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Comparative analysis of coal and coal-shale intrinsic factors affecting spontaneous combustion

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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-shales · Spontaneous combustion · Liability index · Statistical analysis and correlation coefficient

1 Introduction

Spontaneous combustion causes an increase in the temperature of a thermally segregated accumulation of coal or other combustible materials due to the chemical reactions between this material and oxygen (Davidson 1990). The low-temperature oxidation exists when the heat produced is absorbed by the surrounding environment (Kim and Sohn 2012). Spontaneous combustion of coal will eventually occur if nothing is done to minimize it (Onifade and Genc 2018c; Phillips et al. 2011). The liability of coal to spontaneous combustion is a function of the coal properties, geological, environmental and mining factors, which are in turn functions of various contributory factors (Smith and Glasser 2005).

The recent challenge faced by a number of coalfields in South Africa is spontaneous combustion of coals and coal-shales (Onifade et al. 2018; Onifade and Genc 2018d, e; Fig. 1), which frequently causes loss of revenue and at several intervals it has led to loss of precious resources, increase in production cost, loss of properties, and an increase in rehabilitation cost. Sedimentary rocks such as coals and coal-shales contain different volumes of organic and inorganic matter in which pore spaces are embedded in the solid together with carbon-rich matter (Alpern and de

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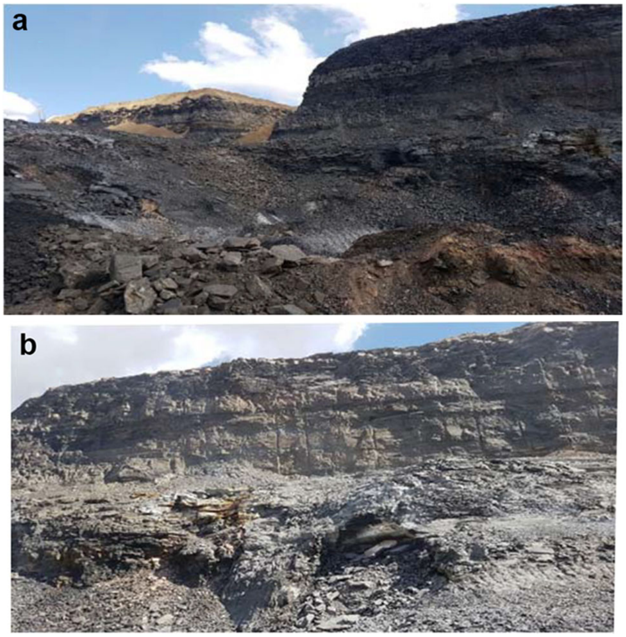

Fig. 1 **a** Spontaneous combustion of highwall and **b** effects of shale spontaneous combustion at at Khwezela Mine (Bokgoni Pit), Witbank, South Africa

Sousa 2002; Dullien 1979; Onifade and Genc 2018e). This renders the rock to be permeable to water and air, and with the increased surface area, the organic materials may have reactive oxidation sites (Dullien 1979). Studies reported by Mastalerz et al. (2010), Restuccia et al. (2017) and Rumball et al. (1986) indicated that for coal-shale to experience self-heating, it may contain varying proportions of sulphur (forms of sulphur), organic matter, reactive nature and rank of associated coal. Research on spontaneous combustion of coal has been examined by Beamish and Blazak (2005), Falcon (2004), Genc and Cook (2015), Gouws and Wade (1989a, b), Kaymakci and Didari (2002), Panigrahi and Sahu (2004), Panigrahi and Sexana (2001) and etc. However, a detailed investigation on the relationships between intrinsic properties of coals and coal-shales towards spontaneous combustion is limited. There is limited information to compare and contrast the intrinsic properties and spontaneous combustion liability of coal-shales in relation to coals (Onifade et al. 2018; Onifade and Genc 2018d, e). For this study, selected experimental tests on coals and coal-shales intrinsic properties (moisture, ash, volatile matter, ash, maceral compositions, total sulphur and forms of sulphur and etc.) were carried out according to the procedures of the American Society for Testing and Materials (ASTM) and International Organization for Standardization (ISO). A broad understanding of the inherent characteristic of coal-shales in relation to coal properties may be used to provide reliable information on the causes of spontaneous combustion of coals and coal-shales.

2 Materials and methods

2.1 Sample collection and preparation

Samples of coal and coal-shale from four open cast mines in the Witbank Coalfield, South Africa using the ply sampling method were experimentally examined. A full description of sample and collections and preparation for both petrographic and chemical analyses tests is extensively described in the studies reported by Onifade and Genc (2018d, e).

2.2 Wits-Ehac tests

The Wits-Ehac Index has been developed to measure the spontaneous combustion liability of coal since the late 1980s and has been widely used in South Africa [Eroglu (1992), Genc et al. (2018), Genc and Cook (2015), Gouws and Wade (1989a, b), Onifade et al. (2018), Onifade and Genc (2018a, b, d), Uludag et al. (2001) and Wade (1989)]. Full details of the Wits-Ehac experimental procedure (Fig. 2a) are extensively explained in the studies reported by Wade et al. (1987) and Onifade and Genc (2018e). The index is calculated from the formula shown in Eq. (1) and MS Excel is used to calculate the stages and generates the thermogram (Fig. 2b).

$$\text{Wits-Ehac Index} = (\text{Stage II slope/XPT}) * 500 \qquad (1)$$

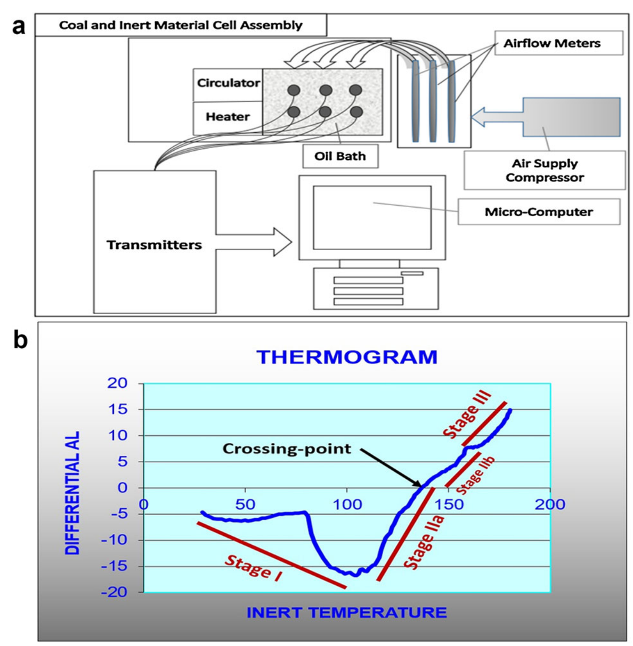

Fig. 2 **a** Schematic of the Wits-Ehac test apparatus setup (Wade et al. 1987). **b** Typical differential thermogram of a coal sample

3 Intrinsic properties and spontaneous combustion liability of coal and coal-shale samples

This study used the set criterion (Table 1) documented by Onifade and Genc (2018e) to evaluate and compare the linear relationships between the intrinsic properties and spontaneous combustion liability of coals and coal-shales. Full details of the statistical analysis is reported by Onifade and Genc (2018e). The data set is divided into dependent (Wits-Ehac Index) and independent (intrinsic properties obtained from proximate, ultimate, total and forms of sulphur and petrographic analysis) variables to enable simple interpretation and analyses. The R-squared values and the correlation coefficients were used to determine the trends of relationships between the intrinsic properties and the liability index (Tables 2, 3, 4, 5, 6).

The overall database involved the Wits-Ehac Index and intrinsic properties of 14 coals and 14 coal-shales. Data were analysed using the set criterion. Tables 2, 3, 4, 5 and 6 present the results of the linear regression analyses for both the coal and coal-shale samples.

The experimental data were analysed with the use of a linear regression analysis to establish whether any of the intrinsic properties were linearly correlated to the spontaneous combustion liability index. The relationships between independent and dependent variables using the correlation coefficients and R-squared values are seen in Tables 2, 3, 4, 5 and 6. The linear regression analysis identifies linear relationships between dependent and independent variables. The analysis of variable pairs (dependent and independent) indicated consistent trends, i.e. an increase in the liability index with increasing volatile matter and vice versa for these materials. Coals and coal-shales show inertinite as the major constituent among the maceral (Tables 3, 6). Weak linear relationships were noted between the Wits-Ehac Index and petrographic properties (total vitrinite, total inertinite, and total liptinite) for both coals and coal-shales (Table 7).

Table 1 Criterion for factors influencing spontaneous combustion liability of coals and coal-shales (Onifade and Genc 2018e)

Category	Criterion	Remarks
1	Correlation coefficient/R-squared value between 0.95 and 1 or − 0.95 to − 1	Variable indicate a perfect positive or negative linear relationship
2	Correlation coefficient/R-squared value between 0.51 and 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 and 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 and 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/R-squared value of zero	Variable indicate no linear relationship at all

Table 2 Relationships between independent (proximate and ultimate analysis, wt%-ad) and dependent variables for the coal samples

Independent variables	Range	Dependent variables	
		Correlation coefficients Wits-Ehac Index	R-squared values Wits-Ehac Index
Moisture	1.6–2.5	− 0.0637	0.0041
Volatile matter	16.7–26.9	0.5164	0.2666
Ash	13.7–48.4	− 0.6884	0.4739
Carbon	36.1–69.7	0.6572	0.4318
Hydrogen	2.55–4.21	0.6616	0.4377
Nitrogen	0.85–1.63	0.6945	0.4823
Oxygen	5.65–10.4	− 0.0686	0.0047
Sulphur	0.59–5.30	− 0.0115	0.0001
Pyritic sulphur	0.13–4.13	0.0869	0.0076
Sulphate sulphur	0.003–0.422	− 0.6422	0.4124
Organic sulphur	0.28–1.09	− 0.1787	0.0319

Table 3 Relationships between independent (petrographic analysis, vol%) and dependent variables for the coal samples

Independent variables	Range	Dependent variables	
		Correlation coefficients Wits-Ehac Index	R-squared values Wits-Ehac Index
Vitrinite and its group			
Total vitrinite	7.0–49.4	0.2591	0.0671
Total vitrinite (mmf)	7.9–60.0	0.0874	0.0076
Collotelinite	1.6–39.0	0.1481	0.0219
Collotelinite (mmf)	1.7–44.4	0.0439	0.0019
Collodetrinite	2.8–13.3	0.3944	0.1556
Collodetrinite (mmf)	3.1–18.8	0.1725	0.0298
Inertinite and its group			
Total inertinite	11.7–84.6	0.1679	0.0282
Total inertinite (mmf)	36.9–90.1	− 0.0735	0.0054
Fusinite	0.8–7.6	0.5663	0.3207
Fusinite (mmf)	1.2–12.1	0.4392	0.1929
Secretinite	0.8–6.0	0.1284	0.0165
Secretinite (mmf)	1.1–6.7	0.0064	0.0001
Reactive semifusinite	0.2–7.7	0.1993	0.0397
Reactive semifusinite (mmf)	0.3–8.4	0.1874	0.0351
Inert semifusinite	5.1–41.4	0.2518	0.0634
Inert semifusinite (mmf)	7.4–43.8	0.0882	0.0078
Total semifusinite	5.5–43.4	0.2898	0.0840
Total semifusinite (mmf)	14.1–45.9	0.1460	0.0213
Reactive inertodetrinite	0–4.6	0.1353	0.0183
Reactive inertodetrinite (mmf)	0–45.7	0.1232	0.0152
Inert inertodetrinite	3.2–47.1	− 0.1111	0.0123
Inert inertodetrinite (mmf)	10.0–50.9	− 0.3103	0.0963
Total inertodetrinite	3.2–49.4	− 0.0894	0.0080
Total inertodetrinite (mmf)	10–55.5	− 0.2768	0.0766
Liptinite and its group			
Total liptinite	0.6–3.8	0.0280	0.0008
Total liptinite (mmf)	0.6–4.4	− 0.2228	0.0496
Sporinite	0.4–3.4	0.2321	0.0539
Sporinite (mmf)	0.7–3.8	0.2116	0.0448

mmf mineral matter free basis

Table 4 Relationships between independent (total reactive maceral, total maceral and total mineral matter-vol%) and dependent variables for the coal samples

Independent variables	Range	Dependent variables	
		Correlation coefficients Wits-Ehac Index	R-squared values Wits-Ehac Index
Total reactive maceral	11.2–53.0	0.3148	0.0991
Total reactive maceral (mmf)	12.8–64.4	0.1229	0.0151
Total maceral	31.7–94.6	0.3785	0.1433
Total mineral matter	5.4–68.4	− 0.3782	0.1430

Total reactive is the sum of total vitrinite, total liptinite, reactive semifusinite and reactive inertodetrinite

Table 5 Relationships between independent (proximate and ultimate analysis, wt%-ad) and dependent variables for the coal-shale samples

Independent variables	Range	Dependent variables	
		Correlation coefficients Wits-Ehac Index	R-squared values Wits-Ehac Index
Moisture	0.8–1.7	0.7715	0.5952
Volatile matter	8.5–16.6	0.6389	0.4082
Ash	51.5–88.7	− 0.8352	0.6975
Carbon	2.66–33.7	0.7962	0.6339
Hydrogen	0.75–2.87	0.5795	0.3358
Nitrogen	0.08–0.96	0.6446	0.4155
Oxygen	5.01–11.85	− 0.3212	0.1031
Total sulphur	0.12–6.90	0.5791	0.3353
Pyritic sulphur	0.04–4.26	0.5704	0.3254
Sulphate sulphur	0.003–0.45	0.5365	0.2878
Organic sulphur	0.05–2.19	0.5933	0.3519

Table 6 Relationships between independent (petrographic analysis-vol%) and dependent variables for the coal-shale samples

Independent variables	Range	Dependent variables	
		Correlation coefficients Wits-Ehac Index	R-squared values Wits-Ehac Index
Vitrinite and its group			
Total vitrinite	0.4–8.4	0.1230	0.0151
Total vitrinite (mmf)	2.4–39.3	− 0.2632	0.0693
Collotelinite	0–4.2	0.2206	0.0487
Collotelinite (mmf)	0–19.6	− 0.1073	0.0115
Collodetrinite	0–2.6	0.2105	0.0443
Collodetrinite (mmf)	0–12.1	− 0.2330	0.0543
Inertinite and its group			
Total inertinite	4.9–46.1	0.7360	0.5418
Total inertinite (mmf)	44.9–91.7	0.2143	0.0459
Fusinite	0–4.0	− 0.4038	0.2194
Fusinite (mmf)	0–16.9	− 0.5898	0.3479
Secretinite	0–1.8	0.6003	0.3604
Secretinite (mmf)	0–4.8	0.3304	0.1092
Reactive semifusinite	0–1.1	− 0.2795	0.0781
Reactive semifusinite (mmf)	0–4.6	− 0.3862	0.1492
Inert semifusinite	0.2–8.0	0.7688	0.5910
Inert semifusinite (mmf)	1.3–40.7	0.2970	0.0882
Total semifusinite	0.2–8.0	0.7776	0.6046
Total semifusinite (mmf)	1.3–40.7	0.2970	0.0882
Reactive inertodetrinite	0–3.6	0.3292	0.1083
Reactive inertodetrinite (mmf)	0–18.5	− 0.0829	0.0069
Inert inertodetrinite	1.9–34.8	0.7460	0.5565
Inert inertodetrinite (mmf)	14.8–76.6	0.2480	0.0615
Total inertodetrinite	3.3–35.2	0.7749	0.6005
Total inertodetrinite (mmf)	25.2–80.5	0.2367	0.0560
Liptinite and its group			
Total liptinite	0.4–5.5	0.3360	0.1129
Total liptinite (mmf)	2.6–25.0	− 0.0113	0.0001
Sporinite	0.4–5.5	0.3463	0.1199
Sporinite (mmf)	2.6–25.0	0.0059	0.0001

Table 7 Relationships between independent (total reactive maceral, total maceral analysis and total mineral matter-vol%) and dependent variables for the coal-shale samples

Independent variables	Range	Dependent variables	
		Correlation coefficients Wits-Ehac Index	R-squared values Wits-Ehac Index
Total reactive maceral	1.6–12.8	0.2923	0.0850
Total reactive maceral (mmf)	9.6–63.9	− 0.2552	0.0651
Total maceral	9.9–53.1	0.7653	0.5857
Total mineral matter	46.9–90.1	− 0.7653	0.5857

From Tables 2, 3, 4, 5 and 6, according to the criterion set, contents of volatile matter, 0.5164; ash, − 0.6884; carbon, 0.6572; hydrogen, 0.6616; nitrogen, 0.6945; sulphate sulphur, 0.6422; and inertinite macerals-fusinite, 0.5663; with strong effects on self-heating potential are factors affecting spontaneous combustion liability of coals, while contents of moisture, 0.7715; volatile matter, 0.6389; ash, − 0.8352; carbon, 0.7962; hydrogen, 0.5795; nitrogen, 0.6446; total sulphur, 0.5791; and its forms [pyritic, 0.5704; sulphate, 0.5365; and organic sulphur, 0.5933], total inertinite, 0.7360; and its constituents [fusinite, mmf, 0.5898; total semifusinite, 0.7776; secretinite, 0.6003; and

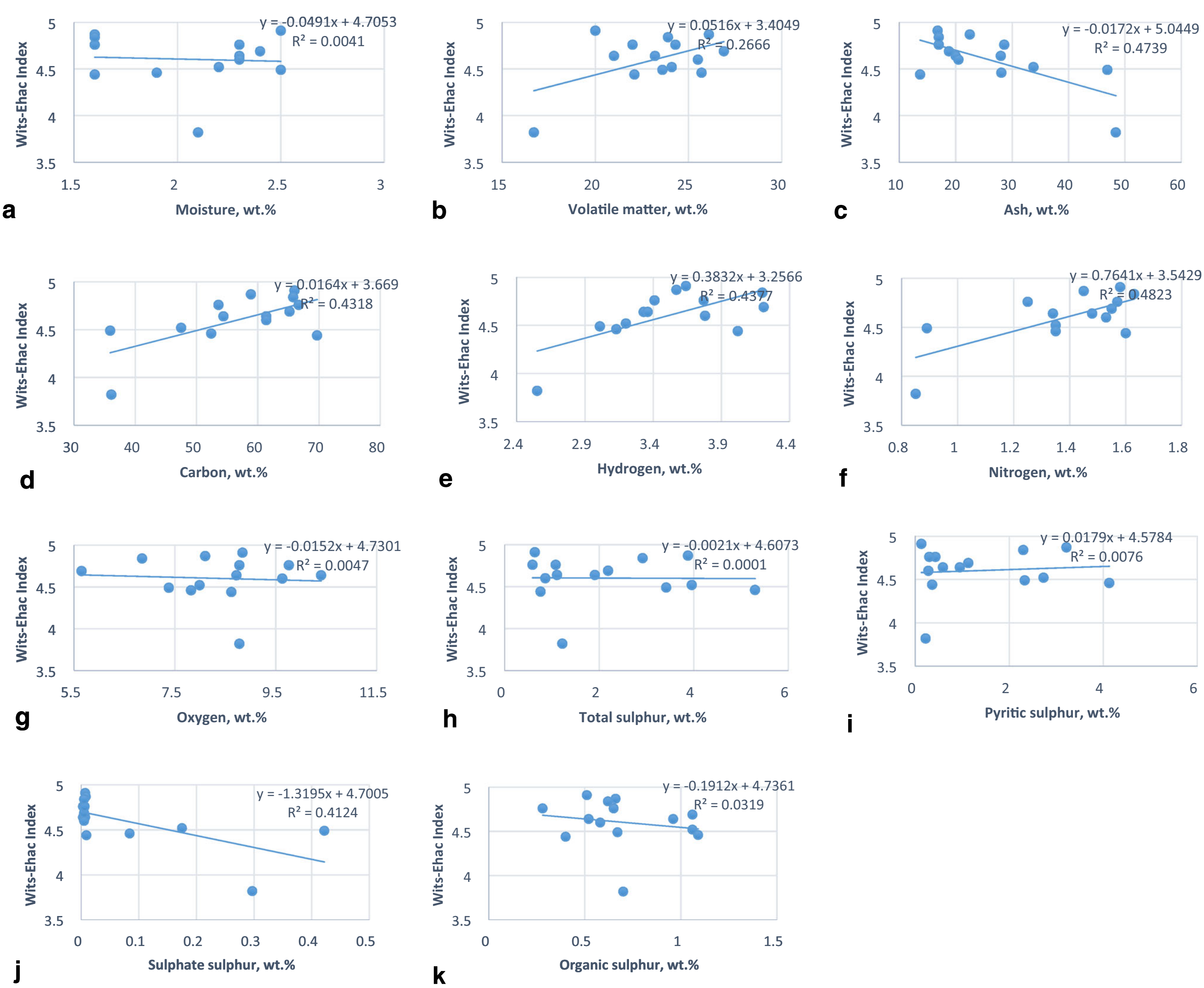

Fig. 3 Influence of proximate and ultimate analysis on spontaneous combustion liability of coals

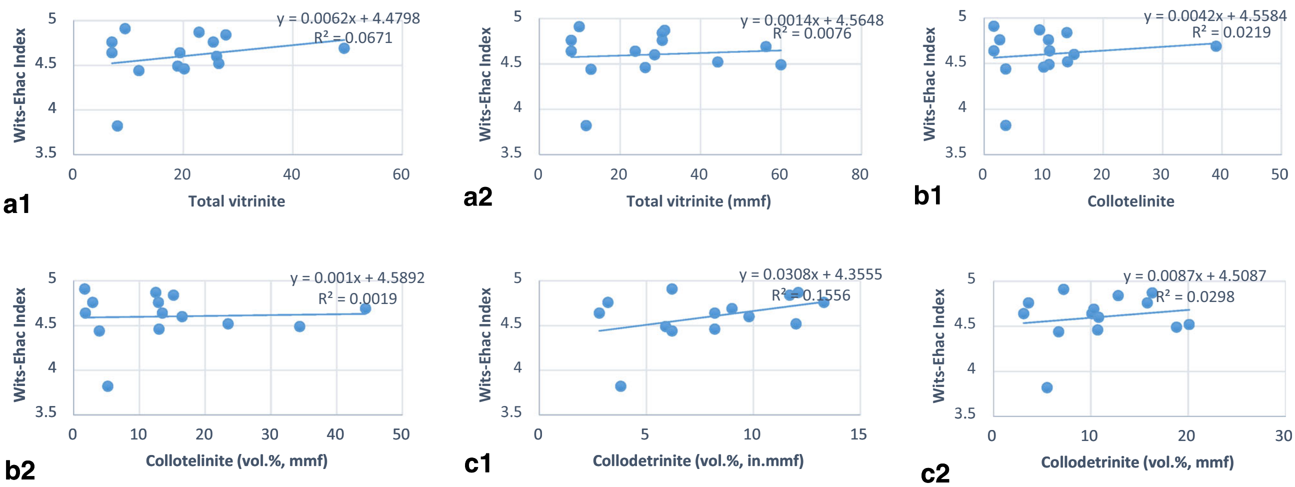

Fig. 4 Influence of vitrinite and its group on spontaneous combustion liability of coals

total inertodetrinite, 0.7749; total maceral, 0.7653; and mineral matter, − 0.7653] with strong effects on self-heating potential are factors affecting spontaneous combustion liability of coal-shales. Ash, some macerals and mineral matter contents for both coals and coal-shales show negative trends. Coal-shale intrinsic properties show better linear relationships to spontaneous combustion liability than the coals and hence, identifies the intrinsic properties influencing these materials toward spontaneous combustion. Despite the low contents of moisture, volatile matter, carbon, hydrogen, nitrogen and total sulphur in coal-shales compared to coals, the coal-shales shows significant correlation to the spontaneous liability index than the coals. The influence of intrinsic properties on spontaneous combustion liability of coals and coal-shales using statistical analysis has been extensively reported in a study by Onifade and Genc (2018e). The study created models which combined the effects of the main intrinsic properties affecting spontaneous combustion liability of these materials for predictive purposes (Onifade and Genc 2018e).

From Figs. 3, 4, 5, 6, 7, 8, 9 and 10, there is an increase in contents of moisture, volatile matter, ash, carbon, total sulphur, calculated oxygen, pyritic sulphur, organic sulphur, inertinite macerals and mineral matter in both coals and coal-shales. However, this appears to be more noticeable for coal-shales than for the coals, while coals seem to be more distinct in terms of hydrogen, nitrogen and sulphate sulphur content than coal-shales. It was found that spontaneous combustion liability of coals and coal-shales could be affected by varying proportions of one or more intrinsic properties.

4 Conclusion

This study has evaluated, compared and identified the relationships between intrinsic properties and spontaneous combustion liability of coals and coal-shales. The influence of selected intrinsic properties towards spontaneous combustion liability indicated a better linear relationship for the

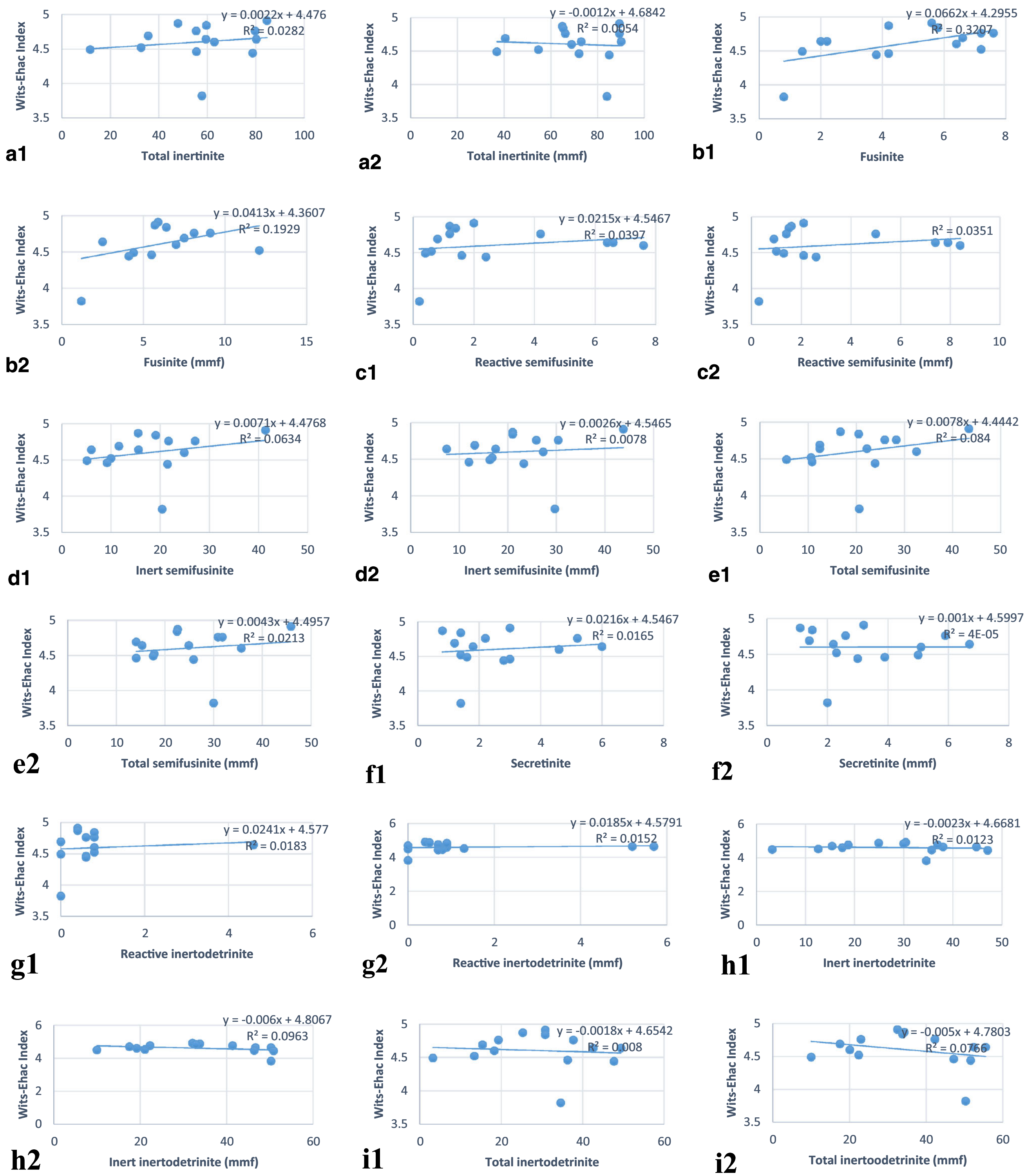

Fig. 5 Influence of total inertinite and its group on spontaneous combustion liability of coals

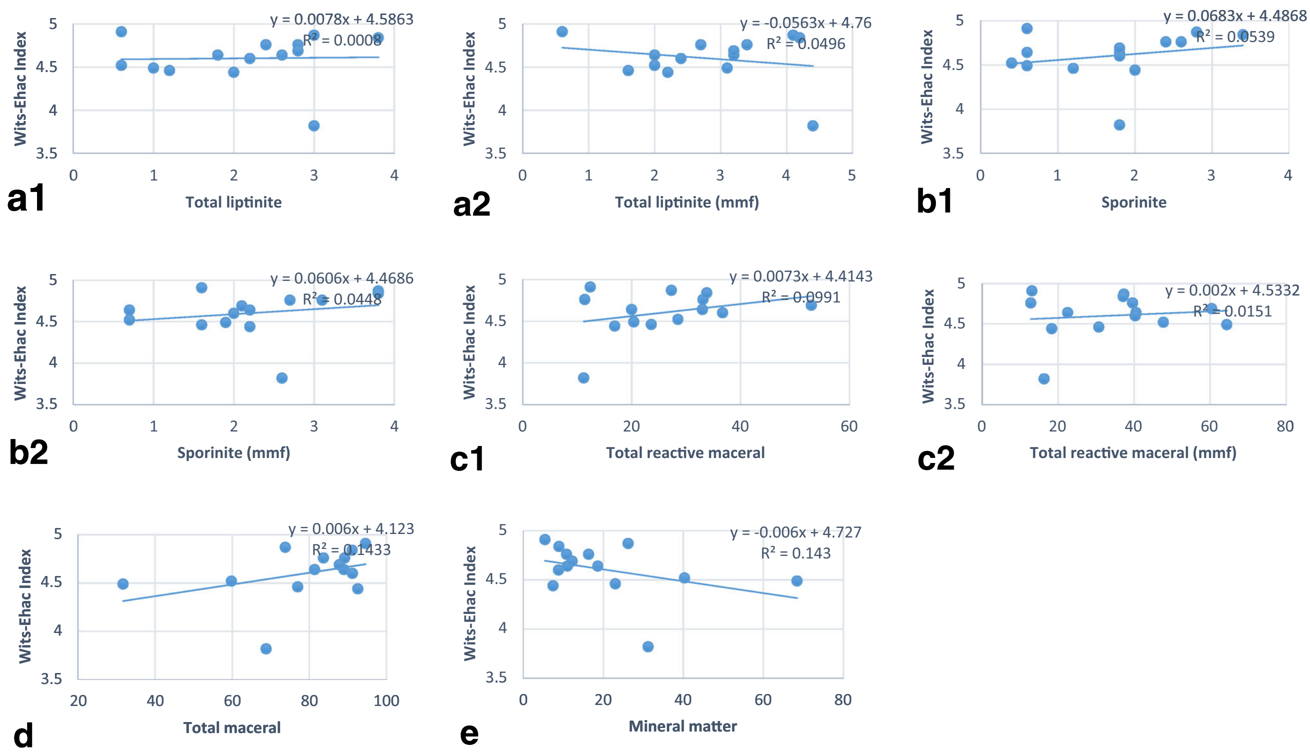

Fig. 6 Influence of total liptinite and its group, total reactive maceral, total macerals, and mineral matter on spontaneous combustion liability of coals

coal-shales than the coals, thus they may have a greater effect to cause spontaneous combustion of coal-shales. The linear regression analysis shows that among the macerals, the inertinite macerals indicated a stronger linear relationship to spontaneous combustion liability. Thus, the spontaneous combustion liability index of coals and coal-shales may be influenced by the proportion of each maceral composition. A definite positive or negative correlation coefficient exists between the intrinsic factors and spontaneous combustion liability index. This paper has established a comparative analysis between the dependence of spontaneous combustion liability index on intrinsic properties of selected coal-shales and associated coals. The results obtained from the petrographic and chemical analyses may be used as a tool to predict spontaneous combustion liability and may serve as of reference when comparing characteristics of coals and coal-shales from different coalfields.

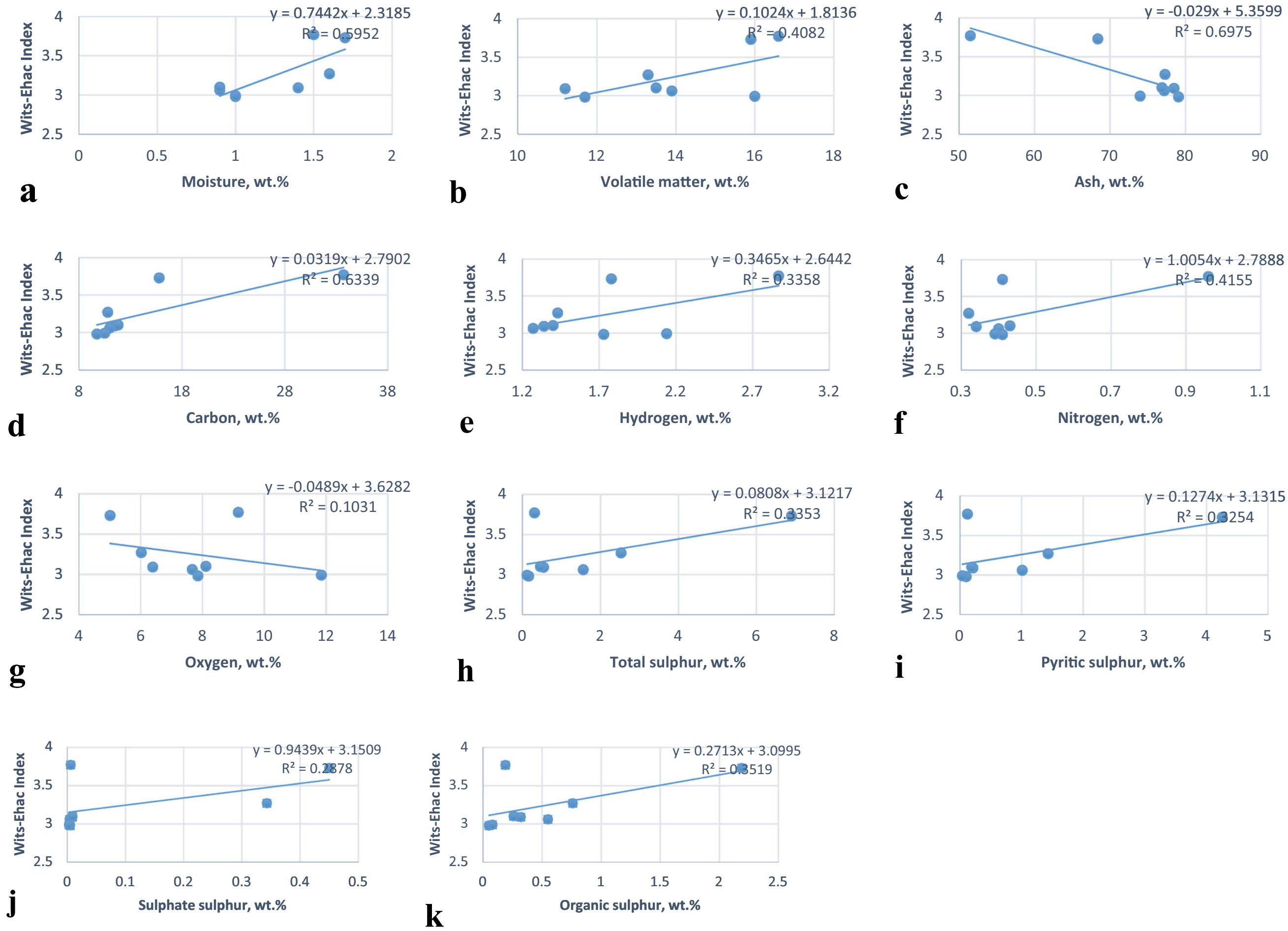

Fig. 7 Influence of proximate and ultimate analysis on spontaneous combustion liability of coal-shales

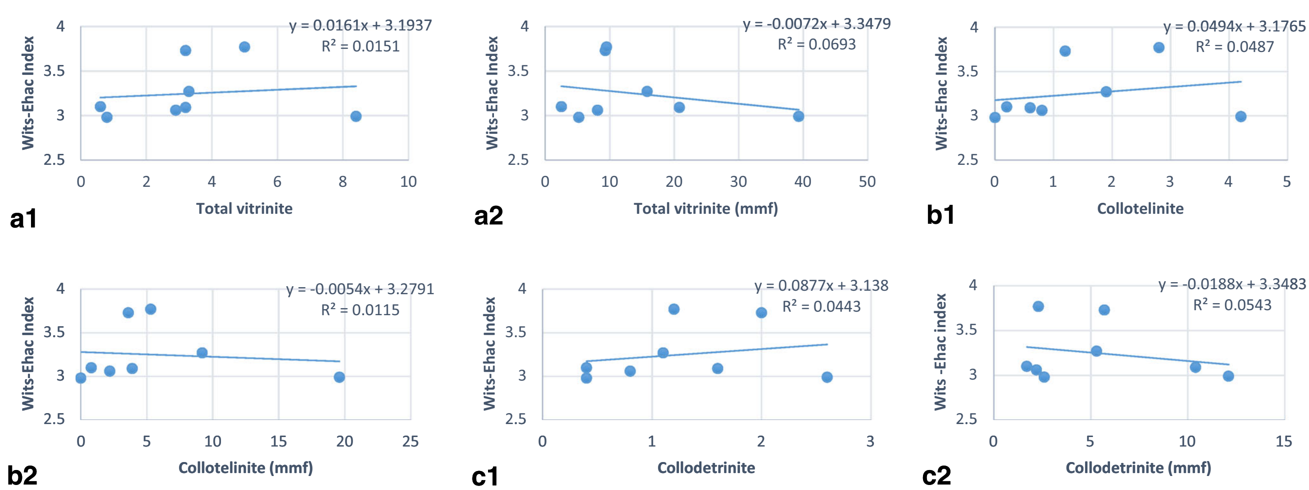

Fig. 8 Influence of total vitrinite and its group on spontaneous combustion liability of coal-shales

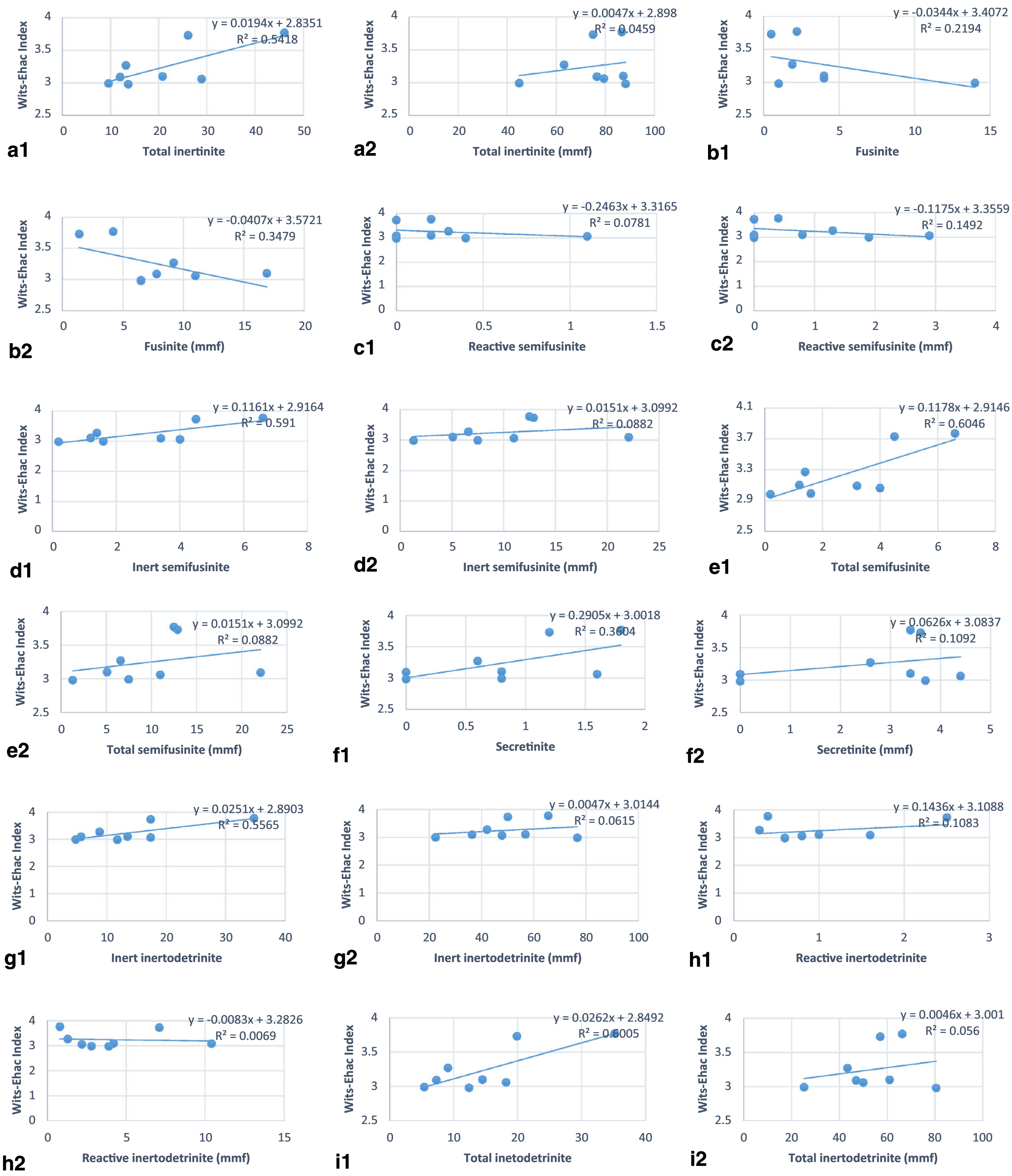

Fig. 9 Influence of total inertinite and its group on spontaneous combustion liability of coal-shales

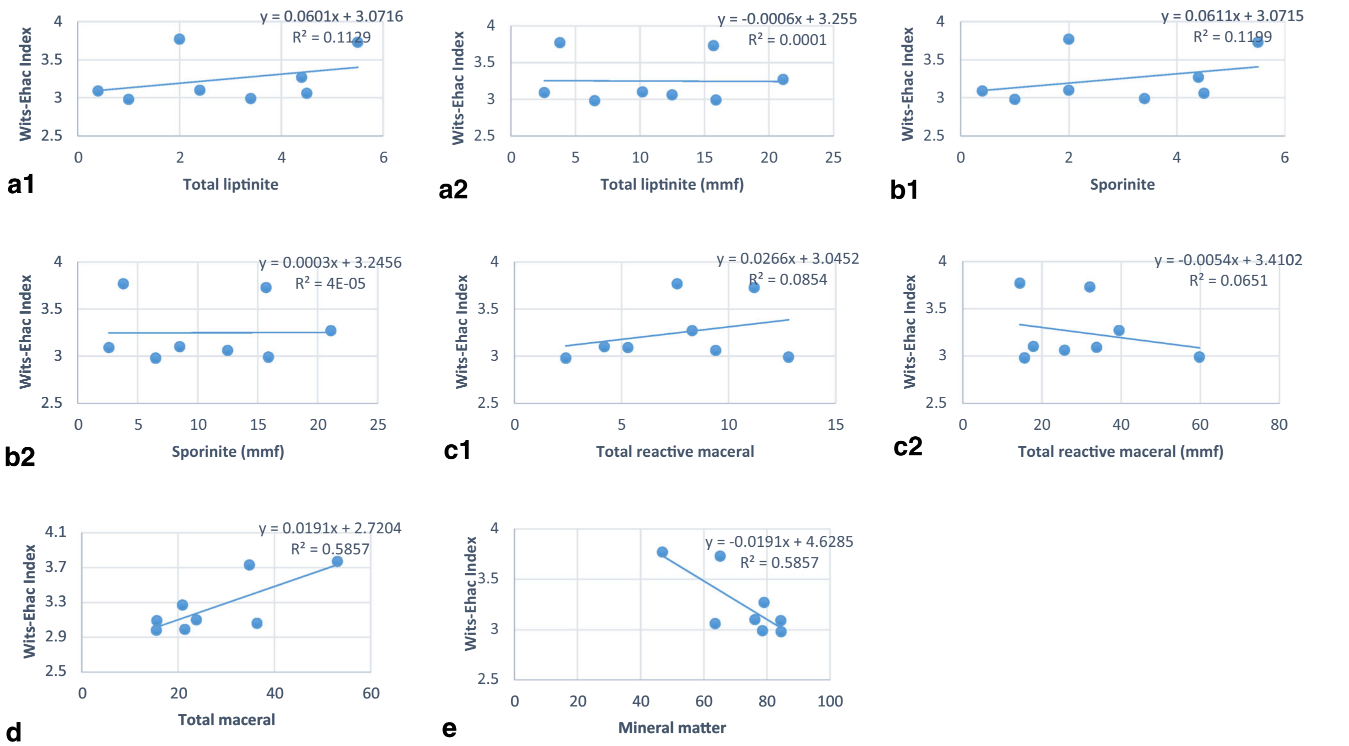

Fig. 10 Influence of total liptinite, sporinite, total reactive macerals, total macerals, and mineral matter on spontaneous combustion liability of coal-shales

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