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Spontaneous combustion liability of coal and coal-shale: a review of prediction methods

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Abstract This study presents a review of the various methods to predict the spontaneous combustion liability of coal and coal-shale. The relative propensity of coal to undergo self-heating can be established by different methods. These methods are well established in their usage, but the fact that no particular test method has become a standard to predict the spontaneous combustion liability 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. Comprehensive studies that centres on the international position on research being conducted by academics, different research institutes and industries on spontaneous combustion of coal and coal mine fires were evaluated. Relationships between the geochemical analysis (proximate and ultimate analysis, forms of sulphur, petrographic properties, X-ray diffraction and X-ray fluorescence) and spontaneous combustion testing methods (numerical and experimental approaches) used to predict the spontaneous combustion liability of coal were reviewed. The combination of these tests provides a better understanding of the mechanism that controls the spontaneous combustion phenomena. However, irrespective of the extensive studies that have been conducted over time, spontaneous combustion is still a major problem in the coal value chain.

Keywords Coalfields · Spontaneous combustion liability · Macerals · Numerical and geochemical analysis

1 Introduction

Coal oxidizes 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 and occurs in almost every coalfield around the globe, however, the rate of occurrence varies from one country to the other. Over the years, spontaneous combustion is one of the challenges faced by both coal producers and users during mining, storage and transportation. 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 coal temperature and promotes the oxidation reaction. As a result of the origin 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.

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; Onifade et al. 2018a; Restuccia et al. 2017). These materials have pore spaces together with the occurrence of carbon (Dullien 1979; Onifade et al.

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2018b). This enables the rock to be porous to air, and with an increase in the surface area, the organic particles will promote self-heating through absorption and diffusion (Onifade and Genc 2018a; Dullien 1979). Self-heating of coal and coal-shale has been reported in South African coal mines to be capable of starting spontaneous combustion (Onifade and Genc 2018b, c, d). 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; Rumball et al. 1986).

Experimental investigations on spontaneous combustion of bulk coal samples have been carried out under a medium to large scale test (Akgun and Arisoy 1994a; Beamish et al. 2001a, b; Chen 1991; Cliff et al. 1998; Monazam et al. 1998; Smith et al. 1991; Stott 1980) with a heating system used to initiate the self-heating process. Studies on spontaneous combustion of coal stockpiles under the effect of atmospheric conditions are documented by Fierro et al. (1999), (2001), Ozdeniz and Sensogut (2006), Ozdeniz and Yilmaz (2009), 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. Small-scale tests involving the use of relatively small amounts of pulverized, dry coal samples has been used for more than 30 years in South Africa and some parts of the world to measure the spontaneous combustion liability of coal (Avila 2012; Banerjee 2000; Genc et al. 2018; Nimaje and Tripathy 2016; Ren et al. 1999). 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 to large scale test that considered the self-heating of a substantial coal mass under atmospheric conditions.

Different works have been conducted to predict the spontaneous combustion liability of coal. This is possible because an understanding of the fundamental mechanisms of self-heating is achieved by research-based investigations. To make this review more coherent, comparisons surrounding the experimental studies to predict the spontaneous combustion liability of coal and coal-shale causing spontaneous combustion in coal mines have been evaluated.

2 History of the characterization and developments

Self-heating of coal takes place when adequate oxygen from the air is sufficient to support the reaction between coal and oxygen. The heat created by low-temperature oxidation of coal is not sufficiently dispersed either by conduction or convection, thereby causing an increase in temperature within the coal mass. The influence of oxygen on coal at low temperatures is exothermic as a whole, while some reactions are endothermic (Shi et al. 2005; Wang et al. 2003). Self-heating is the cause of heat that leads to spontaneous combustion in coal mines. Exothermic processes such as microbial metabolism, the reaction of coal with moist and pyrite oxidation can promote self-heating if the heat produced by coal oxidation is not sufficiently dispersed to the environments through conduction, convection and radiation. The conditions that necessitate the occurrence of spontaneous combustion in coalfields exclusive of the effect of external factors are;

(1) Reaction of coal and oxygen;
(2) Exothermic reaction followed by the generation of heat; and
(3) Heat generated must exceed the heat dissipated.

Once coal surface contacts with oxygen, adsorption/absorption/reaction will occur, releasing heat and gaseous products at the same time. The chemical reaction is expressed as follows (Schmal 1989; Singha and Singh 2005; Stracher and Taylor 2004).

$$\text{Coal} + \text{Oxygen} \rightarrow \text{Oxygen} + \text{Heat} \rightarrow \text{Gas product} \quad (1)$$
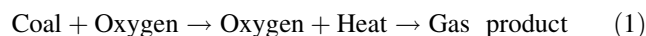
The occurrence of spontaneous combustion is not limited to coal but the phenomenon is known to take place in a number of coal-shale, pyritic black shale and coal refuse (Kim and Chaiken 1990; Onifade and Genc 2018e, f, g; Restuccia et al. 2017; Rumball et al. 1986). The principles that control spontaneous combustion are similar in all cases, irrespective of the material that is involved. The rate of oxygen adsorption in coal is dependent on the coal rank because of the degree of metamorphism (coalification). The heat generated by coal is the cause of coal oxidation and it depends on a number of factors such as the amount of organic and inorganic constituents (Brooks et al. 1988a, b). Coal exposed to oxygen in the air for long periods of time is less reactive than freshly exposed coal (Brooks et al. 1988a, b; Phillips et al. 2011). Brooks et al. (1988a, b) indicated that fine coal particles require limited amounts of oxygen to flow due to its low permeability, while coarse coal is very reactive due to its high permeability and specific surface area. Oxygen can penetrate into coal seams from the top and portions that are in contact with oxygen in the air. 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.

2.1 Areas of coal self-heating

Self-heating is a challenge in the coal value chain (Arisoy and Akgun 2000; Banerjee 1985; Beamish et al. 2001a, b; Brooks et al. 1988a, b; Fierro et al. 1999; Genc and Cook 2015; Phillips et al. 2011). Self-heating in coal stockpiles usually occur in the long-term storage of thermal power stations, in open cast mines, underground old workings, highwall, waste dumps and transportation in cargo ships or trains covering long distances (Carras and Young 1994; Moghtaderi et al. 2000; 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 reported for a better understanding on the mechanism of self-heating in large-scale tests (Cliff et al. 1998; Ozdeniz et al. 2011, 2014; Ozdeniz and Sensogut 2006). Coal stockpiles experience low self-heating which minimizes the calorific value and the amounts of coal stocked. The difference in the ambient temperature due to sun radiation and variation in stockpile particle sizes may affect spontaneous combustion (Cliff et al. 1998; Fierro et al. 1999, 2001; Nelson and Chen 2007; Ozdeniz et al. 2015).

Coal barge/silo/bunker causing subsequent unplanned closure is another area, which is known to cause self-heating. Coal stored inside a silo for a period of time may oxidize and begin to self-heat. The time taken for a coal to self-heat is dependent on the intrinsic properties of a particular coal type (Falcon 2004). Areas in which spontaneous combustion have occurred are shown in Figs. 1 and 2.

## 2.2 Factors affecting spontaneous combustion liability of coal

It is a known fact that self-heating of coal depends on two main factors, such as intrinsic and extrinsic factors. These factors occur as a result of the cumulative effects of various intrinsic and extrinsic factors. The intrinsic factors are related with the inherent properties or origin of the coal, i.e. its physico-chemical characteristics, petrographic properties and mineral composition. The extrinsic factors are associated with the atmospheric, geological and mining conditions prevailing during coal mining operations and these are mainly site specific and difficult to determine. In this study, an attempt has been made to evaluate the role of the intrinsic factors affecting the spontaneous combustion of coal. Each factor is, in turn, the combined influence of a number of sub-factors. The factors influencing the spontaneous combustion liability of coal can be classified as follows;

(1)   Geological factors;
(2)   Seam factors; and
(3)   Mining factors.
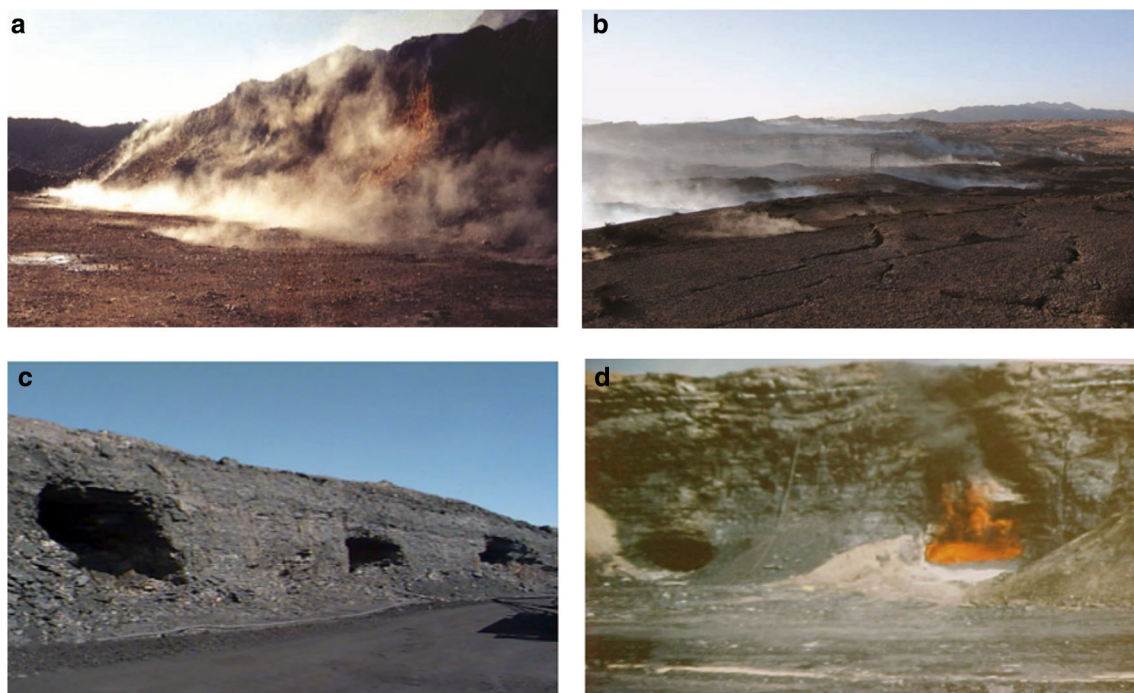
Fig. 1 a Coal waste heap during spontaneous combustion b symptom of self-heating in large fracture commonly observed in Wuda coal mining area (Singha and Singh 2005), c spontaneous combustion of open bords and d highwall at New Vaal, South Africa (Stenzel 2002)
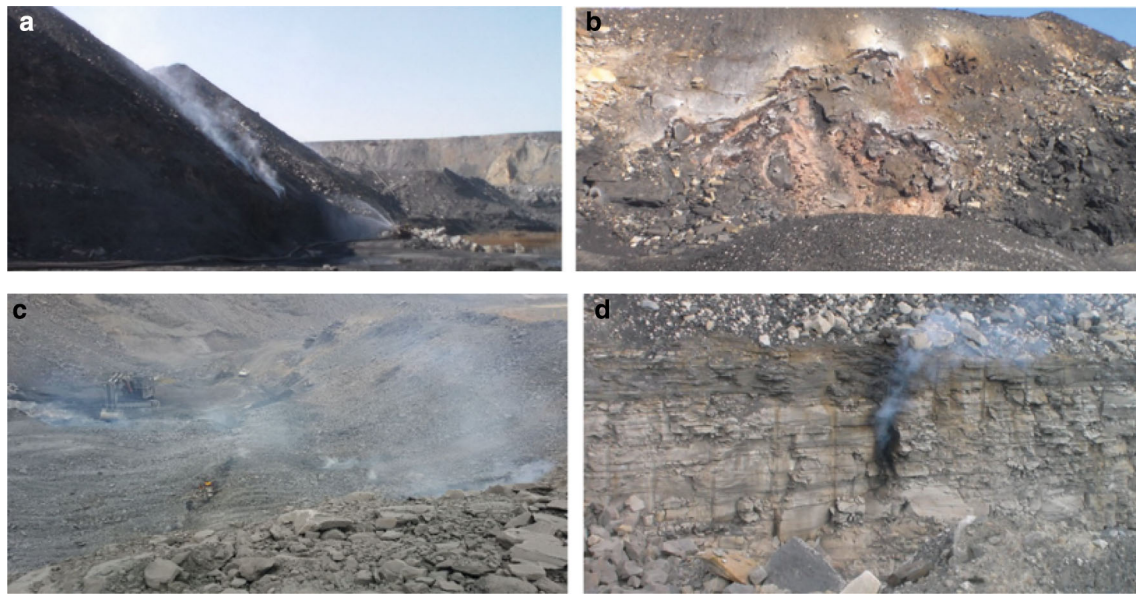
Fig. 2 **a** Self-heating of run-of-mine and **b** spoil heaps **c** signs of self-heating in coal seams and **d** self-heating of in-seam, Witbank Coalfields, South Africa (Onifade et al. 2018a; Onifade and Genc 2018b)

The mining factor is extrinsic, while the seam and geological factors are intrinsic (Bhattacharyya 1982; Eroglu 1992; Phillips et al. 2011).

2.2.1 Geological factors

These factors include seam thickness, seam gradient, coal burst, geothermal gradient, depth of cover, coal friability, faulting, caving characteristics, surrounding strata conditions and geological discontinuities (fissures, joints and cracks). A deep insight into the mechanism of the effects of these factors are analysed below;

(1) *Seam thickness* In a situation where the seam thickness is greater than that which cannot be mined completely in one part or as a whole, the area is more liable to spontaneous combustion, since the un-mined area tends to be exposed to sluggish ventilation flow. Spontaneous combustion is reported to be dependent upon the physical factors involved by the thickness of the seam, the methods of working, type of ventilation and the friable nature of the coal. In thick seams certain bands within the section can also be more liable to spontaneous combustion than others. The thicker the seam, the more difficult it becomes to avoid leaving relatively high risk coal within the goaf area (Bhattacharyya 1982; Eroglu 1992).

(2) *Seam gradient* Board and pillar and long wall methods are usually used in flat seams which are less prone to spontaneous combustion. In an inclined seam, control of combustion becomes more complex, since convection current resulting from the differential temperature tend to cause air currents in the goaf (Bhattacharyya 1982; Eroglu 1992).

(3) *Coal outbursts* This commonly occur in the harder formations and higher rank coals rather than in the softer and lower rank coals. However, great care must be taken where there is a likelihood of coal outburst and spontaneous combustion occurring concurrently, as the danger of the products of an outburst are very intense.

(4) *Geothermal gradient* Geo-thermal gradient doesn't directly influence the self-heating process; although, where geothermal gradients are high, the strata temperature in the workings will increase more rapidly with an increasing depth of working than where the geothermal gradient is low. If a seam is at a higher geothermal gradient zone, then the liability towards spontaneous combustion is high (Bhattacharyya 1982; Eroglu 1992).

(5) *Faulting* The faulted group normally has an influence on the spontaneous combustion liability of coal. Any excavating action along the fault plane resulting in the formation of the fine coal and the ventilating air which enters with that fault to the seam, or oxidises that fine coal, will result in spontaneous combustion. A fault generally slows down the rate of face advance to a safe minimum, with the incidental risk of heat development.

(6) *Depth of cover* The depth of cover does not necessarily influence the incidence of spontaneous combustion. In practice, the greater the depth of

cover, the higher the natural strata temperature and hence, the higher is the base temperature of the coal in question.

(7) *Coal friability* The more friable the coal is, the larger the surface area exposed to oxidation, thus tending to generate more heat per unit volume of coal. As coal breaks easily after extraction, the surface area also increases according to that. If the surface area is increased, then more surface area is exposed to the air. So, with the increase of the surface area the spontaneous combustion liability also increases (Bhattacharyya 1982; Eroglu 1992).

2.2.2 Seam factors

Some of the seam factors affecting the spontaneous combustion liability of coal include moisture, volatile matter, ash/mineral matter, coal rank, sulphur content, heat due to earth movement, particle size/surface area, porosity, sulphur, weathering, bacteria, temperature, ventilation, petrographic properties, thermal conductivity and pyrite content (Eroglu 1992; Falcon 2004; Kaymakci and Didari 2002; Mastalerz et al. 2010; Onifade and Genc 2018b, e, f; Phillips et al. 2011; Restuccia et al. 2017; Rumball et al. 1986). Factors such as the oxygen concentration and temperature (Van der Plaats et al. 1984), inherent moisture content (Bhattacharyya 1971), the presence of mineral particles (Sujanti and Zhang 2001) and the surface area (Singh and Demirbilek 1987) can also affect the spontaneous combustion liability. The oxidation reaction can be affected by the amount of each maceral (Mastalerz et al. 2010; Misra and Singh 1994; Onifade and Genc 2018e, f, g; Scott 2002; Scott and Glasspool 2007), coal rank (vitrinite reflectance) (Misra and Singh 1994) and volatile matter and chemical composition of coal (Marinov 1977; Onifade and Genc 2018e, f).

Particle size and porosity affect the spontaneous combustion liability of coal (Itay et al. 1989; Mastalerz et al. 2010). Nugroho et al. (2000) confirmed that coal mass with different particle diameter (0.18–2.67 mm) will combust faster than those with a fine particle size (0.18 mm). This is because large particles possess the influence of reduced specific surface area and bulk density. It was found in the study that the self-heating characteristics of high-rank coal is dependent on particle sizes. Kucuk et al. (2003) and Ren et al. (1999) indicated that the spontaneous combustion liability of coal increases with a decrease in particle sizes. 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 is based on a complex relationship between various intrinsic and

extrinsic factors of coal (Onifade and Genc 2018c; Uludag 2007). Bhat and Agarwal (1996) indicated that the coal particle size may influence heat loss via 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 and Arisoy 1994b; Arisoy and Akgun 2000; Bhattacharyya 1971; Bhat and Agarwal 1996; Didari 1988; Onifade et al. 2018a; Panigrahi et al. 2000; Ren et al. 1999). The unusual wetting and drying of coal on stockpile surfaces accelerate spontaneous combustion due to adsorption (Beamish and Hamilton 2005; Clemens and Matheson 1996). Akgun and Arisoy (1994b) investigated the effects of air humidity and inherent moisture content affecting spontaneous combustion of coal. It was observed that when dry air is transported over 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 and 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. During the wetting and drying of coal, heat is transferred between coal and oxygen in the air. An increase or a decrease in heat transfer affects the oxidation rate and moisture content.

Panigrahi and Sahu (2004) and Onifade and Genc (2018b, e) indicated that as ash content increases, the spontaneous combustion liability of coal decreases. Beamish and 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. A similar study by Onifade and Genc (2018b, e) indicated a negative relationship between spontaneous combustion liability indices (Wits-Ehac Index and Wits-CT Index) and ash content. Sia and Abdullah (2012) and Zivotic et al. (2013) reported that a high ash content is due to the presence of a peat depositional environment where the condition of flooding of the paleomire occurs intermittently during deposition. Blazak et al. (2001) and Onifade and Genc (2018b, e) 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 Snyman and Botha (1993) and Van Niekerk et al. (2008) indicated that the air-dried ash content of South African coal varies between 21.3 and 38.8 wt%. Choudhury et al. (2007) found that some Indian coal has ash content greater

than 45 wt%. Studies shows that the carbon content of coal varies between 40 and 52 wt% (Roberts 2008), 56.7 wt% and 69.6 wt% (Czaplicki and Smolka 1998), 49.0 wt% and 58.23 wt% (Department of Minerals and Energy South Africa 2004). Hydrogen content in coal ranges between 3.07 and 3.20 wt% (Roberts 2008), 2.60 wt% and 3.13 wt% (Department of Minerals and Energy South Africa 2004) and nitrogen content varies between 0.76 and 1.13 wt% (Czaplicki and Smolka 1998) and 0.56 wt% and 1.44 wt% (Department of Minerals and Energy South Africa 2004).

The presence 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 the spontaneous combustion liability of coal (Kim 1977; Mastalerz et al. 2010). This factor is very complex because it provides the oxygen needed for the oxidation reaction and dissipates 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). Walters (1996) investigated that airflow supplies adequate 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 increase 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 and Glasser 1986).

The mineral matter within a coal has been studied by a number of researchers (Mastalerz et al. 2010; Suarez-Ruiz and Crelling 2008; Ward and French 2005; 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 and Arisoy 2008; Beamish et al. 2005; Humphreys et al. 1981; Smith et al. 1988). Beamish and Arisoy (2008) indicated that the distribution of mineral matter in coal delays 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 matters are in the form of mineral phases and mineral species of different particle sizes. The quantity of trace elements in coal has a great influence on the environmental, economic and spontaneous combustion liability (Finkelman and Gross 1999; Swaine and Goodarzi 1995). The trace elements in coal may exist as organic and inorganic matter (Wagner and Hlatshwayo 2005). The characteristics of trace elements during the utilization and combustion processes of coal can be controlled by the amount of trace elements and their frequency of occurrence (Swaine and Goodarzi 1995). The occurrence of trace elements in a mineral matter within coal influences the spontaneous combustion liability (Finkelman and Gross 1999; Mardon and Hower 2004).

The presence and distribution of sulphur content in coal have been analyzed by various researchers (Chandra et al. 1980; Mastalerz et al. 2010). Studies have shown that the total sulphur in coal varies between 0.93% and 3.35% (William 1994), 0.4% and 1.29% (Wagner and Hlatshwayo 2005), 0.43% and 0.63% (Czaplicki and Smolka 1998), 1.47% and 1.56% (Roberts 2008), 0.59% and 9.45% (Hsieh and Wert 1985), 0.74% and 1.23% (Department of Minerals and Energy South Africa 2004), and 5.4%–15.1% (Olivella et al. 2002) for South African coal and other coal resources around the globe. The sulphur content in deep coal seams has been considered to be higher than shallow coal beds. An excess amount of sulphur in coal promotes self-heating leading to spontaneous combustion. The presence of high sulphur contents in coal has been reported (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 are difficult to distinguish. The inorganic sulphur in coal occurs in the form of pyrite and sulphate. Studies reported on the forms of sulphur shows that the pyrite content in coal varies between 0.11% and 1.15% (William 1994), 3.9% (Hsieh and Wert 1985) and 0.3% and 3.3% (Olivella et al. 2002) for South African coal and some coal samples from around the globe. It was reported that the sulphate content in coal varies between 0.7% and 2.17% (William 1994) and 0.3% and 7.6% (Olivella et al. 2002). Previous studies indicated that the organic sulphur in coal varies between 0.12% (William 1994) and 0.26%–4.30% (Hsieh and Wert 1985). Gupta and Thakur (1977) indicated moisture and sulphur content as factors that cause 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 liability of coal towards spontaneous combustion (Mastalerz et al. 2010; Rumball et al. 1986). Pyrite reacts with oxygen in the presence of water to form hydro-peroxide (H_2O_2) and therefore, initiates oxygen. Pyrite with a concentration above 2% has been reported to promote the coal oxidation (Bhattacharyya 1971). Beamish and 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 as follows (Lain 2009).

$$Fe_2S + 7O_2 + 16H_2O = 2H_2SO_4 + 2FeSO_4 \cdot 7H_2O + 1321 \text{ kJ} \qquad (2)$$
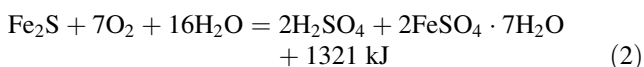
The reaction above is exothermic and occurs at low temperature. The heat generated from the reaction doubled coal with the same amount of oxygen (Garcia 1999; Martinez et al. 2009). Hence, the occurrence of pyrite is a contributory factor affecting spontaneous combustion liability of coal. Chandra et al. (1980) found that the distribution of sulphate sulphur in Assam coal remains the same from the lower to the upper seam. Dutta et al. (1983) indicated an increase in the amount of total sulphur with a decrease in the sulphate sulphur content of coal.

Chandra et al. (1991) indicated that the crossing point temperatures (XPT) and volatile matter decrease with increasing coal rank. Pattanaik et al. (2011) showed an increase in spontaneous combustion liability with a consistent decrease in the degree of coalification. Raju (1988) indicated that an increase in the volatile matter up to a value of 35% with a decrease in XPT. Previous researches used volatile matter and selected intrinsic factors of coal to predict the spontaneous combustion liability (Banerjee 2000).

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 in 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 studies on petrographic properties of coal have been documented by Falcon (1978), Holland et al. (1989), Mastalerz et al. (2010), Snyman and Botha (1993) and Steyn and 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 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 and Blazak (2005) and Ivanova and 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 because of the relationship between coalification and spontaneous combustion of coal. Misra and 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 and Prasad 1990; Morris and 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 provide pathways for the ingress of oxygen in the air within coal seams than in vitrinite (Didari 1988). It was reported that the quantity of exinite, liptinite and vitrinite contents affect the spontaneous combustion liability of coal and, with an increase in the vitrinite reflectance, the liability towards spontaneous combustion progressively increases (Avila 2012; Chandra et al. 1991; 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 and 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 and Mikula (1990); Raju (1988); Ren et al. (1999); 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 and Mikula (1990) indicated that spontaneous combustion should not be based only on coal rank but other properties (thermal conductivity and resistivity, pyrite content, porosity and etc.) should be evaluated. Kim (1977) indicated that as coal rank decreases, the liability toward spontaneous combustion increases.

2.2.3 Mining factors

In open cast mines, the quantity of coal left on the coaling bench, micro and macro cracks in benches and outcrops affect spontaneous combustion (Phillips et al. 2011). In underground mines, factors such as pillar and roof conditions, rate of advance, ventilation system and airflow, waste material in abandoned areas, mining method, multi-seam working, heat from machines, worked out areas, etc. are the mining factors affecting spontaneous combustion of coal (Eroglu 1992; Phillips et al. 2011). A brief description of the influence of mining factors on spontaneous combustion liability are described below:

(1) *Mining methods* Partial pillar extraction underground mining methods, in which part of the coal seam is left in the goaf and in pillars, can promote the likelihood of spontaneous combustion. This is significant to the mining of old workings, since surface mining methods will expose zones where crushed coal has been left over a period of time which can also be a cause of spontaneous combustion. The long wall mining leaves extracted areas lying between the entries serving the working places. The differential pressure in the ventilation system will permit air to flow across these areas, which causes a high risk of combustion (Bhattacharyya 1982; Eroglu 1992; Phillips et al. 2011).

(2) *Rate of advance* The rate of advance during mining operations is one of the causes of spontaneous combustion in the coal mines. Ideally, when a working face is operating normally, any individual piece of coal passes through the zone at a rate equal to the rate of advance of the working face. The time taken for any individual piece of coal from entering or leaving the zone is very critical. If the time is excessive, the oxidation can occur to an unacceptable degree, thereby, causing spontaneous combustion (Bhattacharyya 1982; Eroglu 1992; Phillips et al. 2011).

(3) *Pillar conditions* Pillar size and strength have a direct effect on the spontaneous combustion liability of coal. In practice, pillars should be of a size to avoid crushing. This size depends on the strength of the coal, the depth of the cover and the influence of other workings within the panel. Increase in methane emission is an indication of crushing around pillars that consequently results into self-heating (Bhattacharyya 1982; Eroglu 1992; Phillips et al. 2011).

(4) *Roof conditions* Poor roof releases the shock waves to pass through easily and the developments of cracks increases within the roof and that leads to spontaneous combustion. Poor roof conditions increase the spontaneous combustion liability of coal. Roof fall leave cavities which have to be supported and are regularly filled with timber. Due to their nature, these zones always fill with methane and ignition frequently results to spontaneous combustion (Eroglu 1992; Phillips et al. 2011).

(5) *Ventilation system and air flow rate* Air flow rate is a complex factor because air provides oxygen, while it also carries away the heat generated. There is a critical air quantity that provides sufficient oxygen to permit the coal to oxidize but is not sufficient to prevent the heat produced from accumulating (Kim 1977; Phillips et al. 2011).

(6) *Leakage* This arises where leakage paths exits at air crossings, in and around regulators and doors due to air escapes through fissures, and other similar zones where the outflow is a high pressure gradient and tendency for air to flow through solid coals. Ideally, it is not practicable to rely on making stoppings impermeable and if they were completely impermeable a dangerous pressure of firedamp could be seen accumulating (Kim 1977).

(7) *Multi seam workings* In a situation whereby one seam is working above or below the other seam, spontaneous combustion will develop for roof conditions and leakage of air. Where a multi-seam situation occurs, both during the working of the first seam and of succeeding seams, incidence of spontaneous combustion can occur for the seam currently being worked and any other seam above or below it (Eroglu 1992; Phillips et al. 2011).

(8) *Coal losses* The accumulation of broken coal in worked-out areas is a serious heating problem in which most gob fires result from. There is no mining system that can guarantee that remnants will never be left in a worked-out area. Most mining systems result in a significant loss of coal. In most cases the coal is likely to be crushed, finely divided, and in a location where build-up is possible, must be considered a potential hazard (Bhattacharyya 1982; Eroglu 1992; Phillips et al. 2011).

(9) *Waste material in abandoned areas* The timber props left in the waste caused the coal roof to collapse and formed a saving, therefore encouraging the spontaneous combustion of coal. Presence of timbers in the mines results in the danger of ignition which generates the heat required for spontaneous combustion.

(10) *Heat from machines* Normally heat from machines is dissipated within the ventilating air stream and the temperature rise of the general body of the air is

likely to be very small. In cases where the influence of the heat from machines is secondary, additional air may have to be circulated and this will require a higher ventilating pressure resulting in an increased risk of leakage (Phillips et al. 2011).

(11) *Worked-out areas* Worked-out areas which are not properly sealed by ventilation stoppings are potential causes of spontaneous combustion. They are expected to have suspension in the ventilation system as a result of roof falls or flour lift tending to impair in rib condition and the accumulation of broken coal contributing to potential combustion (Phillips et al. 2011).

3 Comparisons of prediction methods for the spontaneous combustion liability of coal

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

(1) *Differential scanning calorimetry (DSC)* Studies reported by Mahajan and Walker (1971) and Mohalik et al. (2009) used DSC to predict the spontaneous combustion liability of coal. This method determined the changes in energy inputs provided to a sample and a reference material with respect to temperature when these materials are kept at a constant temperature. The experimental investigations of this technique on coal spontaneous combustion liability were restricted to the evaluation of only four Indian coal samples (Mahajan and Walker 1971). This method indicated that there was no uniformity on the laboratory variables reported by different studies in carrying out the differential scanning calorimetry (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.

(2) *Thermogravimetric analysis (TGA)* TGA has been applied in the power industry to examine the reactivity of coal, obtain kinetics parameters (activation energy and pre-exponential factors) of fuel and to estimate physical characteristics such as ash, volatile matter, moisture, carbon content (Avila 2012; Elder 1983; Kizgut and Yilmaz 2004; Sima-Ella et al. 2005; Vaan Graan and Bunt 2016). Studies reported by Avila (2012) and William (1986) indicated the possibility to obtain reliable results with the use of different heating ramps during low-temperature oxidation, to estimate oxygen absorbed and released, amount of moisture and volatile matter contents and calculate the reactive or non-reactive constituents of coal. This method estimates the loss in weight of coal samples at variable temperatures due to self-heating. A specific weight of coal is heated through a fixed heating ramp and plotted against the time/temperature. The results are called 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.

(3) *Russian U index* This method estimates the amount of oxygen absorbed by individual coal samples over 24 h. 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 system is that the volume of oxygen absorbed by a coal 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.

(4) *Differential thermal analysis (DTA)* Reports by Whitehead and Breger (1950) and Mohalik et al. (2009) have used this technique to predict the propensity of coal towards spontaneous combustion. The method includes heating a small coal sample at a fixed 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 certain temperature and the properties of the material in question [Mohalik et al. (2009) and Whitehead and Breger (1950)]. It was found that there was no uniformity on the laboratory parameters selected in the use of this method.

(5) *Crossing point temperature (XPT)*: Many studies have used this method to determine the propensity of various coal towards spontaneous combustion with respect to their ignition temperature which is similar to the XPT (Gouws 1987; 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 inherent properties (moisture, ash, volatile matter and carbon contents) of coal and the design of the testing equipment. Barve and Mahadevan (1994) indicated a relationship between ash (dry), moisture and XPT as seen in Eq. (3) which is not in-line with Humphrey's opinion.

$$XPT = 168.8 - 10.3M + 0.12A + 0.69\,M^2 - 0.06MA + 0.01A^2 \tag{3}$$

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

(6) *X-ray diffractometer (XRD)* 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). The results obtained from this method were related with the results obtained from petrographic properties (macerals) and other thermal techniques to confirm the validity of this method. It was indicated that the variations in iron minerals within coal during the oxidation process may be caused by the catalytic reactions of the iron minerals with oxygen. This technique showed that pyrite among other mineral matter may be easily altered during coal oxidation and the oxidation alteration of pyrite vary with the forms of sulphur and its mineralogy (Ribeiro et al. 2016).

(7) *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 fine coal (0.4 g) is heated at a persistent rate in a quinolone steam bath with the flow of air through the coal pellet. The time against the temperature curve is logged up till a temperature of 235 °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 a relationship between the XPT, flammability temperature (FT), wet oxidation potential (WOE), proximate and ultimate analysis (Nimaje and Tripathy 2015, 2016; Nimaje et al. 2013).

(8) *Adiabatic calorimetric method* This technique 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 towards 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, to the extent that the heat is not dispersed 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 and the liability of coal towards spontaneous combustion was evaluated. 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.

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

(9) *Wits-Ehac test* This test has been used to forecast the propensity of coal and coal-shale towards spontaneous combustion (Eroglu 1992; Genc et al. 2018; Genc and Cook 2015; Gouws and Wade 1989a, b; Onifade et al. 2018a; Onifade and Genc 2018a, b, c, d, e, f; Wade 1989). The apparatus consists of an oil bath, six-cell assemblies, (three for the coal and 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 Fig. 3. Freshly pulverized (− 212 μm) dried coal of 20–25 g is used for each coal cell. The system incorporates the determined XPT and DTA from the coal with periodic measurement of
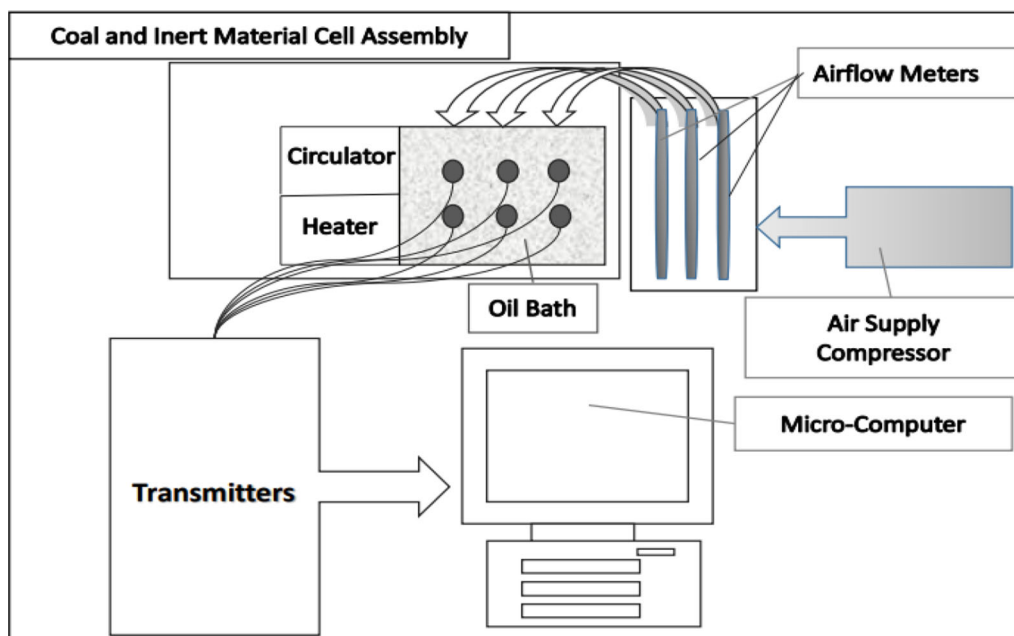
Fig. 3 Representation of the Wits-Ehac equipment (Wade et al. 1987)

temperatures to obtain a reliable spontaneous combustion liability index referred as the Wits-Ehac Index. The coal temperatures are recorded every 30 s by the computer for the oil in the bath to be heated to 200 °C. When using the DTA, the difference in temperatures between the coal and an inert material is measured through a data logger, stored in a computer and plotted against the temperature of the inert material. When the temperature difference between the inert material and the coal is plotted against the inert temperature, the part of the graph where the coal is heating faster than the inert material (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 is higher than the temperature of the coal (Stage I), which is dependent on the cooling effect of the evaporation of the coal moisture content. Secondly, during the evaporation of the moisture content, the coal 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 baseline and is referred to as the XPT. Uludag et al. (2001) reported 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. The index is calculated from the formula in Eq. (4). The thermogram in Fig. 4 illustrates the stages and the XPT for a given coal sample.

$$\text{Wits} - \text{Ehac Index} = (\text{Stage II slope}/\text{XPT}) * 500 \tag{4}$$

(10) *Wits-CT tests* This test was developed because the Wits-Ehac Index failed to provide results during the testing of some coal-shale samples because of their low liability towards spontaneous combustion. This test involves chemical reactions between coal and coal-shale and oxygen (Onifade et al. 2018a). A new index, referred to as the Wits-CT Index was developed (Onifade et al. 2018a). The liability of different samples to self-heat is evaluated for 24 h under the influence of oxygen. This experiment measures the temperature differences within the shortest period of time in a coal or coal-shale mass. The full Wits-CT testing experimental procedure is documented by Onifade et al. (2018a) and Onifade and Genc (2018e). An illustration of the experimental setup is indicated in Fig. 5. The index is
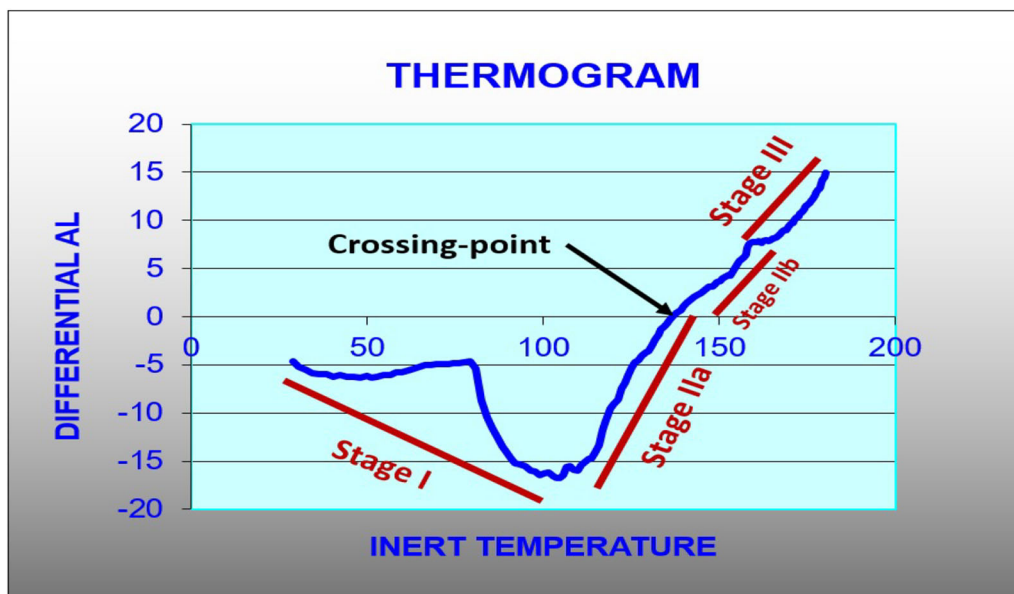
Fig. 4 A characteristic differential analysis thermogram of a coal (Onifade and Genc 2018e)
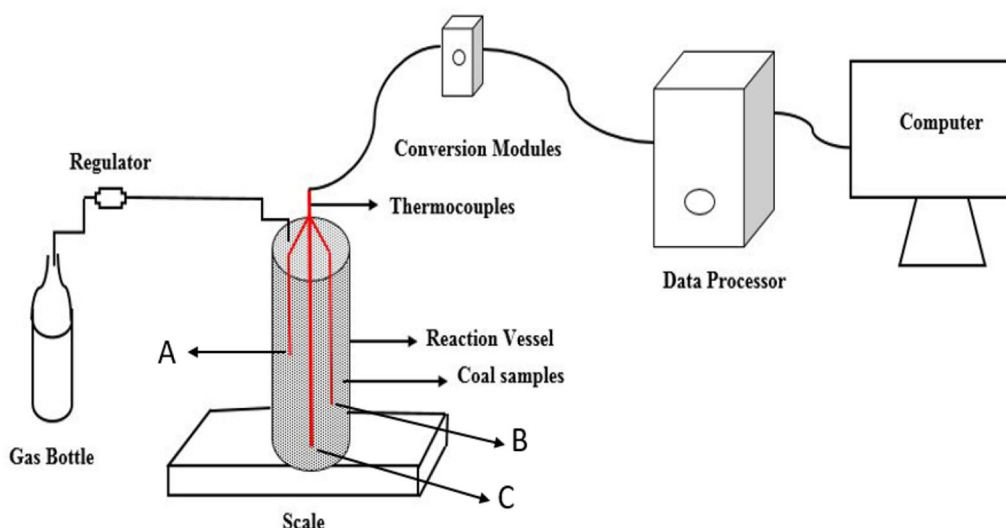

Fig. 5 Schematic of the Wits-CT apparatus (Onifade et al. 2018a; Onifade and Genc 2018e)

calculated from the formula in Eq. (5) Onifade et al. (2018a).

$$\text{Wits} - \text{CT Index} = (T_M/24 + T_R) * \%C_{ad} \qquad (5)$$

where: T_M is the difference between the sum of maximum temperatures of each thermocouple and room temperature (22 °C), T_R is the difference between the maximum temperature and initial temperature during oxidation reaction in degree Celsius, $\%C_{ad}$ is the air-dried percentage of carbon, * is a multiplication sign and 24 is the test duration and is constant.

The Wits-Ehac Index and the Wits-CT Index are used to predict the spontaneous combustion liability of coal and coal-shale in South Africa. The advantages of the Wits-Ehac Index includes a simple and cheap apparatus and rapid test time, while the disadvantages are that the insulation characterisics of the coal are not similar to the practical situation and it is difficult to recognize the ignition point. For the Wits-CT Index, the advantages are a simple and cheap apparatus, while the disadvantage is that due to the lentgthy test time (24 h), the rate of oxygen consumption is assumed as an indication of heat generation.

A number of studies have been conducted to predict the spontaneous combustion liability of coal and coal-shale using different experimental methods both in the laboratory and field and different countries of the world following different methods for this purpose. For example XPT in India, Russian U-Index in Russia, Olpinski Index in Poland, adiabatic calorimetry in USA, Wits-Ehac Index and Wits-CT Index in South Africa and R70 in NewZealand and Australia etc. These simple indices have traditionally been considered for the prediction of spontaneous combustion liability of coal. There is no agreement among researchers for the adoption of a particular method for the evaluation of spontaneous combustion liability of coal. Some of the researchers have suggested that a number of methods may be attempted to determine fairly accurately the degree of proneness of a particular coal to spontaneous combustion by using either the experimental or empirical approaches. Several laboratory techniques exist to establish the relative tendency of different coals to undergo spontaneous combustion. However, none is superior or in general use for predicting hazards due to the spontaneous combustion of coal.

One of the challenges in the coal value chain 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 associated coal-shale intrinsic properties between coal seams influencing spontaneous combustion have been evaluated. The accessibility of sufficient air in waste dump, spoil heaps, highwall, coal-shale and mined out areas where the coal has been left, particularly if it is loose coal (coal fragments larger in size than coal dust), can cause 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 the spontaneous combustion liability of coal under the effects of atmospheric conditions. Exposure of open-pit walls for long periods 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 (Banerjee 1985; Brooks et al. 1988a, b; 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 the 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.

This study established that the occurrence and development of spontaneous combustion is extremely complex and dynamic because of the various factors such as moisture, ash, volatile matter, carbon, sulphur, pyrite, mineral matter, petrographic properties, and mineral compositions. These factors have been the subject of many investigations. Relationships between coal properties and spontaneous combustion liability indices have been documented in previous works and discussed in this study. The major reasons for the difficulties in understanding the mechanism of spontaneous combustion is the presence of different internal and external factors affecting the self-heating and development of the phenomenon. Spontaneous combustion is not only influenced by the seam factor but by multiple factors such as geological factors and mining factors. It is important for the mine ventilation engineer to be conscious of the zones in which spontaneous combustion is most likely to occur. In underground mines, spontaneous combustion can occur along ventilation paths. Leakage can occur in rib fractures around ventilation stoppings, through cracks passing through a pillar or along the bed separation in coal left in the roof. Oxygen may be deposited on these areas where insufficient ventilation exists, resulting in adequate dispersion of heat from oxidisation.

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 environment. The most suitable approach to prevent this event is to have a better knowledge of the coal and its overlying materials. 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 damage to the surrounding areas. This is due to the frequent growth in the event of spontaneous combustion in the affected coal mines across the globe. 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.

Self-heating potentially leading to spontaneous combustion, continues to be a hazard that must be minimized by coal mine operators to create a safe work environment. This can be managed by introducing a spontaneous combustion management plan containing data that can be used to evaluate the intrinsic and extrinsic factors of coal to self-heat, i.e. obtaining results from both small-scale and large-scale tests. The spontaneous combustion liability of coal and related coal-shale is a major factor in evaluating how fires are spread in coalfields and has a direct bearing on the usage of fire control methods. These fires provide unsafe and environment hazards. Eliminating such fires by conventional methods is expensive, difficult and often ineffective because of the nature of coal and related coal-shale.

The relative propensity of coal to undergo self-heating can be established using different methods. These methods are well established in their usage but the fact that no specific test method has become a standard indicates that doubt still exists as to the validity of all of them. Therefore, it is recommended to carry out an extensive study on a group of coal and coal-shale from different countries and establish relationships between the results of the various spontaneous combustion test methods used in predicting their liability indices. It will be more interesting if the results of many of these spontaneous combustion test methods may be similar based on the actual mine conditions considering the effects of factors such as mining, geological and environmental conditions.

4 Conclusion

To provide a better understanding of the spontaneous combustion phenomena, the prediction of the spontaneous combustion liability in coal mines using reliable testing methods were reviewed, and relationships between the intrinsic and extrinsic properties of coal and coal-shale were established. Furthermore, it was found that the intrinsic and extrinsic factors of coal and coal-shale are the parameters that have been used to investigate the cause and predict spontaneous combustion incidents. This study showed that measured experimental factors can be linked to potential self-heating and indicated that self-heating is a very complex process than the differences in heat generation and heat loss rates. On the basis of the findings obtained from this review, in relation to spontaneous combustion, the influence of the gas content obtained during self-heating should be investigated.

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