Ash, Volatile Matter and Carbon Content Influence on Spontaneous Combustion Liability of Coal-Shales

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1 Introduction

The event of spontaneous combustion is one of the major problems in the coal value chain (Liu and Zhou 2010; Onifade and Genc 2018a, b; Phillips et al. 2011). It is well-known that coals and coal-shales undergo self-heating when exposed to oxygen in the air (Onifade and Genc 2018b, c; Onifade et al. 2018). The heat produced may become faster than the heat dissipated to the surrounding (Kim and Chaiken 1990; Dullien 1979). The first mechanism of spontaneous combustion involves heat produced during the physical and chemical processes, which play a significant role in the increase of coal temperature until the ignition point, and the second mechanism involves the reaction of oxygen with these materials either as a system of successive oxidation reactions or in the form of a single constant process (Onifade et al. 2018). Most of the heat transferred may be caused by conduction to surrounding strata (Akande and Onifade 2013; Akande et al. 2013). Spontaneous combustion of coal-shales and spoil heaps fire may result from the accumulation of heat generated and heat dissipated to the surroundings (Onifade and Genc 2018a; Phillips et al. 2011). Spontaneous combustion in waste dumps, spoil heaps and coal-shales is comparable to coal oxidation in which carbon oxide and carbon dioxide are produced (Onifade and Genc 2018b; Onifade et al. 2018; Kim and Chaiken 1990; Restuccia et al. 2017; Rumball et al. 1986).

Coals and coal-shales which contain variable proportions of organic matter (macerals) and inorganic materials (mainly crystalline) may undergo self-heating (Onifade and Genc 2018c; Restuccia et al. 2017; Rumball et al. 1986). This enables the rock to be porous to air, and with the increased surface area, the organic particles have reactive oxidation sites (Dullien 1979). The reasons for coal-shales to

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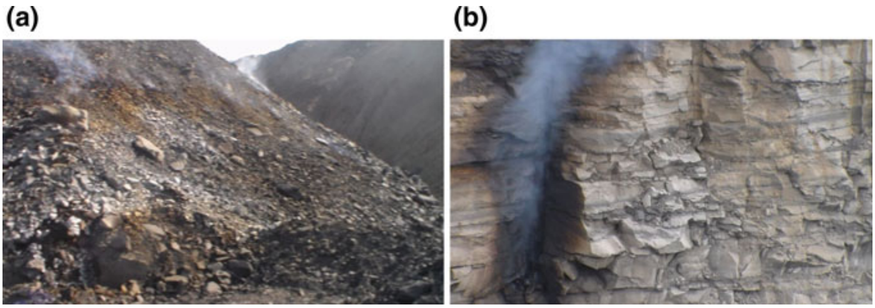

Fig. 1 **a** Burning spoil heaps at Tweefointein Mine and **b** Symptoms of shale self-heating at Goedgevonden Colliery, Witbank, South Africa (Onifade and Genc 2018b, c; Onifade et al. 2018)

undergo spontaneous combustion could be caused by the volume of pyrite, organic composition, nature and rank of the associated coal (Restuccia et al. 2017; Rumball et al. 1986; Mastalerz et al. 2010). Spontaneous combustion of coal-shales has been witnessed in Witbank coalfield, South Africa to cause self-heating in selected bands of coal seams (above and below), spoil heaps, waste dumps and highwalls (Onifade and Genc 2018b, c; Onifade et al. 2018) (Figs. 1 and 2). Coal seam and spoil heaps comprise of weathered coal, clays, pyritic shales, coal-shales, slate and other strata associated with the coal seam (Onifade and Genc 2018b; Onifade et al. 2018; Kim and Chaiken 1990; Restuccia et al. 2017; Rumball et al. 1986). Limited research has been carried out to understand the characteristics of coal-shales exposed to an oxidizing environment (Onifade and Genc 2018b, c,d; Onifade et al. 2018; Restuccia et al. 2017; Rumball et al. 1986). Many studies have been carried out on

Fig. 2 Symptoms of self-heating at mine face at Khwezela Mine (Bokgoni Pit), Witbank, South Africa (Onifade and Genc 2018b, c, d; Onifade et al. 2018)

spontaneous combustion liability of coals both experimentally and computationally (Genc et al. 2018; Genc and Cook 2015; Gouws and Wade 1989a, b; Onifade and Genc 2018; Stracher and Taylor 2004). However, no sufficient information has been reported on spontaneous combustion liability of coal-shales.

This paper presents the results of some intrinsic properties (ash, volatile matter and carbon content), and spontaneous combustion tests carried out on selected coal-shales in South African coal mines. This will be suitable to identify the influence of these intrinsic factors on spontaneous combustion liability and provides better understanding in predicting the initiation of coal-shale fires in coal mines.

2 Characterization and Spontaneous Combustion Tests

2.1 Sample Collection

The coal-shales used in this research were gathered (using ply sampling technique) from three different coal mines, South Africa, and to avoid further oxidation, collected samples were kept in airtight bags. Each ply sample bag was individually labelled. Six samples representative of in situ coal-shales were collected from areas known to be self-heating (highwalls and selected bands between coal seams). Characterization tests (proximate and ultimate analysis) and spontaneous combustion liability test (Wits-CT tests) were conducted on the coal-shales.

2.2 Sample Characterization

The sample lumps were reduced with the use of a crusher and ball mill to −250 µm for geochemical and −212 µm for spontaneous combustion tests. Similar preparation is related to studies reported by Onifade and Genc (2018b) and (c). The determination of the volatile matter, ash and carbon content of samples was carried out according to the American Society for Testing and Material Standards (ASTM) (ASTM, D-3173-17a 2017; ASTM, D-3174-11 2011; ASTM, D-3175-17 2017). Fixed carbon was obtained by subtracting the sum of the percentage of volatile matter, ash and moisture from 100. The carbon content was determined using a LECO TruSpec CHNS analyzer (Fig. 3) after calibration with sulfamethazine based on the International standard organization (ISO) (2001). The results pertaining to the coal-shales are discussed therein, and detail description on the characteristics of the samples is extensively described in the study by Onifade and Genc (2018b).

Fig. 3 LECO TruSpec CHN analyzer

2.3 Wits-CT Tests

The Wits-Ehac index was developed at the University of the Witwatersrand to measure the spontaneous combustion liability of coal. However, when testing some coal-shales, in some cases the Wits-Ehac index failed to produce results due to the low reactivity of the tested samples. To overcome this problem, a new equipment was developed to test coal and coal-shale undergoing chemical reactions with air, and a new index is obtained. This new index called the Wits-CT index (Onifade et al. 2018). The equipment has been used to test the spontaneous combustion liability of coals and coal-shales (Onifade and Genc 2018a, b, c, d; Onifade et al. 2018). The details of the experimental procedure are reported in study by Onifade et al. (2018). An illustration of the experimental setup is indicated in Fig. 4. The index is calculated from the formula in Eq. 1 (Onifade et al. 2018).

$$\text{Wits}-\text{CT Index} = (\text{TM}/24 + \text{TR}) * \%\,\text{Cad} \tag{1}$$

where: TM is the difference between the sum of maximum temperatures of each thermocouple in the autoclave and room temperature (22 °C), TR is the difference between the peak temperature and initial temperature during oxidation reaction in degree Celsius, %Cad is the air-dried percentage of carbon content of the sample, * is a multiplication sign, and 24 is the test duration and is constant.

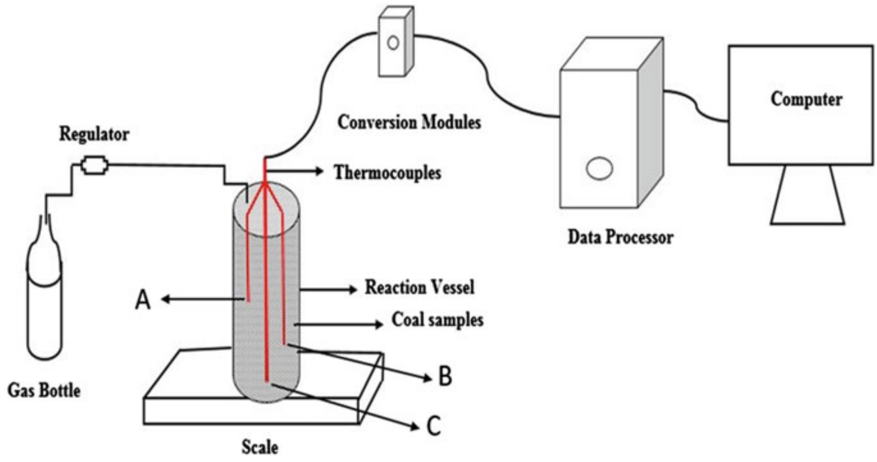

Fig. 4 Schematic of the Wits-CT apparatus (Onifade et al. 2018)

3 Results and Discussion

The results for the geochemical data (proximate and elemental analysis) and spontaneous combustion tests obtained from six coal-shale samples are presented in Table 1. The coal-shales are rich in mineral matter as indicated by the high ash content (Table 1). All samples are hence classified as carbonaceous shales (ash above 50 wt%). Sample SN has the lowest ash content, highest carbon content and highest spontaneous combustion liability index.

Table 1 Results of ash, volatile matter, carbon content (wt%, ad) and spontaneous combustion tests for coal-shale samples

Samples	Moisture	Ash	Volatile matter	Fixed carbon	Carbon	Wits-CT index
SH	0.8	88.7	8.5	2	2.66	0.27
SG	0.8	84.3	10.7	4.2	6.02	0.67
SA	1.4	78.5	11.2	8.9	11.5	1.33
SK	1	79.1	11.7	8.2	9.75	1.18
SM	0.8	76.9	11.7	10.6	12.5	1.44
SN	1.5	51.5	16.6	30.4	33.7	3.99

3.1 Results

The results of the selected intrinsic factors and spontaneous combustion tests carried out on the coal-shale samples are presented in Table 1. The ash content ranges between 51.5 and 88.7 wt%, respectively. The cause of the high ash content is due to the peat depositional environment where the condition of flooding of the paleomire occurred periodically during deposition. This is in line with the studies reported by Onifade and Genc (2018b), Restuccia et al. (2017), Rumball et al. (1986) and Mastalerz et al. (2010). It was observed that the ash contents vary from one another between bands of coal seams. Sample SN has the lowest ash content among the samples analyzed. The ash content of samples SH and SG is considerably high compared to the other samples. Sample SH with the highest ash contents has the lowest liability index, while sample SN samples with the lowest ash content has the highest spontaneous combustion liability index. The low and high liability index of the samples could be due to the influence of minerals absorbing heat within the coal-shales. Similar studies are reported by Onifade and Genc (2018b, d). It was found that the spontaneous combustion liability of coal-shale decrease with increasing ash content and vice versa. This characteristic is similar to those exhibited by coals in studies reported by Onifade and Genc (2018b, d). Therefore, coal-shale samples display similar ash behaviour as coal with respect to spontaneous combustion. The air-dried volatile content for the samples varies between 8.5 and 16.6 wt%, respectively. Sample with the highest volatile matter content has high spontaneous combustion liability index, while sample with low volatile matter has low liability index. Similar studies are reported by Onifade and Genc (2018b) and Restuccia et al. (2017). Coal-shale SN has the highest volatile matter content than the other samples. It was found that coal-shale with high volatile matter content is more liable to spontaneous combustion. This characteristic of coal-shale with high volatile matter content corresponding to high liability index is similar to coals with high volatile matter content (Onifade and Genc 2018b, d).

The determined air-dried carbon content varies between 2.66 and 33.7 wt%, respectively. Sample SH has the lowest carbon and the lowest liability index, while sample SN with the high carbon content has the highest spontaneous liability index. It was observed that the analyzed coal-shale samples have low carbon content when compared with some coals. Similar studies are reported by Onifade and Genc (2018b, d), Restuccia et al. (2017) and Rumball et al. (1986). It is more likely to suggest that the ability of coals, coal-shales and other carbonaceous materials to undergo self-heating is directly related to the amount of organic and inorganic matter present within them. If the self-heating rate of carbonaceous materials is related with the heat generated and heat dissipated from a single process, it is expected that the rise in temperature increases the reaction rate of that process. The carbon contents found in the coal-shales varied from one sample to the other. The results indicate that different reactions with different activation energies take place during spontaneous combustion. Coal-shale SH has the lowest Wits-CT, and coal-shale SN has the highest Wits-CT index among the coal-shale samples.

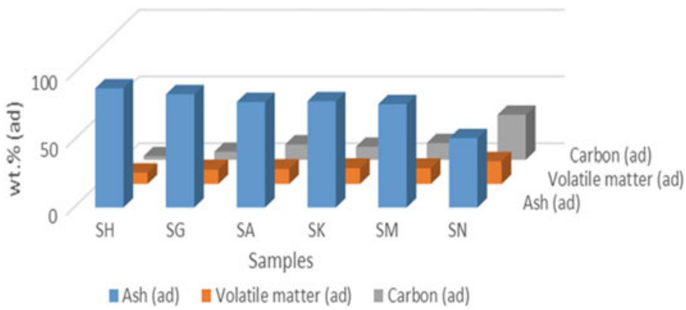

Fig. 5 Representation of ash, volatile matter and carbon content of coal-shale samples

The study shows that high-risk coal-shales have high Wits-CT value. The results show that the event of spontaneous combustion in coal mines is caused by a number of organic and inorganic constituents within coal-shale. Similar study is reported by Mastalerz et al. (2010). Coal-shales undergo self-heating when they absorb sufficient oxygen and moisture under the influence of atmospheric conditions. The heat generated due to the influence of oxygen exceeds the heat dissipated to the surrounding (via conduction, convection and radiation) and accumulates to cause spontaneous combustion (Onifade and Genc 2018b, d). Figure 5 shows that coal-shales with lower ash content, high carbon and high volatile matter are more liable to spontaneous combustion. Therefore, the influence of these factors may play a significant role in evaluating the occurrence of spontaneous combustion.

The decreasing ash content affects the self-heating potential by causing high spontaneous combustion liability index for the coal-shales as shown in Fig. 6a. Similar studies are reported by Onifade and Genc (2018c, d). Figure 6a shows that coal-shales with high ash content are showing low liability index, while coal-shales with low ash content seem to have high spontaneous combustion liability index. When the effect of the ash content is considered on linear regression analysis, it is negative on spontaneous combustion liability index. Similar studies are reported by Onifade and Genc (2018c, d). The spontaneous combustion liability for the coal-shales was found to increase with increasing volatile matter as shown in Fig. 6b. The results obtained from the linear regression analysis showed a positive effect on the self-heating potential. Therefore, as the volatile matter increases, the oxidation capacity of coal-shale increases in general. Similar studies are reported by Onifade and Genc (2018c, d). The sharp fall in the linear graph (Fig. 6b) could be caused by high mineral matter, which might act as heat absorbing capacity of the mineral within the coal-shale and hence suppress the self-heating rate. The increasing carbon content affects the self-heating potential of the coal-shales by causing a high spontaneous combustion liability as shown in Fig. 6c. The study indicated that as the carbon content increases, the spontaneous combustion liability

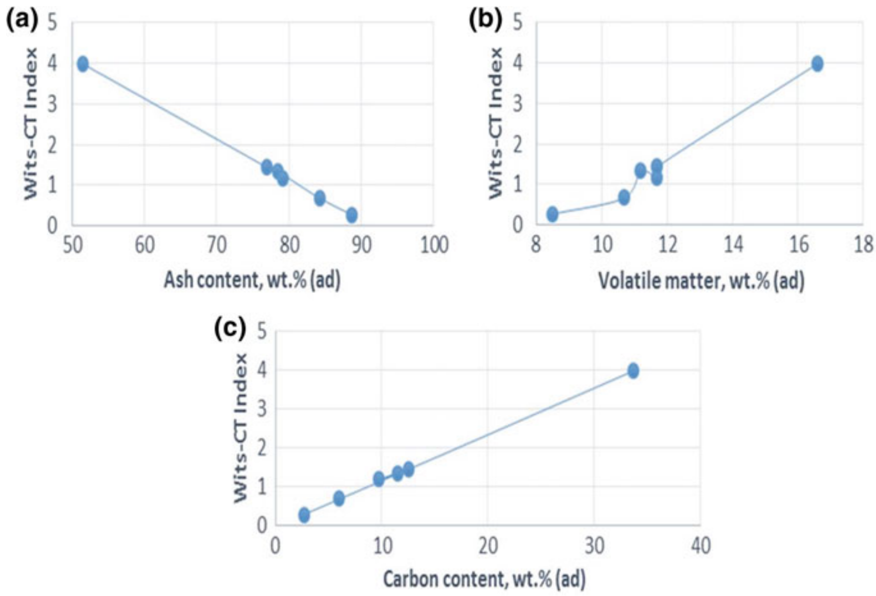

Fig. 6 Effects of **a** ash content **b** volatile matter content **c** carbon content on Wits-CT index

index of coal-shales increased. The results obtained from the linear regression analysis indicated a positive linear effect on the self-heating potential. Similar studies are reported by Onifade and Genc (2018c, d).

4 Conclusion

In this paper, the influence of carbon, volatile matter and ash contents on spontaneous combustion liability of coal-shales was investigated. The Witbank coal-shales were assessed using the Wits-CT spontaneous combustion liability index. It was found that ash shows a negative effect on the spontaneous combustion liability, while volatile matter and carbon showed positive effects with the self-heating potential. Coal-shales with high volatile matter and carbon content and low ash content are more liable to spontaneous combustion liability index and vice versa. The study indicated that the intrinsic properties evaluated affect spontaneous combustion liability of coal-shales and may be used as a reference in predicting their likelihood towards coal-shale spontaneous combustion.

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References

Akande, J.M., Onifade, M.: Modeling of Okaba underground coal mining ventilation system. Int. J. Eng. Technol. **3**(7), 766–772 (2013)

Akande, J.M., Onifade, M., Aladejare, A.E.: Determination of airflow distributions in Okaba underground coal mine. J. Min. World Expr **2**(2), 40–45 (2013)

ASTM, D-3174-11. Standard test method for ash in the analysis sample of coal and coke from coal and standard classification of coals by rank. ASTM International, West Conshohocken, PA (2011). www.astm.org

ASTM, D-3175-17. Standard test method for volatile matter in the analysis sample of coal and coke. ASTM International, West Conshohocken, PA (2017). www.astm.org

ASTM, D-3173-17a. Standard test methods for moisture in the analysis sample of coal and coke. ASTM International, West Conshohocken, PA (2017). www.astm.org

Dullien, F.: Porous Media Fluid Transport and Pore Structure, p. 79. Academic Press (1979)

Genc, B., Cook, A.: Spontaneous combustion risk in South African coalfields. J. South. Afr. Inst. Min. Metall. **115**, 563–568 (2015)

Genc, B, Onifade. M, and Cook.: A Spontaneous combustion risk on South African coalfields: part 2. In: Proceedings of the 21st International Coal Congress of Turkey 'ICCET' April 11–13, 2018, Zonguldak, Turkey, pp. 13–25 (2018)

Gouws, M.J., Wade, L.: The self-heating liability of coal: predictions based on simple indices. Mining Science and Technology **9**, 75–80 (1989a)

Gouws, M.J., Wade, L.: The self-heating liability of coal: predictions based on composite indices. Min. Sci. Technol. 9, 81–85 (1989b)

ISO 12902:2001: Determination of total carbon, hydrogen and nitrogen

Kim, A.G., Chaiken, R.F.: Relative self-heating tendencies of coal, carbonaceous shales and coal refuse. In: West Virginia, Paper presented at the 1990 Mining and Reclamation conference and Exhibition, Charleston, West Virginia, pp. 535–542 (1990)

Liu, L., and Zhou. F.B.: A comprehensive hazard evaluation system for spontaneous combustion of coal in underground mining. Int. J. Coal Geol. **82**, 27–36 (2010)

Mastalerz, M., Drobniak, A., Hower, J. C., O'keefe, J.M.K.: Spontaneous combustion and coal petrology. In: Stracher, G.B., Sokol, E.E., Prakash, A (eds) Coal and Fires: A Global Perspective. Coal-Geology and Combustion **1**, 47–62 (2010)

Onifade, M., Genc, B.: A review of spontaneous combustion studies—South African context. Int. J. Min. Reclam. Environ. 2–17 (2018a)

Onifade, M., Genc, B.: Spontaneous combustion of coals and coal-shales. Int. J. Min. Sci. Technol. (2018b) https://doi.org/10.1016/j.ijmst. 2018.05.013

Onifade, M., Genc, B.: Modelling spontaneous combustion liability of carbonaceous materials. Int. J. Coal Sci. Technol. (2018c). https://doi.org/10.1007/s40789-018-0209-2

Onifade, M., Genc, B.: Prediction of the spontaneous combustion liability of coal and coal-shale using statistical analysis. In: Society of Mining Professors, 6th Regional Conference, March 12–13, Johannesburg, South Africa, 2018, pp. 63–82 (2018d)

Onifade, M., Genc, B.: Establishing relationship between spontaneous combustion liability indices. In: Proceedings of the 21st International Coal Congress of Turkey 'ICCET' 11–13 April 2018, Zonguldak, Turkey, 1–11 (2018e)

Onifade, M., Genc, B., Carpede, A.: A new apparatus to establish the spontaneous combustion propensity of coals and coals. Int. J. Min. Sci. Technol. https://doi.org/10.1016/j.ijmst. 2018. 05.012 (2018)

Phillips, H., Chabedi, K., Uludag, S.: Best practice guidelines for South African Collieries, 1–129 (2011)

Restuccia, F., Ptak. N., Rein. G.: Self-heating behaviour and ignition of shale rock. Combustion and Flame **176** 213–219 (2017)

Rumball, J.A., Thomber, M.R., Davidson. L.R.: Study of chemical reactions leading to spontaneous combustion of pyritic black shale at MT Whaleback, Western Australia. In: Western Australia, Symposia series, Australasian Institute of Mining and Metallurgy, pp. 133–139 (1986)

Stracher, G.B., Taylor, T.P.: Coal fires burning out of control around the world: thermodynamic recipe for environmental catastrophe. Int. J. Coal Geol. **59**(1), 7–17 (2004)