	6.22	3.42

6.3 Discussion

The application of four (4) chemical inhibitors to minimize self-heating and spontaneous combustion liability of coal and coal-shale highly prone to spontaneous combustion was investigated experimentally. The effect these inhibitors had on spontaneous combustion liability was investigated with the aim to develop a guideline in search of suitable inhibitors for minimizing spontaneous combustion and to prove the concept that inhibitors can minimize spontaneous combustion. A total of seven samples (three (3) coal samples and one coal-shale sample from this study, and three (3) coal samples recently reported with high spontaneous combustion liability indices) were examined with the chemical inhibitors. The PE of selected inhibitors based on the results obtained from XPT, Stage II slope and Wits-Ehac Index both for the untreated and treated samples are shown in Table 6.1 and Table 6.2. The Differential analysis thermogram for untreated and treated sample are illustrated in Figures 6.1 to 6.3, while Figures 6.4 to 6.8 shows the Wits-Ehac Index of each sample with respect to the inhibitors.

The liability index of treated coal with $MgCl_2$ decreased from 4.87 to 3.28 for coal sample CL, 4.84 to 3.41 for coal sample CN, 4.91 to 2.85 for coal sample CG, 3.77 to 2.41 for coal-shale

sample SN, 6.44 to 3.52 for coal sample KCD, 5.67 to 3.44 for coal sample KCC, and 6.07 to 2.92 for coal sample KCA respectively. The Wits-Ehac Index of treated samples with $CaCO_3$ decreased from 4.87 to 4.56 for coal sample CL, 4.84 to 4.58 for coal sample CN, 4.91 to 4.38 for coal sample CG, 3.77 to 3.18 for coal-shale sample SN, 6.44 to 5.09 for coal sample KCD, 5.67 to 5.24 for coal sample KCC, and 6.07 to 4.87 for coal sample KCA respectively. The Wits-Ehac Index of treated coal with $NH_4H_2PO_4$ decreased from 4.87 to 4.26 for coal sample CL, 4.84 to 4.32 for coal sample CN, 4.91 to 4.40 for coal sample CG, 3.77 to 2.80 for coal-shale sample SN, 6.44 to 4.68 for coal sample KCD, 5.67 to 4.58 for coal sample KCC, and 6.07 to 4.44 for coal sample KCA respectively. The Wits-Ehac Index of treated coal with $Na_2SiO_3.5H_2O$ decreased from 4.87 to 4.37 for coal sample CL, 4.84 to 4.49 for coal sample CN, 4.91 to 4.31 for coal sample CG, 6.44 to 6.22 for coal sample KCD, increased from 3.77 to 4.0 for coal-shale sample SN, increased from 5.67 to 6.72 for coal sample KCC, and increased from 6.07 to 4.99 for coal sample KCA respectively.

The XPT of treated coal with $MgCl_2$ increased from 148.6^oC to 181.1^oC for coal sample CL, 139^oC to 183.1^oC for coal sample CN, 140.7^oC to 183.1^oC for coal sample CG, 161.8^oC to 163.7^oC for coal-shale sample SN, 136.4^oC to 176.6^oC for coal sample KCA, 137.1^oC to 160.1^oC for coal sample KCC and 135.6^oC to 186.7^oC for coal sample KCD respectively. The XPT of treated coal with $CaCO_3$ decreases from 148.6^oC to 131.8^oC for coal sample CL, from 139^oC to 126.5^oC for coal sample CN, increases from 140.7^oC to 148.5^oC for coal sample CG, decreases from 161.8^oC to 135.2^oC for coal-shale sample SN, increases from136.4^oC to 149.1^oC for coal sample KCA, decreases from 137.1^oC to 132.5^oC for coal sample KCC and decreases from 135.6^oC to 134.7^oC for coal sample KCD respectively. The XPT of treated coal with $NH_4H_2PO_4$ decreases from 148.6^oC to 140.9^oC for coal sample CL, from 139^oC to 129.3^oC for sample CN, increases from 140.7^oC to 150.3^oC for coal sample CG, decreases from 161.8^oC to 134.6^oC for coal-shale sample SN, increases from136.4^oC to 143.2^oC for coal sample KCA, decreases from 137.1^oC to 131.1^oC for coal sample KCC and decreases from 135.6^oC to 129.6^oC for coal sample KCD respectively. The XPT of treated coal with $Na_2SiO_3.5H_2O_4$ increases from 148.6^oC to 178.9^oC for coal sample CL, 139^oC to 176.6^oC for coal sample CN, 140.7^oC to 180.9^oC for coal sample CG, 161.8^oC to 174.2^oC for coal-shale sample SN, 136.4^oC to 179.2^oC for coal sample KCA, 137.1^oC to 177.3^oC for coal sample KCC and 135.6^oC to 168.5^oC for coal sample KCD respectively. The study shows that the XPT values of the treated samples varies from one sample to the other.

The Stage II slope of the sample decreases from 1.4486 to 1.1880 for sample CL treated with $MgCl_2$, decreases from 1.4486 to 1.2020 for sample CL treated with $CaCO_3$, decreases from 1.4486 to 1.2005 for sample CL treated with $NH_4H_2PO_4$ and increases from 1.4486 to 1.5636 for sample CL treated with $Na_2SiO_3.5H_2O$. The Stage II slope of the sample decreases from 1.3460 to 1.2487 for sample CN treated with $MgCl_2$, decreases from 1.3460 to 1.1587 for sample CN treated with $CaCO_3$, decreases from 1.3460 to 1.1172 for sample CN treated with $NH_4H_2PO_4$ and increases from 1.3460 to 1.5859 for sample CN treated with $Na_2SiO_3.5H_2O$. The Stage II slope of the sample decreases from 1.3815 to 1.0437 for sample CG treated with $MgCl_2$, decreases from 1.3815 to 1.3009 for sample CG treated with $CaCO_3$, decreases from 1.3815 to 1.3226 for sample CG treated with $NH_4H_2PO_4$ and increases from 1.3815 to 1.5594 for sample CG treated with $Na_2SiO_3.5H_2O$. The Stage II slope of the sample decreases from 1.2245 to 0.7890 for sample SN treated with $MgCl_2$, decreases from 1.2245 to 0.8599 for sample SN treated with $CaCO_3$, decreases from 1.2245 to 0.7538 for sample SN treated with $NH_4H_2PO_4$ and increases from 1.2245 to 1.3936 for sample SN treated with $Na_2SiO_3.5H_2O$. The Stage II slope of the sample decreases from 1.6559 to 1.0313 for sample KCA treated with $MgCl_2$, decreases from 1.6559 to 1.4522 for sample KCA treated with $CaCO_3$, decreases from 1.6559 to 1.2716 for sample KCA treated with $NH_4H_2PO_4$ and increases from 1.6559 to 1.7884 for sample KCA treated with $Na_2SiO_3.5H_2O$. The Stage II slope of the sample decreases from 1.5547 to 1.1015 for sample KCC treated with $MgCl_2$, decreases from 1.5547 to 1.3886 for sample KCC treated with $CaCO_3$, decreases from 1.5547 to 1.1851 for sample KCC treated with $NH_4H_2PO_4$ and increases from 1.5547 to 2.3829 for sample KCC treated with $Na_2SiO_3.5H_2O$. The Stage II slope of the sample decreases from 1.7465 to 1.3122 for sample KCD treated with $MgCl_2$, decreases from 1.7465 to 1.3712 for sample KCD treated with $CaCO_3$, decreases from 1.7465 to 1.2131 for sample KCD treated with $NH_4H_2PO_4$ and increases from 1.5547 to 2.0961 for sample KCD treated with $Na_2SiO_3.5H_2O$. The study shows that the Stage II slope values of the treated samples varies from one sample to the other.

The results obtained from the XPT, Stage II slope and the Wits-Ehac Index values indicated that the characteristics of coal and coal-shale samples treated with inhibitor varies from one to the other. The XPT and Stage II slope values obtained from samples treated with $MgCl_2$ show that as XPT increases, the Stage II slope and Wits-Ehac Index decreases.

The data for the PE of each inhibitor on spontaneous combustion liability was obtained from the spontaneous combustion liability tests of treated coal and coal-shale samples as provided

in Table 6.1. The study indicated that the spontaneous combustion liability index decreases due to the blend of the samples with the inhibitors, such that the treated samples show a consistent decrease in the liability indices except for samples SN and KCC blended with $Na_2SiO_3.5H_2O$. The outcome of the test indicates that the blend of the inhibitors on coal and coal-shale decreases the spontaneous combustion liability and hence, as predicted, the presence of the $MgCl_2$ inhibitor gives a better performance of the thermal stability of the samples compared with $CaCO_3$, $NH_4H_2PO_4$ and $Na_2SiO_3.5H_2O$ as shown in Table 6.1 and Figures 6.1 to 6.8. The spontaneous combustion liability indices of the treated samples were lower than that of the untreated samples. This could be that the inhibitors created an oxidative barrier during low temperature oxidation and preventing moisture and high volatile organics from escaping the internal surface of the coal and coal-shale. $NH_4H_2PO_4$ shows better improvement in the inhibiting effect on the samples than $Na_2SiO_3.5H_2O$ and $CaCO_3$.

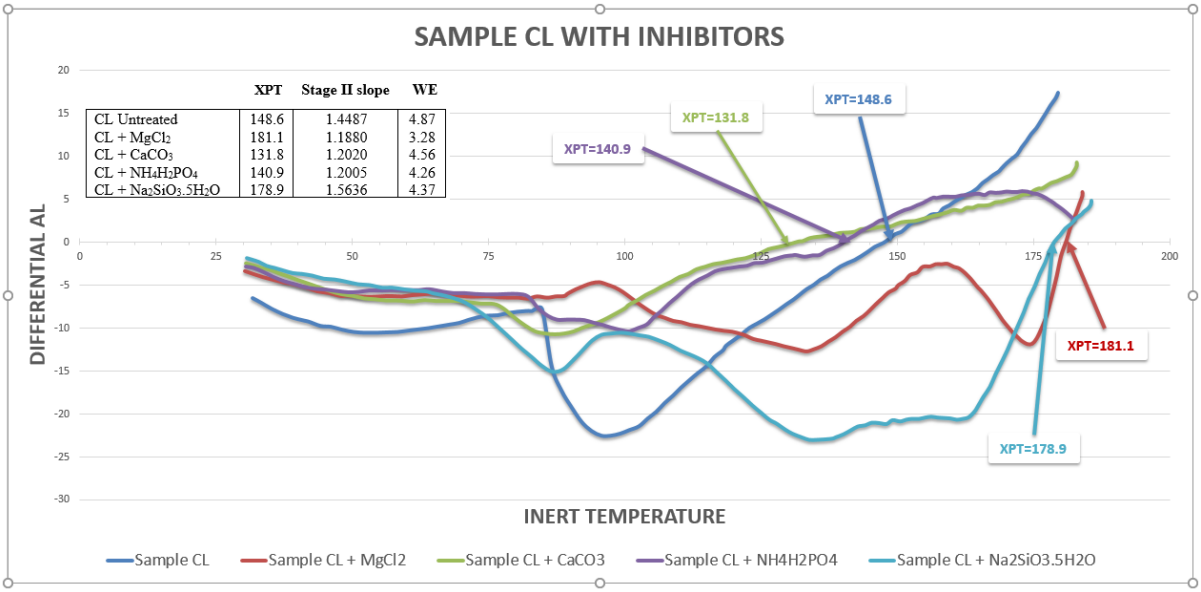

Figure 6.1 Differential analysis thermogram for untreated and treated sample CL

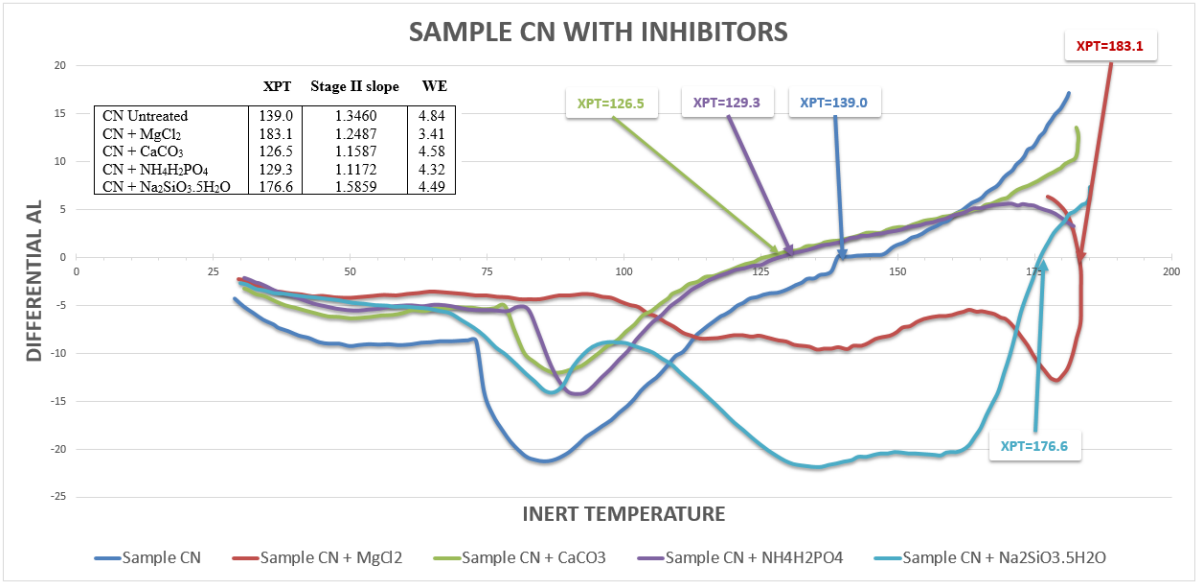

Figure 6.2 Differential analysis thermogram for untreated and treated sample CN

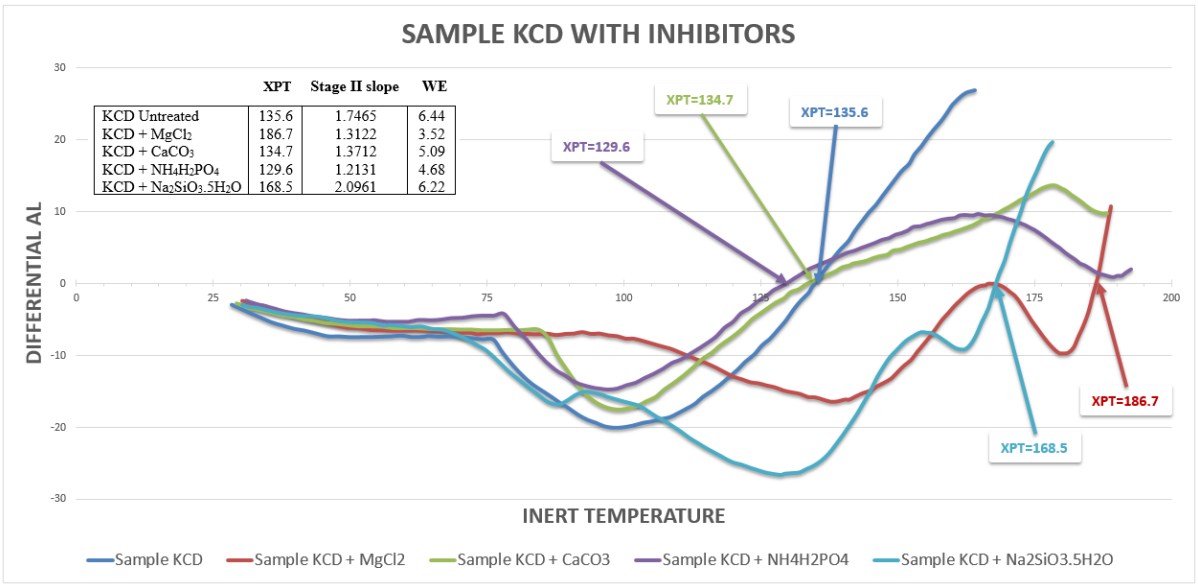

Figure 6.3 Differential analysis thermogram for untreated and treated sample KCD

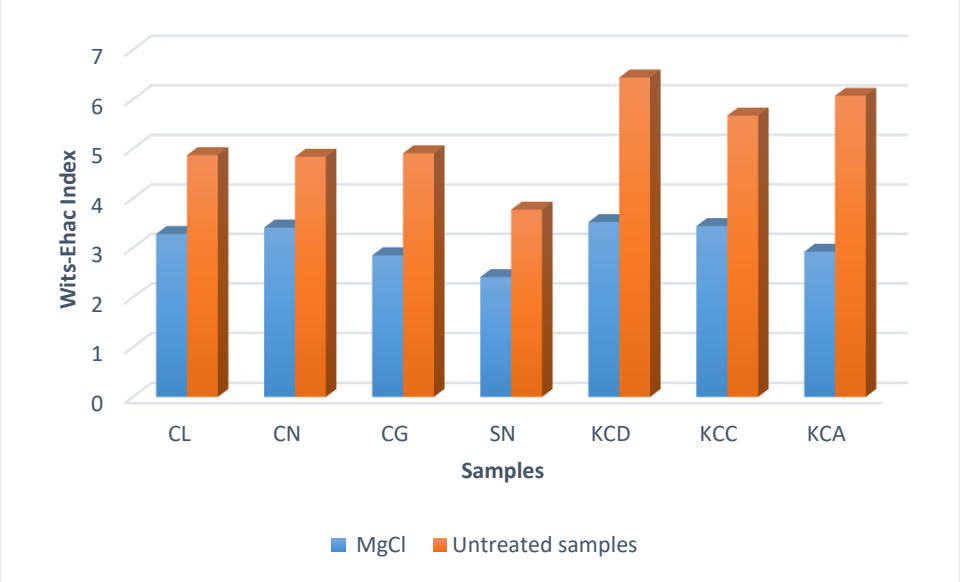

Figure 6.4 Wits-Ehac Index for untreated and treated ($MgCl_2$) coal and coal-shale samples

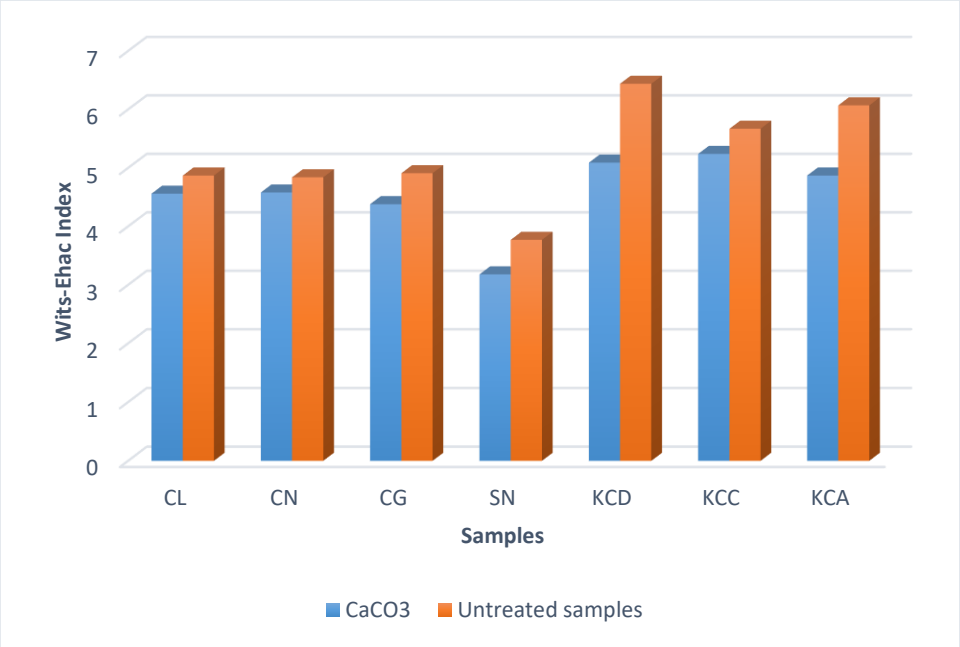

Figure 6.5 Wits-Ehac Index for untreated and treated ($CaCO_3$) coal and coal-shale samples

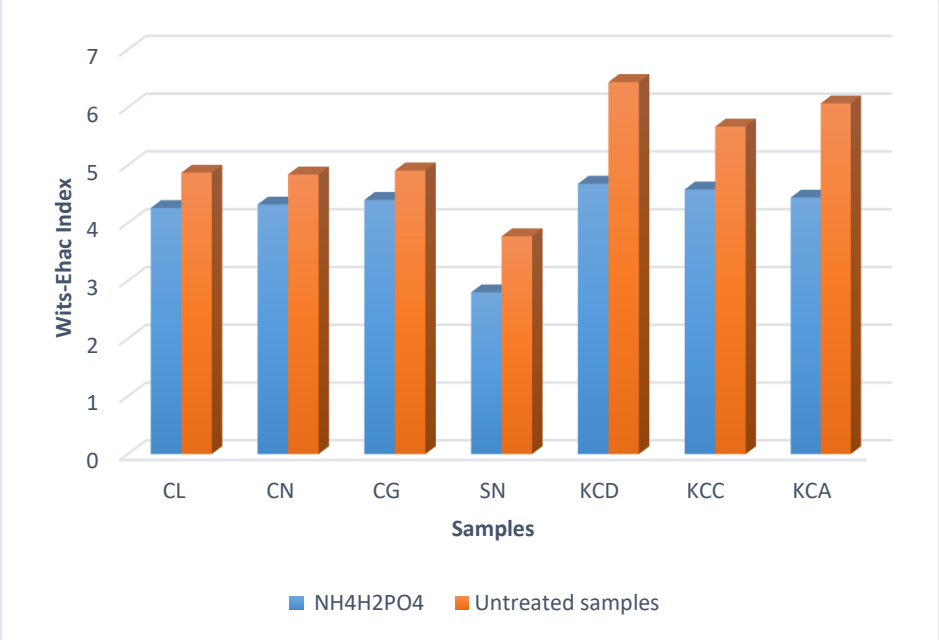

Figure 6.6 Wits-Ehac Index for untreated and treated ($NH_4H_2PO_4$) coal and coal-shale samples

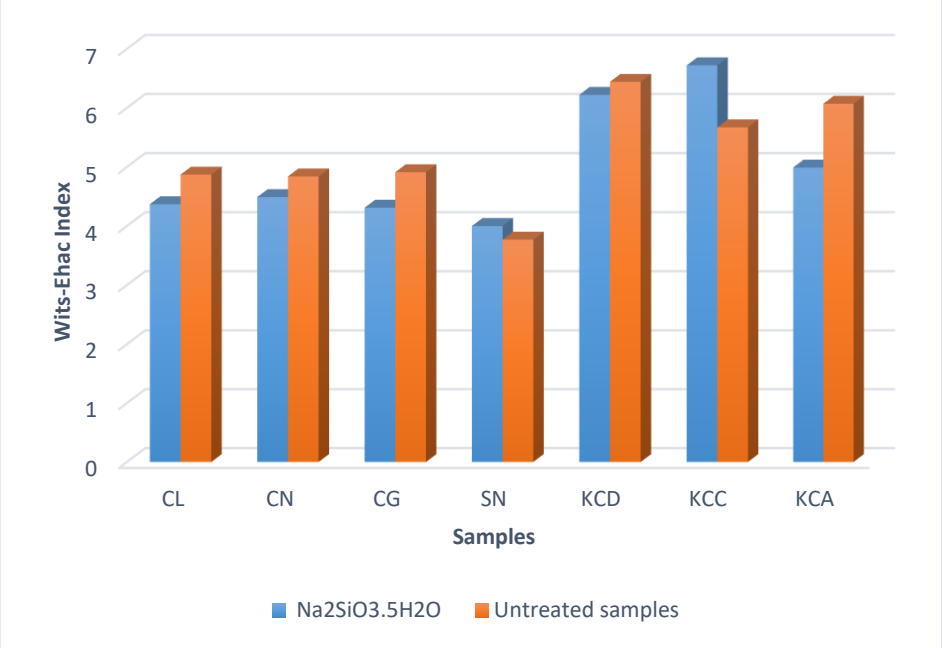

Figure 6.7 Wits-Ehac Index for untreated and treated ($Na_2SiO_3.5H_2O$) coal and coal-shale samples

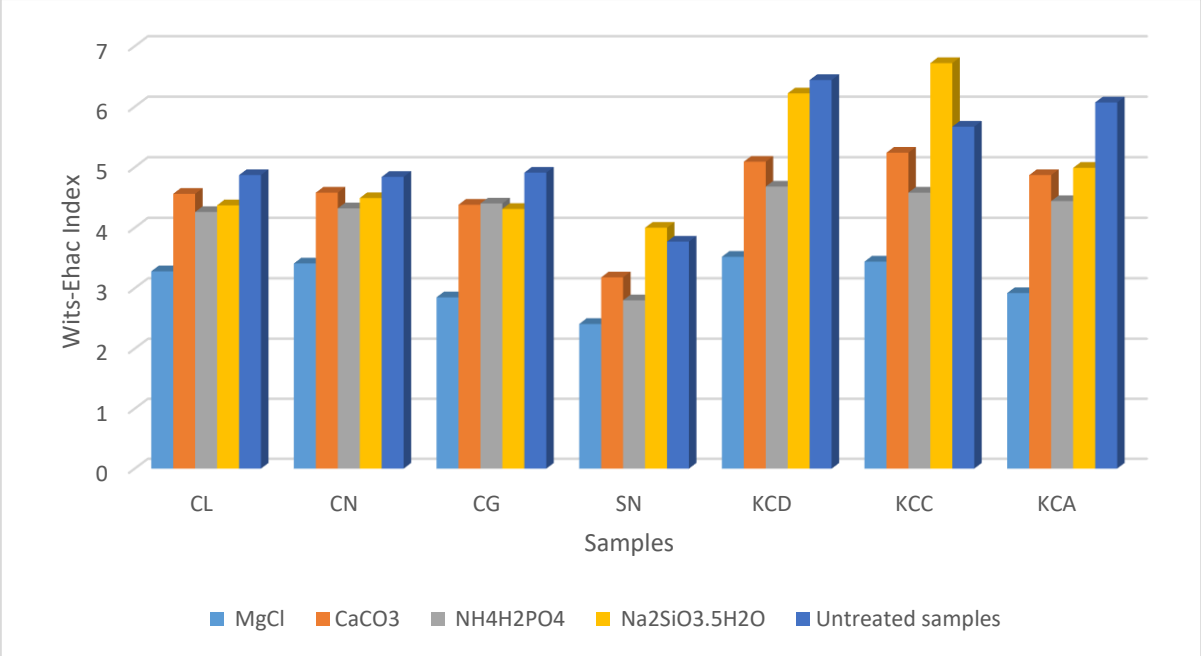

Figure 6.8 Wits-Ehac Index for untreated and treated coal and coal-shale samples

6.4 Cost performance evaluation for selected inhibitors

The PE was calculated based on the results obtained from the liability index for untreated and treated samples. The cost of inhibitors per 20g used is shown in Table 6.3.

Table 6.3 Cost of inhibitors per 20g in Rand (R)

Inhibitors	$MgCl_2$	$CaCO_3$	$NH_4H_2PO_4$	$Na_2SiO_3.5H_2O$
Price (R)	4.56	4.56	21.20	3.88

$MgCl_2$ provides a 32.5% liability index reduction for coal CL, while other inhibitors provide a 12.5% or less liability index reduction as shown in Table 6.1. Hence, $MgCl_2$ provides the best inhibition effect based on the liability index reduction for sample CL as compared with the other inhibitors. $MgCl_2$ provides a 29.5% liability index reduction for sample CN, while other inhibitors provide a 10.74% or less liability index reduction. Hence, $MgCl_2$ provides the best inhibition effect based on the liability index reduction for sample CG as compared with the other inhibitors. As shown in Table 6.1 and Table 6.2, $MgCl_2$ provides a 41.95%, 36.1%, 45.3%, 39.3% and 51.9% liability index reduction for samples CG, SN, KCD, KCC and KCA

respectively, while other inhibitors provide a 12.21%, 25.73%, 27.33%, 19.22% and 26.85% or less liability index reduction. In conclusion, $MgCl_2$ indicated the best liability index reduction performance for all the samples considered. The difference in the index reduction when compared to other inhibitors ranges from 10.37% to 29.74%. Considering the CPE of each inhibitor in Table 6.3, $Na_2SiO_3.5H_2O$ is the cheapest but also presents the highest abnormality in PE when compared with the other samples. Apart from $Na_2SiO_3.5H_2O$, $MgCl_2$ and $CaCO_3$ are the inhibitors with lower cost but proved to obtain better liability index reduction than $Na_2SiO_3.5H_2O$. However, $MgCl_2$ provides better PE than $CaCO_3$ with respect to their liability indices reduction. Hence, it can be concluded that $MgCl_2$ produces the best CPE than other selected inhibitors as shown in Tables 6.1 to 6.3. This is based on the premises that of all the inhibitors that produces a consistent liability indices reduction, $MgCl_2$ produces the best performance at the cheapest cost.

6.5 Summary

The treatment with the chemical inhibitors decreased the spontaneous combustion liability. The results suggest that chemical inhibitors can effectively retard the spontaneous combustion liability of coal and coal-shale. Analysis of the inhibitors tests on coal and coal-shale self-heating indicated that among the inhibitors, the presence of $MgCl_2$ blended with the samples markedly reduces the oxygen consumption and thus, decreases the spontaneous combustion liability index more than the other inhibitors. Hence, $MgCl_2$ has the best inhibitory effect on spontaneous combustion liability. $MgCl_2$ is indicated as the best liability index reduction performance for all the samples considered. The difference in the index reduction when compared with the other inhibitors which ranges from 10.37% to 29.74% at the cheapest cost. The capacity of the inhibitors to prevent the spontaneous combustion in this study can be arranged as $MgCl_2$, $NH_4H_2PO_4$, $CaCO_3$ and $Na_2SiO_3.5H_2O$. This implies that $MgCl_2$ may minimize the oxygen adsorption during low-temperature oxidation. The study shows that some inhibitors can minimize the oxidation processes of coal and coal-shale and thus, can be used to minimize the self-heating and spontaneous combustion in coal mines.

The choice of selecting an effective chemical inhibitor to control spontaneous combustion in coal mines continue to be a major challenge in the coal industry due to varying microscopic and inorganic constituents present in coal and coal-shale. The reliability of this study was analysed by a system of standard measurements to evaluate the performance of selected coal and coal-shale known to be highly prone to self-heating with and without chemical inhibitors.

The results show that $MgCl_2$ being one of the selected inhibitors minimize the spontaneous combustion liability of tested samples. The impeding effect of the selected inhibitors were evaluated using the Wits-Ehac tests to confirm the selection techniques. The spontaneous combustion liability index obtained from the treated samples is less than that obtained from the raw samples, thus indicating that treated samples absorbed less oxygen than the untreated samples. Chemical inhibitors, such as inorganic salts, can be used to minimise spontaneous combustion. The spontaneous combustion liability index of treated coal with $MgCl_2$ decreased from 4.87 to 3.28 for coal CL, 4.84 to 3.41 for coal CN, 4.91 to 2.85 for coal CG, 3.77 to 2.41 for coal-shale SN, 6.44 to 3.52 for coal KCD, 5.67 to 3.44 for coal KCC, and 6.07 to 2.92 for coal KCA respectively. The results confirmed that blends of inhibitors with coal and coal-shale can be used as a retardant material for spontaneous combustion in coal mines.

The investigation on the effect of selected inhibitors on the spontaneous combustion liability prove the concept that inhibitors can minimise the spontaneous combustion liability at a minimum cost under experimental conditions. Proper prediction and prevention of spontaneous combustion in coal mines is critical and serves a very essential role in the whole coal mining industry. The need for the prediction, assessment and management of the spontaneous combustion is significant for the efficient working and better productivity of Coalfields. The application of other inhibitors to retard the spontaneous combustion liability of coal and coal-shale has a significant scope for future work. Therefore, it will be more appropriate for further research to examine more inhibitors and evaluate their effects on the coal quality and properties on both laboratory and large-scale tests.

7 CONCLUSIONS AND RECOMMENDATIONS

7.1 Introduction

One of the challenges in the coal mining industry is the self-heating of coal as this can become a problem during mining operations, stockpiling and transportation. For a better understanding of the problem, this study reviewed the mechanism of spontaneous combustion, areas of spontaneous combustion, various factors influencing spontaneous combustion and different experimental studies to predict the occurrence of spontaneous combustion. Apart from coal itself, burning coal-shale is becoming a problem in some South African coal mines. Serious incidents of spontaneous combustion were reported as a result of the spontaneous combustion of coal-shale. Coal-shale can start spontaneous combustion depending on the various organic matter within the material and together with the nature of the associated coal or other carbonaceous materials. Limited studies have been conducted to evaluate factors affecting the spontaneous combustion liability of coal-shale in coal mines, therefore establishing techniques to predict and minimize the occurrence of spontaneous combustion was necessary and has been evaluated in this study. Different experimental methods reported in the past research have been reviewed, the Wits-Ehac Index and the Wits-CT Index were identified as being reliable and suitable methods to measure the spontaneous combustion liability of coal and coal-shale. This thesis conducted experimental tests to evaluate the cause of spontaneous combustion in coal-shale under conditions that might be found in practice.

The spontaneous combustion liability indices of coal and coal-shale were predicted with the Wits-Ehac apparatus. The tests involve a simultaneous determination of the XPT and DTA with an apparatus designed in the School of Mining Engineering, University of the Witwatersrand. However, the spontaneous combustion liability indices of some coal-shale samples could not be determined with the Wits-Ehac apparatus, therefore, this necessitated the development of a new device referred to as the Wits-CT apparatus. This test used the carbon content and the temperature differences due to the reaction of carbonaceous material and oxygen to establish an index termed the Wits-CT Index. An understanding of coal-shale intrinsic factors may be important in understanding the process of spontaneous combustion in mines. On the basis of these, both small and medium-scale spontaneous combustion tests and geochemical tests (proximate, ultimate, total sulphur, forms of sulphur, petrographic, XRF and

XRD analysis) were carried out on fourteen (14) representative *in-situ* bituminous coal and 14 coal-shale samples in order to establish relationships between them.

Statistical analyses were carried out to evaluate the relationships between the intrinsic properties and the spontaneous combustion liability indices of coal and coal-shale. The results obtained from the mentioned tests may be used as a reference when comparing coal-shale from different coal mines to predict their likelihood towards spontaneous combustion. This would enable an extensive database involving characterisation of intrinsic factors of coal and coal-shale to be established with regards to spontaneous combustion.

In addition, investigation of spontaneous combustion liabilities of some untreated and treated samples more liable to spontaneous combustion were conducted in order to prove the concept that inhibitors can be used to minimize spontaneous combustion. The results obtained from the spontaneous combustion liability indices indicated that the characteristics of coal and coal-shale treated with inhibitors varied from one sample to the other.

This thesis aim was to study the "spontaneous combustion liability of coal and coal-shale in the South African Coalfields". In order to provide answers to the research question in section 1.5, the main objectives and required methodologies were all successfully completed. From this study, the evidence suggests that coal and coal-shale tend to undergo spontaneous combustion when exposed to oxygen in the air. A research plan involving six (6) sections was formulated as the basis to answer the research question raised in Section 1.5 of this thesis. A general summary with the precise findings that answered the research questions is given as follows:

7.2 Key Findings of the Research

- A widely accepted spontaneous combustion liability index in South Africa (Wits-Ehac Index) was used to predict the spontaneous combustion liability of coal and coal-shale. The spontaneous combustion liability of some coal-shale could not be obtained due to their low reactivity. This necessitated the development of a new device that measured the spontaneous combustion liability under chemical reactions with oxygen. This device measures the spontaneous combustion liability of coal and coal-shale from which a liability index, referred to as the Wits-CT Index was established. The index, known as the Wits-CT Index was based on the results of fourteen (14) coal and 14 coal-

shale samples and proved to be effective in identifying materials known to be liable to spontaneous combustion;

- The results from the Wits-CT Index and the Wits-Ehac Index were compared with the incidents of spontaneous combustion in the mines and are in-line. The Wits-CT Index is established based on the experience of spontaneous combustion observed in the mines and proved to be reliable in identifying coal prone to spontaneous combustion and coal-shale which is not determined by the Wits-Ehac Index due to their low reactivity. The results of the Wits-CT Index are used to set a "standard" for the spontaneous combustion risk with respect to prehistorical information of spontaneous combustion incidents in South African coal mines. The high-risk zones were identified as the zones from which the samples had been collected. The samples varied in their spontaneous combustion liability indices and were found from zones which were likely to cause an incident at that specific mine;
- The XPT and Stage II slope values obtained from the DTA ranges from 139^{o}C to 167.2^{o}C for the coal samples and from 161.8^{o}C to 194.1^{o}C for the coal-shale samples, while the Stage II slope values ranges from 1.2763 to 1.4487 for the coal samples and 1.1050 to 1.2245 for the coal-shale samples respectively. The Wits-Ehac Index obtained from the values of the XPT and Stage II slope had a range from 3.82 to 4.91, while the coal-shale samples had a range from 2.98 to 3.77. The investigation shows that samples with higher Wits-Ehac Index values represent a high risk to spontaneous combustion. The spontaneous combustion test results show that coal samples CN (4.84), CM (4.79), CL (4.87), CG (4.91) and CE (4.76) will undergo spontaneous combustion faster than the other coal samples due to their high liability indices. However, for the coal-shale, samples SN (3.77) and SE (3.73) undergo spontaneous combustion faster than the other coal-shale samples based on their higher Wits-Ehac Indices. The Wits-CT Index varies between 4.05 to 9.59 for the coal samples and 0.27 to 3.77 for the coal-shale samples respectively. Coal samples CL (9.59), CM (7.27), CN (7.91), CG (7.53) and CK (9.1) will undergo spontaneous combustion faster than the coal samples with lower Wits-CT Indices. For the coal-shale, samples SN (3.99) and SE (1.66) with higher Wits-CT indices will also undergo spontaneous combustion faster than coal-shale samples with lower Wits-CT Indices. The results obtained from the spontaneous combustion tests indicated that the coal and coal-shale samples varies in their spontaneous combustion

liability indices. Hence, samples with higher liability indices will undergo spontaneous combustion faster;

- The study established a detailed experimental methodology to evaluate the spontaneous combustion liability of coal and coal-shale under laboratory conditions. It was found that coal has a higher spontaneous combustion liability index than coal-shale. Both materials show that an increase in carbon, moisture, hydrogen, volatile matter, nitrogen and a decrease in the ash content can complement spontaneous combustion liability. The petrographic results indicated that both coal and coal-shale have high amounts of inertinite, followed by vitrinite and liptinite content. It was found that coal-shale has higher liptinite and petrographically observable mineral matter than coal. The relationships between the various petrographic properties and the spontaneous combustion liability are not clearly indicated because each sample consists of a number of varying microscopic organic constituents (vitrinite, inertinite and liptinite) and inorganic constituents (mineral matter). It was found that the macerals in each sample differs, which is more likely to cause each sample to have a different spontaneous combustion liability index. These results obtained enabled further studies to be conducted on various constituents of macerals and established relationships with the spontaneous combustion liability indices;
- The results obtained from the XRF analysis indicated that the ash oxides differ considerably from one sample to the other and was generally dominated by SiO_2 and Al_2O_3. The XRD analysis shows that the coal and coal-shale samples differ in mineralogical composition and indicated that samples with a low ash content correspond to a low quartz content. Coal contains a higher amount of pyrite, dolomite and calcite than coal-shale, while coal-shale contains higher amounts of quartz, siderite, kaolinite, muscovite, microcline and plagioclase than coal. The study shows that the XRD and XRF are complementary for the identification of the mineralogical composition of coal and coal-shale;
- The study measured and determined the relationship between intrinsic properties and the spontaneous combustion liability indices using statistical analysis. The evaluation shows that the influence of selected intrinsic properties of coal-shale in relation to coal could be a tool to predict spontaneous combustion risk in coal mines. The results provide quantitative descriptions and showed relationships between the dependence of the spontaneous combustion liability indices and intrinsic properties. It was found that

the spontaneous combustion test results indicated linear relationships with some of the intrinsic factors and thus, identifies the major intrinsic factors affecting the spontaneous combustion liability. A definite positive or negative correlation coefficient exists between the intrinsic factors and the spontaneous combustion liability;

- The multiple regression analysis of the spontaneous combustion liability index on 12 independent variables (moisture, volatile matter, ash, carbon, hydrogen, nitrogen, sulphur, oxygen, vitrinite, liptinite, inertinite and mineral matter content) has developed acceptable and accurate predictive models as indicated by the R-squared values, the coefficient of correlations and standard error of estimates. The multiple regression analysis indicates that the Wits-Ehac Index and intrinsic properties show significant correlation coefficients of 0.939 and 0.998, R-squared values of 0.882 and 0.997, and a low standard error of estimates of 0.336 and 0.094 for coal and coal-shale respectively. The Wits-CT Index with intrinsic properties shows significant correlation coefficients of 0.992 and 0.999, R-squared values of 0.983 and 0.999 and a higher standard error of estimate of 0.772 and 0.123 for coal and coal-shale respectively;
- The models derived from the samples produced high correlation coefficients and a low standard error of estimates. The use of the models derived for the studied samples is more likely to give more accurate predictions for the spontaneous combustion liability of coal and coal-shale. Confidence in the predictive models was found by indicating that they established results similar to the actual liability indices. The influence of the intrinsic properties affecting spontaneous combustion liability may be evaluated more effectively by these models than by the linear regression. The results obtained may be used as a reliable tool to support previous works on the role of intrinsic properties on spontaneous combustion;
- The statistical analysis showed that the spontaneous combustion liability of coal and coal-shale can be determined by a model consisting of various intrinsic factors. The developed models enable mine management to identify the most important factors affecting spontaneous combustion and establishes relationships between them. Hence, no single factor is sufficient to influence the spontaneous combustion liability of coals and coal-shales but a cumulative effect of favourable factors may decide whether a material will be liable to spontaneous combustion or not;

- Treatment with the chemical inhibitors decreased the spontaneous combustion liability of coal and coal-shale. The investigation proved the concept that inhibitors can minimize spontaneous combustion liability;
- The liability index of treated coal with $MgCl_2$ decreased from 4.87 to 3.28 for coal sample CL, 4.84 to 3.41 for coal sample CN, 4.91 to 2.85 for coal sample CG, 3.77 to 2.41 for coal-shale sample SN, 6.44 to 3.52 for coal sample KCD, 5.67 to 3.44 for coal sample KCC, and 6.07 to 2.92 for coal sample KCA respectively. The XPT of the samples treated with $MgCl_2$ increases from 148.6^{o}C to 181.1^{o}C, 139^{o}C to 183.1^{o}C, 161.8^{o}C to 163.7^{o}C, 136.4^{o}C to 176.6^{o}C and 137.1^{o}C to 160.1^{o}C for all the treated samples respectively. The Stage II slope values decreases from 1.4486 to 1.1880, 1.3460 to 1.2487, 1.3815 to 1.0437, 1.2245 to 0.7890, 1.6559 to 1.0313, 1.5547 to 1.1015 and 1.7465 to 1.3122 respectively for all the treated samples. The XPT and Stage II slope values obtained from the samples treated with $MgCl_2$ show that as the XPT increases, the Stage II slope and Wits-Ehac Index decreases, while other inhibitors show variation in this findings; and
- $MgCl_2$ is indicated as the best liability index reduction performance for all the samples considered. The difference in the index reduction when compared to other inhibitors ranges from 10.37% to 29.74%. In addition, $MgCl_2$ produces the best CPE at the cheapest cost than the other selected inhibitors.

7.3 Research Contribution

- This study established that determined experimental factors can be linked to spontaneous combustion liability. The complex nature of coal and coal-shale has impeded the understanding of their chemical reactions with oxygen. However, the study showed that coal and coal-shale varies in their spontaneous combustion liabilities when exposed to oxygen in the air;
- The evidence of self-heating in highwalls and between bands of coal seams leading to spontaneous combustion is found in coal-shale;
- The spontaneous combustion liability and intrinsic properties of coal and coal-shale between coal seams vary throughout its horizontal extent. The relationships between the spontaneous combustion liability index and geochemical data were established and compared. The intrinsic properties of the coal and coal-shale have been identified to be the main factors contributing towards spontaneous combustion in coal mines;

- The study showed no single factor is sufficient to influence the spontaneous combustion liability of coal and coal-shale but a cumulative effect of favourable factors may influence whether a coal and coal-shale will be liable to spontaneous combustion. This finding is supported by the predictive models;
- There were significant differences in the various organic and inorganic constituents found in coal and coal-shale under microscopic studies. The importance of organic matter within coal and coal-shale lies in the fact that each macerals varies in degrees of their reactivity and oxidation rate. The specific constituents within the inertinite macerals of coal and coal-shale are more reactive than the other macerals and constitute a very important portion of the organic matter. The petrographic analysis has been found to be significant when evaluating the spontaneous combustion liability and hence, need to be carefully studied in the same way specific minerals and their resultant ashes during combustion are been evaluated;
- Coal and coal-shale contain a similar collection of common minerals, but the distribution and quantities varies from one sample to the other;
- The study established and evaluated relationships between the dependence of the spontaneous combustion liability indices on coal and coal-shale intrinsic properties which may serve as a point of reference when comparing their characteristics;
- Selected chemical inhibitors proved the concept that inhibitors can minimize spontaneous combustion liability at minimum cost. $MgCl_2$ produces the best CPE than the other selected inhibitors. The difference in the index reduction when compared to the other inhibitors ranges from 10.37% to 29.74% at the cheapest cost; and
- Spontaneous combustion liability can be minimized by altering the self-heating characteristics (i.e. chemical and physical characteristics of a coal and coal-shale surface) through the use of chemical inhibitors.

7.4 Research Limitation

- There was a number of limitations to this research. Much research has been conducted on spontaneous combustion, but no single study exists which satisfactorily covers all the different aspects of this reaction. For this study, the main restrictions were associated to the number, type and conditions of the samples that are already known to be prone to spontaneous combustion. An extensive search was conducted contacting

mining companies and research groups in South Africa; however, just a few samples were obtained based on the request by the sponsor;

- The access to equipment for the different experimental tests and the limited amount of time has restricted the extent of this study. This type of study deals with several uncontrolled factors and the application of any of the experimental techniques described to identify coal and coal-shale samples prone to spontaneous combustion must be compared with an alternative test; and
- Sampling protocols are not relatively simple and straight-forward for the heterogeneous nature of coal because it complicates the sampling procedures. In order to obtain representative samples for testing and analysis there is a strong emphasis on the need to obtain representative samples. Hence, the various composition of coal offers several challenges to analysts who need to ensure that a sample under investigation is representative of the whole. The substantial variation in coal quality and composition from the top to the bottom of the seam and between bands of coal seams offers challenges. The challenges faced in this study was that coal and coal-shale were collected from a relatively small portion that accurately represents the composition of the samples using the ply sample method. Therefore, samples must be representative of the composition of the whole as represented by the properties or quality of the sample. Measurements of the selected intrinsic properties are expressed as numerical values which requires high accuracy in coal analysis. The measurements need to be accurate in order to prevent negative scientific or economic consequences. This was achieved by using reliable standard tests methods for coal and coal-shale analysis (ASTM, ISO and SANS).
- A limited number of chemical inhibitors were used in this study to prove the concept that inhibitors can be used to minimize spontaneous combustion under laboratory tests.

7.5 Recommendations for Future Research Work

This section presents suggestions for further research. On the basis of the observations, discussion and results obtained so far, the recommendations are as follows:

- Some chemical inhibitors exhibit disadvantages, such as an unstable inhibitory effect when combined with coal and coal-shale obtained from different bands of coal seams. As a result of this, the selection of the optimal inhibitor for any given type of coal and

coal-shale is still a challenging task. Therefore, it is recommended to investigate more chemical inhibitors that have better effects to minimize spontaneous combustion and exhibit a high efficiency and a long active life time with minimum effects on coal quality;

- It would be most beneficial to the industry if further research can investigate the effects of various inhibitors on both laboratory and large-scale tests taking into consideration coal quality, coal properties and CPE on other coal samples; and
- In relation to spontaneous combustion, the influence of the gas content obtained during self-heating should be investigated.

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9 APPENDICES

9.1 Summary of paper abstracts on the research

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

9.1.1 Paper 1

Parts of Chapter 2 were compiled and published as the paper: **Moshood Onifade** & Bekir Genc., 2018. A review of Spontaneous combustion studies - South African context. Published by International Journal of Mining, Reclamation and Environment, https://doi.org/10.1080/17480930.2018.1466402. The following is an extract of the paper abstract:

"Abstract

One of the challenges in the coal mining industry is self-heating of coal, as this can become a problem during mining operations, stockpiling, and transportation. This study reviews the mechanism of self-heating, areas of spontaneous combustion, various factors influencing self-heating, and different experimental studies to predict its occurrence. The technical control measures applied to minimize self-heating with special consideration to South African coal are also discussed. It was observed that the distribution and association of unknown materials accelerating spontaneous combustion at different bands within the seam is yet to be studied. A site evaluation of the effects of these parameters are underway. The work presented here is part of a PhD research study in the School of Mining Engineering at the University of the Witwatersrand.

***Keywords**: Spontaneous combustion, open cast, barges, stockpile, impeded zones, thermodynamic".*

9.1.2 Paper 2

Parts of Chapter 3 and 4 were compiled and published as the paper: **Moshood Onifade**, Bekir Genc, & Andrew Carpede., 2018. A new Apparatus to Establish the Spontaneous Combustion Propensity of Coals and Coal-shales. Published by International Journal of Mining Science and Technology, Volume 28(4), pp. 649-655. The following is an extract of the paper abstract:

"Abstract

Coal and coal-shale both tend to undergo spontaneous combustion under favourable atmospheric conditions. The Wits-Ehac Index has been developed in South Africa since the late 1980's to test the spontaneous combustion liability of coal. However, in some cases, the Wits-Ehac Index fails to produce tangible results when testing coal-shale. To overcome this problem, a new apparatus has been developed to test carbonaceous materials such as coal and coal-shale under chemical reactions with oxygen and an index has been obtained. This index is called the Wits-CT Index. The equipment emulates the influence of oxygen adsorption on carbonaceous material for a period of 24 h without a heating system. The Wits-CT Index uses the total carbon content of the sample and the temperature variations obtained from the samples during reaction with oxygen to predict the spontaneous combustion liability. Eighteen samples have been analyzed using both indices and the results are in-line. It was found that coal and coal-shale with higher values of the Wits-CT Index are more liable to spontaneous combustion. Further research on different coal-shale is underway in order to establish an extensive database for coal and coal-shale, together with known incidences of self-heating.

***Keywords**: Coal; Coal-shale; Spontaneous combustion; Wits-Ehac Index; Wits-CT Index".*

9.1.3 Paper 3

Parts of Chapter 3 and 4 were compiled and published as the paper: **Moshood Onifade** & Bekir Genc., 2018. Spontaneous Combustion of Coals and Coal-shales. Published by International Journal of Mining Science and Technology, DOI: https:// doi.org/10.1016/j.ijmst.2018.05.013. The following is an extract of the paper abstract:

"Abstract

Spontaneous combustion of coal is a well-known phenomena around the globe. Apart from the coal itself, burning coal-shale is becoming a problem in the South African coal mines. Serious incidents of spontaneous combustion have been reported as a result of self-heating of reactive coal-shale. The intrinsic properties and spontaneous combustion tests of 28 selected coal and coal-shale samples were conducted and a relationship between the two has been established. Intrinsic properties were obtained by using the proximate and ultimate analysis, and spontaneous combustion liability tests results were obtained by using the Wits-Ehac and Wits-CT indices. The experimental results show that intrinsic properties of these materials

complement to the spontaneous combustion liability tests results. Comparative analyses of intrinsic properties and spontaneous combustion characteristics indicate similarities between the mechanism of coal oxidation and that of the oxidative processes undergone by coal-shale. For the tested samples, coal samples have a higher intrinsic spontaneous combustion reactivity rating than the coal-shale. Furthermore, an increase in carbon, moisture, hydrogen, volatile matter, nitrogen and a decrease in ash content indicate an increased proneness to self-heating. The concentration of pyrite found in the coal-shale accelerates self-heating. The event of spontaneous combustion can occur if coal-shale absorb sufficient oxygen when subjected to atmospheric conditions.

Keywords: *Spontaneous combustion; Coal-shale; Proximate and ultimate analysis; Wits-Ehac Index; Wits-CT Index".*

9.1.4 Paper 4

Parts of Chapter 3, 4 and 5 were compiled and published as the paper: **Moshood Onifade** & Bekir Genc. Modelling spontaneous combustion liability of carbonaceous materials. Published by International Journal of Coal Science and Technology, Volume 5(2), pp. 191-212. The following is an extract of the paper abstract:

"Abstract

This paper presents predictive models to determine spontaneous combustion liability of carbonaceous materials (coal and coal-shale) using statistical analysis. The intrinsic properties and spontaneous combustion liability index were determined by testing 14 coal and 14 coal-shale from Witbank Coalfields, South Africa. The relationship between these intrinsic properties (obtained from proximate, ultimate and petrographic analysis) and spontaneous combustion liability indices (the Wits-Ehac Index and Wits-CT Index) were established. The influence of the intrinsic properties of coal-shale in relation to coal properties affecting spontaneous combustion has been established using a statistical method. The linear regression analysis indicates better linear relationships between some of the selected intrinsic properties and spontaneous combustion liability index and thus, identifies the major intrinsic factors affecting their liability toward spontaneous combustion. It was found that a definite positive or negative correlation coefficient exists between the intrinsic factors and spontaneous combustion liability. A set of models to predict the spontaneous combustion liability was derived. The best significant correlation along with the most appropriate model as indicated

by R-squared values, the coefficient of correlations and standard error was used to predict the incident of spontaneous combustion.

Keywords: *Coal, coal-shale, statistical analysis, Wits-Ehac Index, Wits-CT Index and spontaneous combustion".*

9.1.5 Paper 5

Parts of Chapter 3 and 4 were compiled and published as the paper: **Moshood Onifade** & Bekir Genc. Comparative analysis of coal and coal-shale intrinsic factors affecting spontaneous combustion. Published by International Journal of Coal Science and Technology, Volume 5(3), pp. 282-294. The following is an extract of the paper abstract:

"Abstract

Coal and coal-shales tend to undergo spontaneous combustion under favourable atmospheric conditions. Spontaneous combustion liability index and intrinsic properties of coals and coal-shales varies between (above and below) coal seams. The spontaneous combustion liability index (obtained from the Wits-Ehac Index) and intrinsic properties (obtained from proximate, ultimate, and petrographic analysis) of fourteen samples representative of in situ coal (bituminous) and fourteen coal-shales obtained in Witbank coalfield, South Africa were experimentally studied. Comparative analysis of the relationships between the spontaneous combustion liability index and intrinsic properties of coals and coal-shales were established to evaluate their effects on self-heating potential. The intrinsic properties show linear relationship with spontaneous combustion liability and therefore, identifies the factors affecting spontaneous combustion of these materials. The influence of coal-shales intrinsic properties towards spontaneous combustion liability shows higher correlation coefficients than the coals. Both coals and coal-shales show inertinite maceral as major constituents than the vitrinite and liptinite macerals, hence the reactivity of inertinite macerals may show greater influence on spontaneous combustion liability. A definite positive or negative trends exists between the intrinsic properties and spontaneous combustion liability index. This research is part of a larger project which is considering the influence of intrinsic properties of coals and coal-shales on spontaneous combustion liability.

Keywords: *Coal-shale, spontaneous combustion, liability index, statistical analysis and correlation coefficient".*

9.1.6 Paper 6

Parts of Chapter 3 and 4 were compiled and published as the paper: **Moshood Onifade** & Bekir Genc. Prediction of the spontaneous combustion liability of coal and coal-shale using statistical analysis. Published by the Southern African Institute of Mining and Metallurgy, Volume 118, pp. 799-808. The following is an extract of the paper abstract:

"Abstract

In this study we investigate the intrinsic factors influencing the propensity of coals and coal shales to undergo spontaneous combustion using statistical analysis. The intrinsic properties were determined by testing 14 in situ bituminous coals and 14 coal shales from the Witbank coalfield, South Africa. The relationships between these intrinsic properties (obtained from proximate and ultimate analysis) and spontaneous combustion liability indices (the Wits-Ehac Index and the Wits-CT Index) were established using linear and multiple regression analysis based on set criteria. The linear regression analyses indicate that moisture, volatile matter, ash, carbon, hydrogen, and nitrogen contents are the main factors affecting the spontaneous combustion liability of coals, while moisture, volatile matter, ash, carbon, hydrogen, nitrogen and total sulphur contents are the factors affecting the spontaneous combustion liability of coal shales. The regression analysis shows either a positive or a negative correlation coefficient between the intrinsic factors and the spontaneous combustion liability index. Multiple regression of the spontaneous combustion liability index on eight independent variables was used to develop acceptable and reliable predictive models as indicated by high R-squared values, high correlation coefficients, and low standard error of estimates. The use of the models derived from this study may enable the spontaneous combustion liability of coals and coal shales to be reliably predicted.

Keywords: *Spontaneous combustion, coal and coal-shale, statistical analysis, Wits-Ehac Index, Wits-CT Index".*

9.1.7 Paper 7

Parts of Chapter 3 and 4 were compiled and published as the paper: **Moshood Onifade**, Bekir Genc, & Nicola Wagner. Influence of organic and inorganic properties of coal-shale on

spontaneous combustion liability by International Journal of Mining Science and Technology (Under Review). The following is an extract of the paper abstract:

"Abstract

Coal and coal-shale undergo low-temperature oxidation when exposed to air, potentially leading to spontaneous combustion. Coal-shale found in association with coal seams vary considerably in their intrinsic properties and spontaneous combustion liability index. Fourteen coal-shale samples collected from four different coal mines in Witbank Coalfield, South Africa, were experimentally investigated. The influence of coal-shale intrinsic properties and spontaneous combustion liability indices (determined by the Wits-Ehac Index and the Wits-CT Index) were established. The liability indices indicate relationships with the intrinsic factors and thus, identifying the major intrinsic factors affecting liability toward spontaneous combustion in these coal-shale samples. The XRF analysis indicated that the coal-shale samples are rich in SiO_2, Al_2O_3 and Fe_2O_3, while the XRD showed that same coal-shale samples are generally dominated with kaolinite and quartz. The coal-shale occurred in association with medium Rank C bituminous coal and contained varying proportion of macerals. The Wits-Ehac Index was unable to reliably determine liability indices of some coal-shale samples, and hence the Wits-CT Index was developed. The results obtained from the characterisation tests may be used as a tool to predict the spontaneous combustion liability in carbonaceous material and may serve as reference when comparing coal-shale from different coal mines.

Keywords: *Wits-Ehac Index, Wits-CT Index, Witbank Coalfield, self-heating, statistical analysis and correlation coefficients**".***

Publications (Published Conference Proceedings)

9.1.8 Paper 8

Parts of Chapter 3 and 4 were compiled and published as the paper: Bekir Genc, **Moshood Onifade**, & Alan Cook. Spontaneous combustion risk on South African Coalfields: Part 2. Proceedings of the 21st International Coal Congress of Turkey 'ICCET' April 11-13, 2018, Zonguldak, Turkey; 2018, pp. 13-25. The following is an extract of the paper abstract:

"Abstract

Spontaneous coal of occurs in nearly every coalfield worldwide, though the frequency and extent of the incidents varies from country to country. The risk of spontaneous combustion is a well-known problem and has been recently reviewed in the South African coal mines between the period of 2013 and 2017. One-hundred and fifty-eight coal (158) samples from different mines have been tested and evaluated to predict the areas with high risk of spontaneous combustion using the Wits-Ehac Index and the crossing point temperatures. The obtained results have indicated that the coal mines possess a medium to high-risk spontaneous combustion. It was observed that the possibility of spontaneous heating to occur is a function of the coal quality and the surrounding in which it is situated. The study has established an extensive database for comparing coal on relative rating scales with respect to spontaneous heating. The database is frequently being updated as more coal samples are tested and will provide the basis for an improved risk evaluation practice for the event of spontaneous combustion.

Keywords: *Spontaneous combustion, Wits-Ehac Index, crossing point temperatures and database**".***

9.1.9 Paper 9

Parts of Chapter 3 and 4 were compiled and published as the paper: **Moshood Onifade** & Bekir Genc. Establishing relationship between spontaneous combustion liability indices. Proceedings of the 21st International Coal Congress of Turkey 'ICCET' April 11-13, 2018, Zonguldak, Turkey; 2018, pp. 1-11. The following is an extract of the paper abstract:

"Abstract

Spontaneous combustion liability index has been one of the tools used to predict the propensity of coal to spontaneous heating in coal mines and other related industry. This paper has established the relationship and interrelationship between spontaneous combustion liability indices (Wits-Ehac Index and Wits-CT Index), Crossing Point Temperature (XPT) and carbon content. The linear regression analysis has been used to establish the relationship between these variables. The linear regression analyses show that carbon content gave better correlation coefficients with the Wits-CT Index, Wits-Ehac Index and the XPT for the coal-shale than for the coal samples. The carbon content has been evaluated and confirmed as one of the intrinsic factors influencing the propensity of coal and coal-shale to spontaneous combustion. It has been found that no clear relationship exists between the results of the Wits-Ehac, the Wits-CT Index, and the crossing point temperatures. However, the two spontaneous combustion liability indices provide reliable results to predict coal and coal-shale liable to self-heating.

Keywords: *Spontaneous combustion, crossing point temperature, Wits-Ehac Index and Wits-CT Index".*

9.1.10 Paper 10

Parts of Chapter 3 and 4 were compiled and published as the paper: **Moshood Onifade** & Bekir Genc. Ash, volatile matter and carbon content influence on spontaneous combustion of coal-shale, 18th International Symposium on Environmental Issues and Waste Management in Energy and Mineral Production, 19-23 November, 2018, Santiago, Chile (Accepted). The following is an extract of the paper abstract:

"Abstract

Self-heating of coal-shale has been reported in the South African coal mines to be a source of spontaneous combustion. Spontaneous combustion tests were conducted on a series of coal-shale samples collected from affected coal mines using a newly developed liability index (Wits-CT Index). The ash content, volatile matter and the carbon content of the samples were investigated and their effect toward spontaneous combustion liability was established. It was found that as ash content increases, the liability towards spontaneous combustion decreases, while as the volatile matter and carbon content increases, the liability towards spontaneous

combustion increases. Furthermore, it was found that the ash content showed a negative effect on the liability index, while volatile matter and carbon content showed positive effect on the liability index. Therefore, coal-shale with low ash content and high volatile matter and carbon content are more likely to be prone to spontaneous combustion.

Keywords: *Spontaneous combustion, coal-shale, liability index and Wits-CT Index*''.

9.2 How to compute correlation coefficient in MS Excel

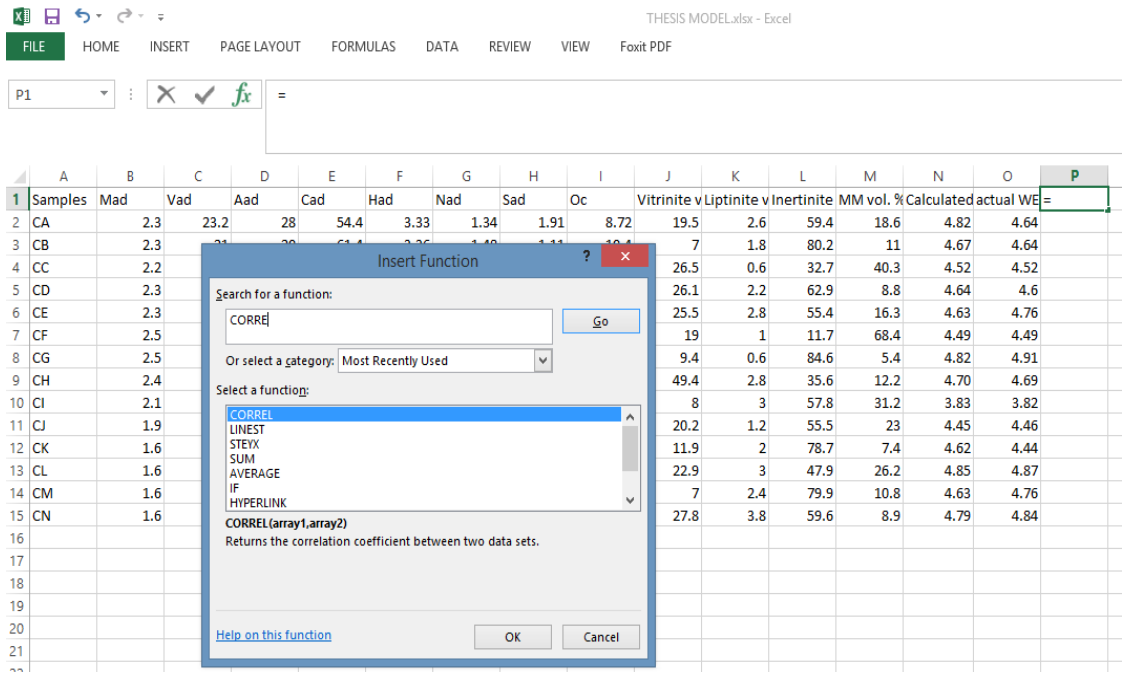

Figure 9.1 Generating correlation coefficient in Ms Excel-step 1 and step 2

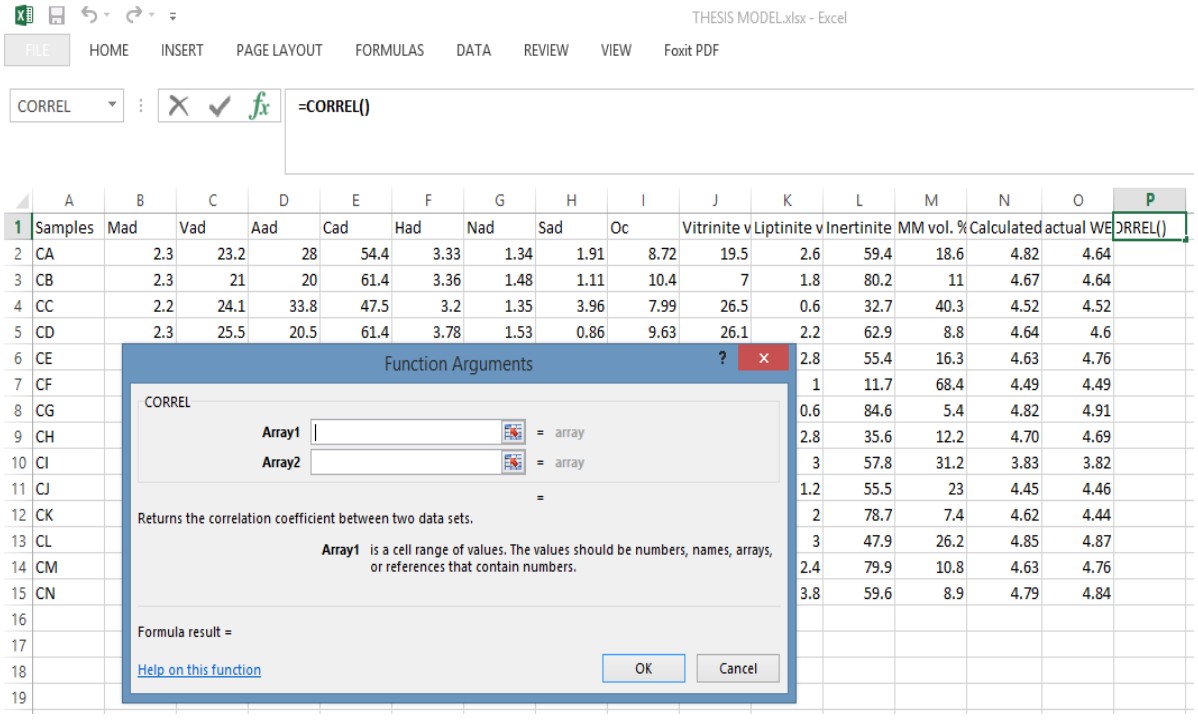

Figure 9.2 Generating correlation coefficient in Ms Excel-step 3 and step 4

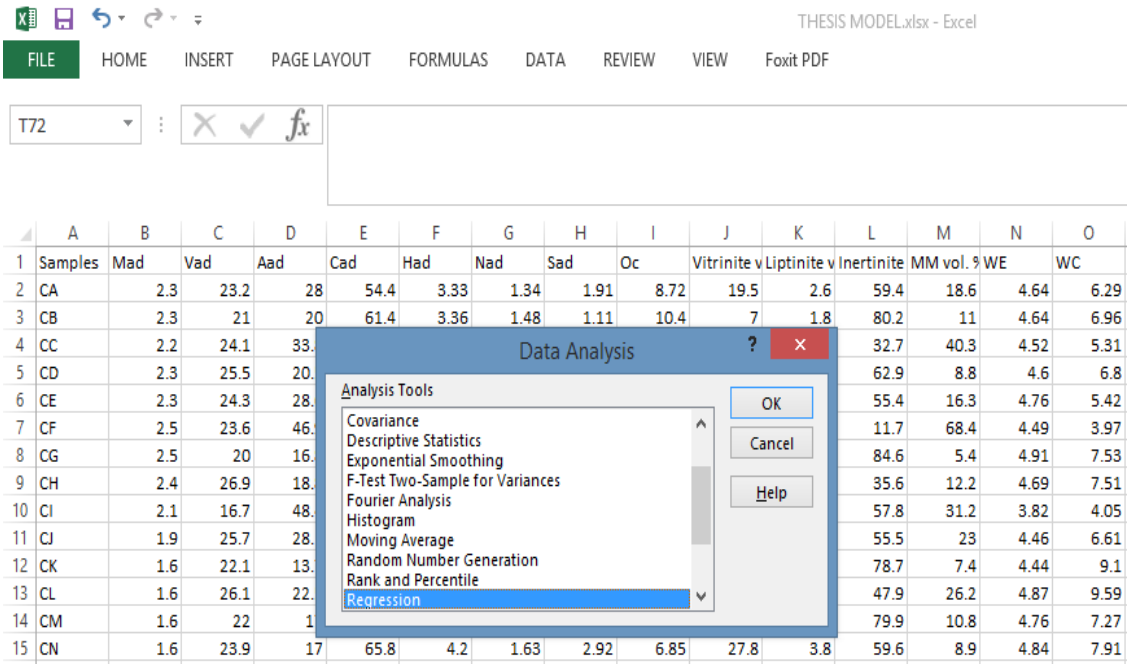

Figure 9.3 Generating multivariable linear regression in Ms Excel-step 1

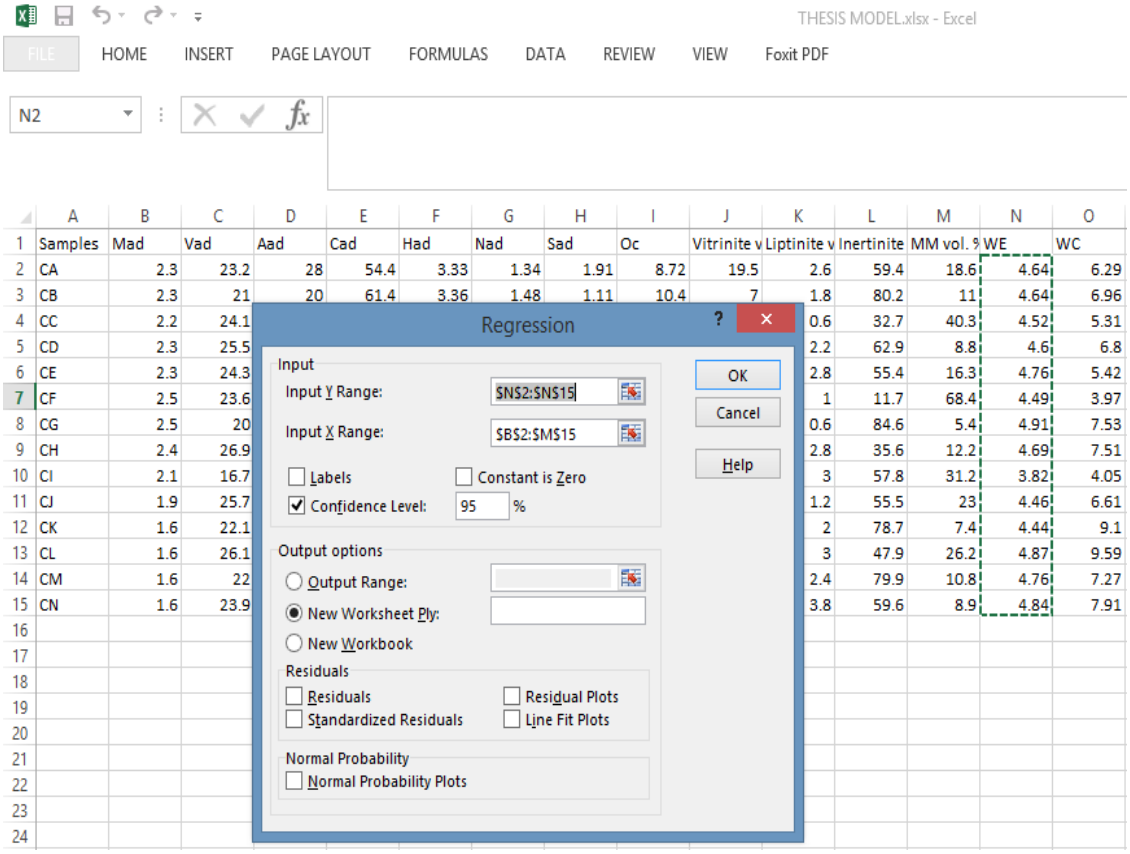

Figure 9.4 Generating multivariable linear regression in Ms Excel step 2

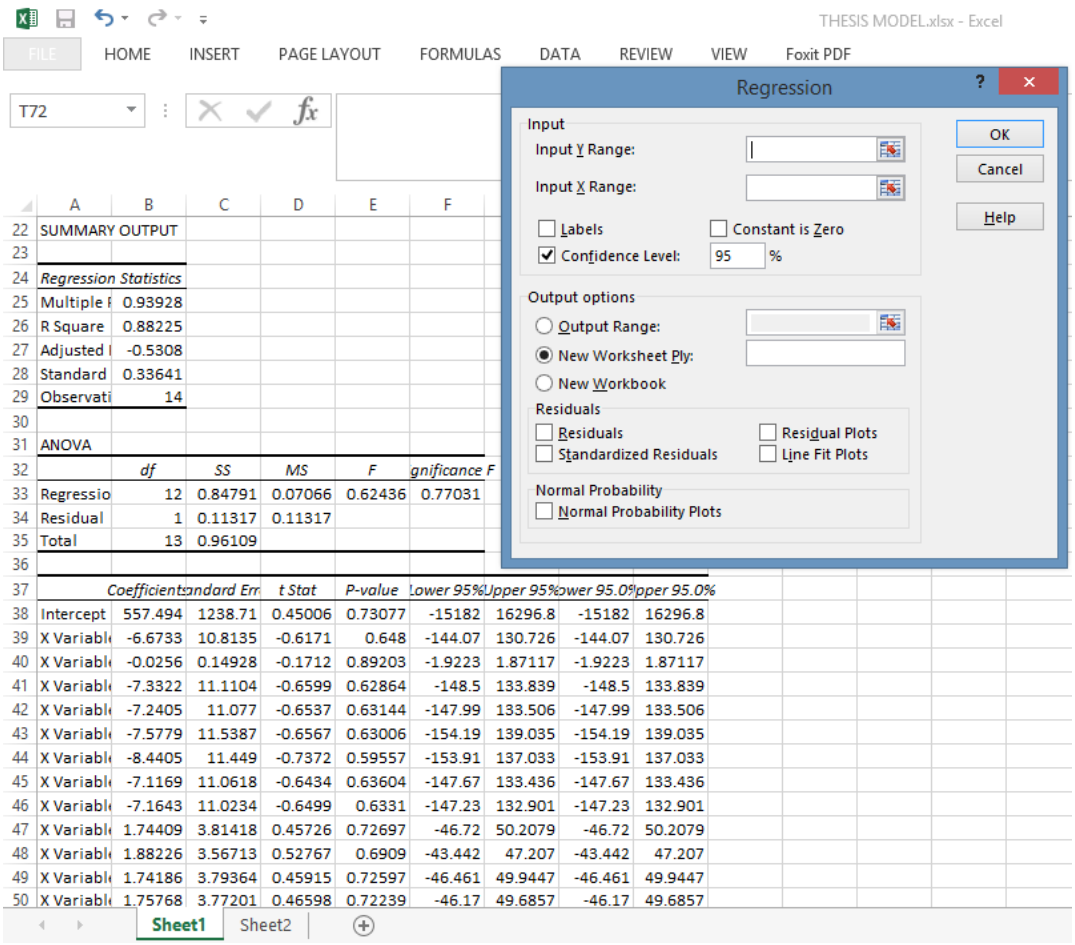

Figure 9.5 Generating multivariable linear regression in Ms Excel-step 3

9.3 Summary of R-squared values obtained from linear regression graphs of intrinsic properties and spontaneous combustion liability indices for coal and coal-shale

Table 9.1 Relationships between independent (proximate and ultimate analysis, total sulphur and forms of sulphur, wt.%-ad) and dependent variables for the coal

Independent variables	**Range**	**Dependent variables Correlation coefficients**	
		Wits-Ehac Index	**Wits-CT Index**
Moisture	1.60-2.50	(0.0637)	(0.5847)
Volatile matter	16.7-26.9	0.5164	0.3273
Ash	13.7-48.4	(0.6884)	(0.8660)
Carbon	35.9-69.7	0.6572	0.8565
Hydrogen	2.55-4.21	0.6616	0.7552
Nitrogen	0.85-1.63	0.6945	0.8382
Oxygen	5.65-10.4	(0.0686)	(0.1016)
Sulphur	0.59-5.30	(0.0115)	(0.0687)
Pyritic sulphur	0.13-4.13	0.0869	0.0553
Sulphate sulphur	0.01-0.42	(0.6422)	(0.7620)
Organic sulphur	0.28-1.09	(0.1787)	(0.2914)

Table 9.2 Relationships between independent (vitrinite and inertinite macerals, vol.%) and dependent variables for the coal

Independent variables	**Range**	**Dependent variables Correlation coefficients**	
		Wits-Ehac Index	**Wits-CT Index**
Vitrinite and its group			
Total vitrinite	7.0-49.4	0.2591	0.0890
Total vitrinite (mmf)	7.9-60.0	0.0874	(0.2755)
Collotelinite	1.6-39.0	0.1481	0.0255
Collotelinite (mmf)	1.7-44.4	0.0439	(0.2451)
Collodetrinite	2.8-13.3	0.3944	0.1580
Collodetrinite (mmf)	3.1-18.8	0.1725	(0.2401)
Inertinite and its group			
Total inertinite	11.7-84.6	0.1679	0.4752
Total inertinite (mmf)	36.9-90.1	(0.0735)	0.2754
Fusinite	0.8-7.6	0.5663	0.2860
Fusinite (mmf)	1.2-12.1	0.4392	0.0447
Secretinite	0.8-6.0	0.1284	0.1143
Secretinite (mmf)	1.1-6.7	0.0064	(0.1791)
Reactive semifusinite	0.2-7.7	0.1993	0.0818
Reactive semifusinite (mmf)	0.3-8.4	0.1874	0.0253
Inert semifusinite	5.1-41.4	0.2518	0.3035
Inert semifusinite (mmf)	7.4-43.8	0.0882	0.0927
Total semifusinite	5.5-43.4	0.2898	0.3098
Total semifusinite (mmf)	14.1-45.9	0.1460	0.1010
Reactive inertodetrinite	0-4.6	0.1353	0.0382
Reactive inertodetrinite (mmf)	0-45.7	0.1232	0.0185
Inert inertodetrinite	3.2-47.1	(0.1111)	0.4274
Inert inertodetrinite (mmf)	10.0-50.9	(0.3103)	0.2640
Total inertodetrinite	3.2-49.4	(0.0894)	0.4065
Total inertodetrinite (mmf)	10-55.5	(0.2768)	0.2504

Table 5.3 Relationships between independent (liptinite, vol.%) and dependent variables for the coal

Independent variables	**Range**	**Dependent variables Correlation coefficients**	
		Wits-Ehac Index	**Wits-CT Index**
Liptinite and its group			
Total liptinite	0.6-3.8	0.0280	0.2621
Total liptinite (mmf)	0.6-4.4	(0.2228)	(0.1108)
Sporinite	0.4-3.4	0.2321	0.4353
Sporinite (mmf)	0.7-3.8	0.2116	0.3235

Table 9.3 Relationships between independent (various petrographic properties, vol.%) and dependent variables for the coal

Independent variables	**Range**	**Dependent variables Correlation coefficients**	
		Wits-Ehac Index	**Wits-CT Index**
Total reactive maceral	11.2-53.0	0.3148	0.1292
Total reactive maceral (mmf)	12.8-64.4	0.1229	(0.2814)
Total macerals	31.7-94.6	0.3785	0.6496
Total mineral matter	5.4-68.4	(0.3782)	(0.6496)

Table 9.4 Relationships between independent (proximate and ultimate analysis, wt.%-ad) and dependent variables for the coal-shale

Independent variables	**Range**	**Dependent variables Correlation coefficients**	
		Wits-Ehac Index	**Wits-CT Index**
Moisture	0.8-1.7	0.7715	0.4507
Volatile matter	8.5-16.6	0.6389	0.6901
Ash	51.5-88.7	(0.8352)	(0.9448)
Carbon	2.66-33.7	0.7962	0.9630
Hydrogen	0.75-2.87	0.5795	0.8799
Nitrogen	0.08-0.96	0.6446	0.9462
Oxygen	5.01-11.85	(0.3212)	0.3243
Sulphur	0.12-6.90	0.5791	0.0242
Pyritic sulphur	0.04-4.26	0.5704	0.0367
Sulphate sulphur	0.01-0.45	0.5365	(0.0403)
Organic sulphur	0.05-2.19	0.5933	0.0147

Table 9.5 Relationships between independent (vitrinite and inertinite macerals, vol.%) and dependent variables for the coal-shale

Independent variables	**Range**	**Dependent variables** **Correlation coefficients**	
		Wits-Ehac Index	**Wits-CT Index**
Vitrinite and its group			
Total vitrinite	0.4-8.4	0.1230	0.2620
Total vitrinite (mmf)	2.4-39.3	(0.2632)	(0.1854)
Collotelinite	0-4.2	0.2206	0.3366
Collotelinite (mmf)	0-19.6	(0.1073)	(0.0928)
Collodetrinite	0-2.6	0.2105	0.1289
Collodetrinite (mmf)	0-12.1	(0.2330)	(0.1933)
Inertinite and its group			
Total inertinite	4.9-46.1	0.7360	0.8029
Total inertinite (mmf)	44.9-91.7	0.2143	0.3262
Fusinite	0-4.0	(0.4038)	0.1313
Fusinite (mmf)	0-16.9	(0.5898)	(0.0427)
Secretinite	0-1.8	0.6003	0.6009
Secretinite (mmf)	0-4.8	0.3304	0.1523
Reactive semifusinite	0-1.1	(0.2795)	(0.0250)
Reactive semifusinite (mmf)	0-4.6	(0.3862)	(0.1234)
Inert semifusinite	0.2-8.0	0.7688	0.2971
Inert semifusinite (mmf)	1.3-40.7	0.2970	(0.2238)
Total semifusinite	0.2-8.0	0.7776	0.2967
Total semifusinite (mmf)	1.3-40.7	0.2970	(0.2238)
Reactive inertodetrinite	0-3.6	0.3292	(0.3140)
Reactive inertodetrinite (mmf)	0-18.5	(0.0829)	(0.4564)
Inert inertodetrinite	1.9-34.8	0.7460	0.8455
Inert inertodetrinite (mmf)	14.8-76.6	0.2480	0.4974
Total inertodetrinite	3.3-35.2	0.7749	0.8388
Total inertodetrinite (mmf)	25.2-80.5	0.2367	0.4460

Table 5.6 Relationships between independent (liptinite, vol.%) and dependent variables for the coal-shale

Independent variables	**Range**	**Dependent variables Correlation coefficients**	
		Wits-Ehac Index	**Wits-CT Index**
Liptinite and its group			
Total liptinite	0.4-5.5	0.3360	(0.0276)
Total liptinite (mmf)	2.6-25.0	(0.0113)	(0.4033)
Sporinite	0.4-5.5	0.3463	(0.0238)
Sporinite (mmf)	2.6-25.0	0.0059	(0.3997)

Table 9.6 Relationships between independent (various petrographic properties, vol.%) and dependent variables for the coal-shale

Independent variables	**Range**	**Dependent variables Correlation coefficients**	
		Wits-Ehac Index	**Wits-CT Index**
Total reactive maceral	1.6-12.8	0.2923	0.0575
Total reactive maceral (mmf)	9.6-63.9	(0.2552)	(0.4294)
Total maceral	9.9-53.1	0.7653	0.8026
Total mineral matter	46.9-90.1	(0.7653)	(0.8029)