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Effect of maceral content on tendency of spontaneous coal combustion using the R70 method

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ABSTRACT

Spontaneous coal combustion (SCC) is one of the significant hazardous phenomena in underground coal mines. The tendency of spontaneous coal combustion is an intrinsic property, due to the presence of the maceral content. Unlike its importance, this matter has not been discussed in detail among the researchers. Therefore, it is necessary to investigate the effect of this parameter on SCC. Maceral content is defined by the original vegetation from which coal is formed. The present study examines the role of maceral content on SCC, based on 51 coal samples with different maceral contents. These samples were collected from several Iranian underground coal fields, and the R70 test was carried out on each coal sample. By examining the results and comparing the R70 values, it was found that with an increase in the vitrinite and liptinite contents and a decrease in the inertinite content, the coal samples showed to have more tendency of spontaneous combustion.

Keywords : *Maceral Content, R70 Test, Spontaneous Coal Combustion*

1. Introduction

Coal is a profitable fossil fuel, which is the major, plentiful, and economical fuel around the world and is considered as the primary energy source in many countries. Coal has a fundamental role in the development and stability of the world economy [1, 2].

Unlike its benefits, coal mining is a complex set. The harsh working situation and the hazardous environment are the most critical parameters that affect a coal mining system.

The spontaneous coal combustion (SCC) is a significant hazard in coal industries. SCC is responsible for endogenic coal fires and is a significant safety point in the coal industry [3-8]. This phenomenon threatens the environment, economy, and society [9-11], which can shift to severe problems in the sustainable development of coal mines throughout their operational lifespan. Thus, this phenomenon is an urgent problem that should be resolved [1].

SCC is an oxidation process that occurs without the involvement of an external heat source, which can be due to several complicated exothermal processes [5, 12]. Approximately, all types of coal cause spontaneous combustion. The intrinsic characteristics of coal are believed to be the primary triggers of this phenomenon [13, 14].

This phenomenon remodels the internal heat characteristics of coal and can progress with the temperature. In the long run, this event may lead to burning the surrounding coals [3, 15-19].

The oxidation mechanism in this event results in releasing heat, and if allowed growing, the rate of oxidation would continually increase and would form an open fire [20-25].

Coal oxidation requires physical (Equation (1)) and chemical (Equation (2)) adsorption of oxygen, which forms oxidized surfaces (Equations (3) to (5)) under specific situations, as shown in Fig. 1. Firstly, these events occur in macropores and mesopores that allow the free expansion of oxygen [18, 22]. This process exhales greenhouse gases (CO_2 and CH_4), toxic gases (e.g., CO, H_2S, NO_x, N_2O, SO_2) and some trace elements (e.g., As, F, Se, Hg), which can seriously threaten the environment [26-29].

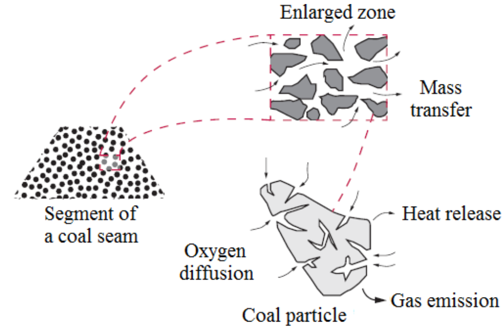

Fig. 1. Schematic illustration of the coal oxidation process.

$$\text{Coal} + O_2 \rightarrow O_2 \text{ (physical adsorption)} \quad (1)$$

$$O_2 \text{ (physical adsorption)} \rightarrow O_2 \text{ (chemical adsorption)} \quad (2)$$

$$O_2 \text{ (chemical adsorption)} \rightarrow \text{Surface oxide} \quad (3)$$

$$\text{Surface oxide} \xrightarrow{\text{primary process}} CO_2 + H_2O \quad (4)$$

$$\text{Surface oxide} \xrightarrow{\text{secondary process}} CO, C_nH_m, H_2 \quad (5)$$

The mechanisms of SCC have been described and discussed by several authors including Banerjee, (1985) [20], Kaymakci and Didari (2001) [30], Stracher and Taylor (2004) [31], Stracher et al. (2004) [32], Smith and Glasser (2005a,b) [33, 34] and Chatterjee (2006) [35].

The most important hazards created by this phenomenon are listed below, and some of which are shown in Figure 2 [9, 13, 14]:

- Climate change and global warming;

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- Personnel casualties;
- Emission of greenhouse gases (CO_2 and CH_4) along with SO_x, NO_x, H_2S, and CO;
- Costs of medicinal and recovery measures.
- Damage to equipment;
- Disturbance in families and communities;
- Loss of reputation and market position;
- Loss of production;
- Loss of valuable energy resources;
- Mine closure;
- Sir, water (surface and groundwater), and soil pollution near the burning coal field;
- Subsidence.

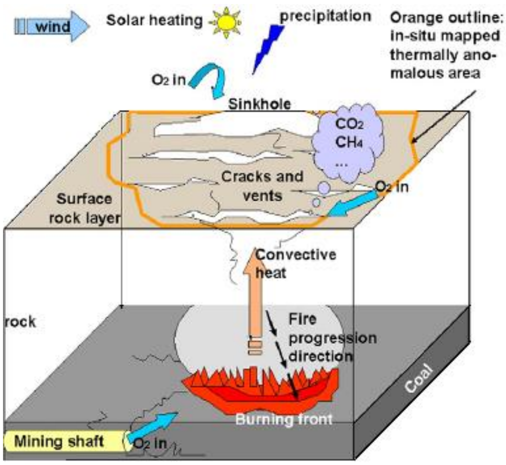

Fig. 2. Different aspects of pollution due to spontaneous coal combustion [36].

The occurrence of SCC is inevitable in coal industries due to the intrinsic characteristics of coal. Coal is a complex substance, mainly composed of materials called *macerals* that are the original vegetation from which coal is formed. There are three main groups of macerals, including vitrinite, liptinite, and inertinite (Figure 3). These three main groups have different chemical compositions due to their different sources. Vitrinite comes mainly from the leaves and stems of land plants and has more oxygen than the other two groups; liptinite comes from algae, pollens, resins, and cuticles and has more hydrogen; and inertinite has more carbon and is formed by the oxidation of vitrinite [37, 38].

The maceral content has a vital role in this phenomenon. In particular, the influence of the maceral content of coal that leads to spontaneous combustion has remained unclear. The maceral content of coal influences coal oxidation, and there are opposite effects of the maceral content on SCC. Humphreys' (1979) [38] tests indicated that the tendency of spontaneous combustion increases as the vitrinite content increases; Cotterell (1997) [37] did not find a correlation between maceral and spontaneous combustion tendency. Arisoy (2010) [39] claimed that the maceral content is not an effective parameter for estimating the liability of SCC.

The results of these studies are contradictory, showing the complex influence of the maceral content on the tendency of spontaneous coal combustion. Therefore, a comprehensive study is required to evaluate the effect of maceral content on this issue. In this study, 51 coal samples with different maceral contents of several Iranian underground coal fields (Tabas Parvadeh coal mines, Eastern Alborz coal mines, Central Alborz coal mines, and Kerman coal mines) were collected, and the R70 test was carried out on each coal sample.

2. Maceral content

Petrographic analysis of macerals provides essential data about the textural characteristics of coal types and is useful for the evaluation of SCC. Van Krevelen (1961) [40] classified the macerals used extensively by organic geochemists, as shown in Figure 3. This figure shows the differences between macerals, the variation within macerals, and the changes in their composition as the coalification progresses.

As seen in Figure 3, liptinite and vitrinite have a high H/C and O/C ratio in comparison to inertinite, respectively. The presence of H and O in coal accelerates the tendency of spontaneous combustion of coal. So, the existence of liptinite and vitrinite in coal samples is responsible for the spontaneous combustion of coal.

The oxidation process first attacks the non-aromatic part of coal molecules [41]. The non-aromatic parts are highly reactive constituents in low-grade coals and are described by high proportions of vitrinite and liptinite. Vitrinite possesses more non-aromatic parts in its molecular structure than inertinite. So, oxygen is combined to the molecular structure more extra per unit area and with lower activation energies, hence influencing to faster forms of oxidation at a lower temperature in preliminary steps than inertinite of the same grade [42].

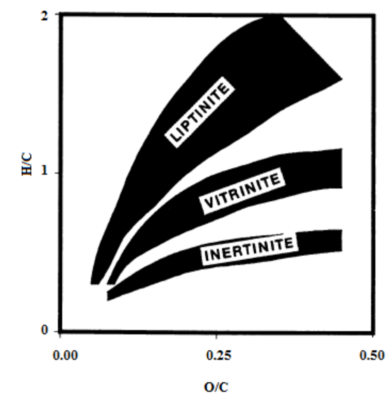

Fig. 3. Approximate bands for the three main maceral groups [40].

- ***Inertinite***

This group of macerals has higher carbon and lower oxygen and hydrogen contents compared to vitrinite. The H/C and O/C ratios in this group is low in comparison to liptinite and vitrinite (Figure 3) [43, 44].

So, coals with high inertinite contents are not susceptible to spontaneous combustion. As the inertinite content in the coal sample increases, the susceptibility of these coals to spontaneous combustion decreases.

- ***Vitrinite***

Humification and gelification are the most important processes in the formation of this group of macerals. Humification affects continuous oxidation slowly, which may be accelerated by the addition of oxygen [45, 46]. As a result, coals with high vitrinite contents are highly susceptible to spontaneous combustion when oxygen reaches these groups of macerals. Moreover, these groups of macerals have high H/C and O/C ratios (Figure 3) [44], in which the existence of H and O in coal accelerates the tendency of coal to spontaneous combustion. Previous studies have shown that as the vitrinite content in a coal sample increases, the susceptibility of these coals to spontaneous combustion increases.

- ***Liptinite***

This group of macerals comes from the relatively hydrogen-rich plant material and has high H/C and O/C ratios (Figure 3) [44], in which the existence of H and O in coal accelerates the spontaneous combustion of coal. So, coals with high liptinite contents are susceptible to spontaneous combustion. As the liptinite content in coal increases, the susceptibility of these coals to spontaneous combustion increases.

3. Materials and method

3.1. Coal samples and laboratory studies

The R70 test has been successfully used for the assessment of the tendency of spontaneous coal combustion. In this study, 51 coal samples were collected, on which laboratory tests (Maceral content and R70 test) were implemented. Fresh coal samples were collected directly from the operating workshops of the mine. The sampling technique followed the guideline provided by Xuyao et al. (2011) [47], and Zhang et al. (2016). These guidelines included the samples' weight and the codes for collecting and transferring the samples to the laboratory by avoiding the possibility of peroxidation [48].

As for the R70 test, the coal samples with a particle size of <212 µm were sieved to satisfy the experimental procedure. The R70 test requires 150 g (±0.01 g) of crushed coal samples. These samples were packed into the coal reaction vessel. In order to minimize the effect of oxidation on fresh surfaces, the test specimens were grinded right before each run.

These samples were prepared from different Iranian coalfields, such as the Tabas Parvadeh coal mines, the Eastern Alborz coal mines, the Central Alborz coal mines, and the Kerman coal mines. Figure 4 shows the location of these case studies. The basic descriptive statistics of the maceral content for this study are given in Table 1-3.

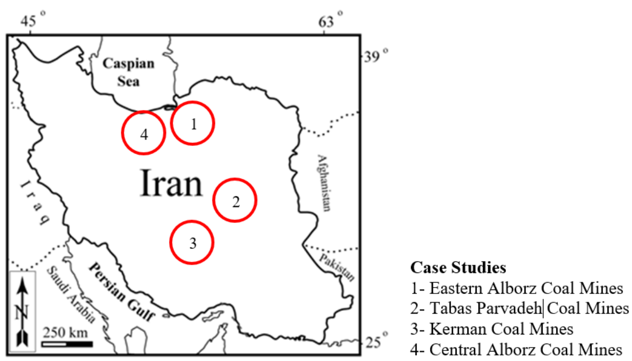

Fig. 4. Location of Case studies.

Table 1. Basic descriptive statistics of the Vitrinite contents for this study.

Statistic	Value	Percentile	Value
Sample Size	51	Min	24.06
Range	60.29	5%	30.796
Mean	59.614	10%	33.738
Variance	327.49	25% (Q1)	45.9
Std. Deviation	18.097	50% (Median)	58.77
Coef. of Variation	0.30357	75% (Q3)	78.97
Std. Error	2.534	90%	81.708
Skewness	-0.09786	95%	82.852
Excess Kurtosis	-1.4005	Max	84.35

Table 2. Basic descriptive statistics of the Liptinite contents for this study.

Statistic	Value	Percentile	Value
Sample Size	51	Min	0
Range	16.91	5%	0.672
Mean	7.1575	10%	1.508
Variance	25.955	25% (Q1)	2.5
Std. Deviation	5.0946	50% (Median)	5.8
Coef. of Variation	0.71179	75% (Q3)	10.86
Std. Error	0.71339	90%	15.102
Skewness	0.4743	95%	15.698
Excess Kurtosis	-1.1337	Max	16.91

Table 3. Basic descriptive statistics of the Inertinite contents for this study.

Statistic	Value	Percentile	Value
Sample Size	51	Min	0
Range	75.94	5%	0
Mean	31.867	10%	0.272
Variance	523.45	25% (Q1)	7.12
Std. Deviation	22.879	50% (Median)	38.75
Coef. of Variation	0.71794	75% (Q3)	48.8
Std. Error	3.2037	90%	63.262
Skewness	-0.0768	95%	67.638
Excess Kurtosis	-1.3141	Max	75.94

3.2. Experimental apparatus and method

3.2.1. R70 method

The R70 method can be summarized as follows. The grinded coal sample (particle size <212 µm) was dried in an adiabatic oven at a temperature of 105-110 ºC for 16 hours under an inert gas flow (nitrogen). Afterward, the coal sample was cooled down to 40 ºC in the same environment and was then stored in an oxygen-rich airflow at the same temperature (40 ºC). The coal oxidation initiated in the latter environment in which the self-heating processes were observed. The average rate of coal heating from 40 to 70 ºC is considered to be the index R70 (ºC/h). The higher the value of the index, the more prone the coal sample is to spontaneous combustion. This method is the most efficient technique regarding the characterization of coal tendency to oxidation and self-heating [49, 50].

3.2.2. Testing system

Figure 5 shows a simplified diagram of the set utilized for the oxidation of coal. Figure 6 shows a schematic sample container (Bomb). The instrument was built in the Shahrood University of Technology in Iran (Faculty of Mining, Petroleum & Geophysics Engineering), and consists of the following parts (see Figure 7 for details):

- Temperature-programmed adiabatic oven, which is used to control the temperature of the coal sample, whose temperature ranges from room temperature up to 400°C, with a precision of 1°C.
- Electric heater
- Fan
- Sample container (Bomb), which is made of pure aluminum and is connected, in order, to an inlet for air supply path, thermocouple for temperature measurement, and an air outlet path (Figure 6).
- 15-meter-long gas preheating copper tube
- Two thermocouples: thermocouple #1 is used to monitor the oven temperature, and thermocouple #2 is used to measure the coal sample temperature)
- JUMO Dicon touch (Control panel), which is consist of:
    - Data logger to record the temperature changes in the coal sample versus time
    - Microcontroller: the programmed adiabatic oven is set to increase the temperature using a microcontroller).
- Computer
- 50kg-heavy O2 gas cylinder as the air supply system.
- 50kg-heavy N2 gas cylinder for drying the coal samples in 105-110 ºC for 16 hours.
- Pressure reduction valve
- Flowmeter

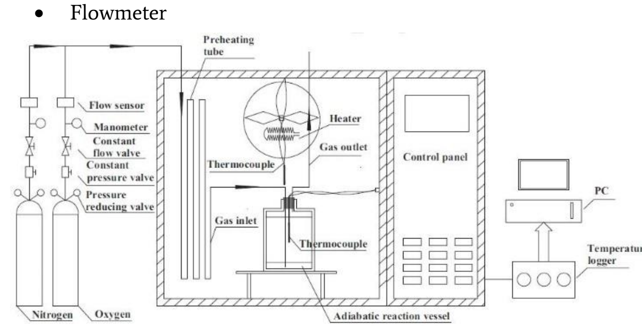

Fig. 5. A schematic diagram of the apparatus applied for the oxidation test system.

4. Modeling and discussion

4.1. Polynomial regression (Single variable)

This section provides a polynomial regression model to show the tendency of spontaneous coal combustion, based on regression analyses.

After conducting the experimental tests, a set of data, including 51 experimental tests, was collected. Additionally, as mentioned earlier, the maceral content was considered as the input data and the R70 test as the output of the SCC tendency model.

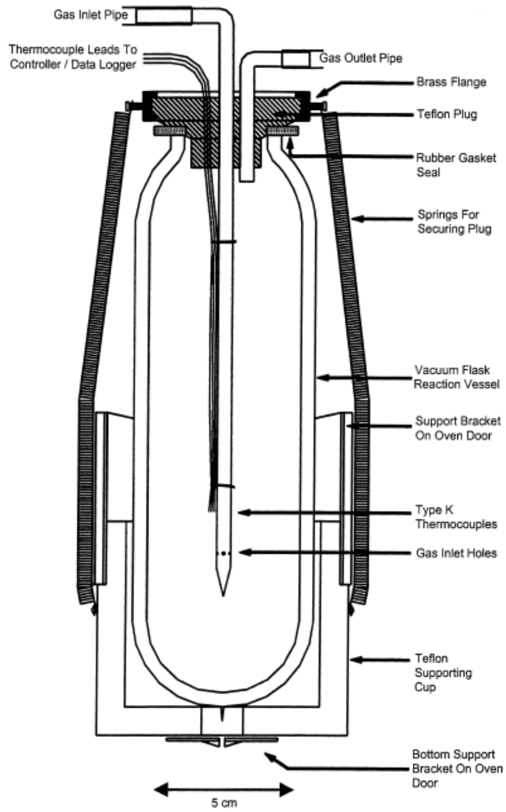

Fig. 6. Sample container (Bomb).

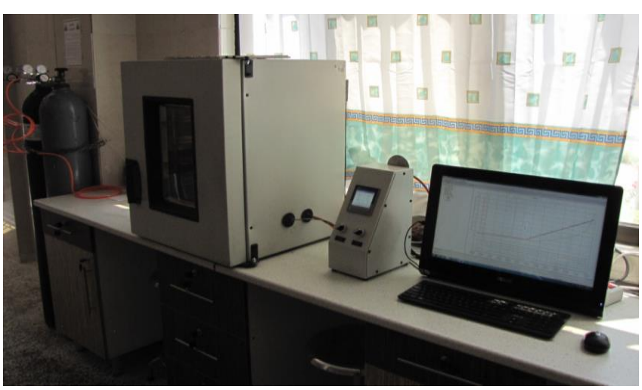

Fig. 7. The testing system.

The results of the maceral content are in agreement with the results of the R70 test, as shown in Figures 8-10. The regression analyses are given in Equations (6) to (8).

$$R70 = 0.0002\ V^3 - 0.0255\ V^2 + 1.4073\ V - 21.273 \qquad R^2 = 0.5356 \qquad (6)$$

$$R70 = 0.007\ L^3 - 0.1738\ L^2 + 1.4263\ L + 1.2153 \qquad R^2 = 0.6603 \qquad (7)$$

$$R70 = -0.0001\ I^3 + 0.0104\ I^2 - 0.3505\ I + 7.8553 \qquad R^2 = 0.7332 \qquad (8)$$

Where, V, L, and I are the Vitrinite, Liptinite, and Inertinite contents, respectively.

Using Equations (6) to (8), the simple relationships between the R70 and the independent variables are important and have a high coefficient of determination (R^2) (see Figures 8, 9, and 10).

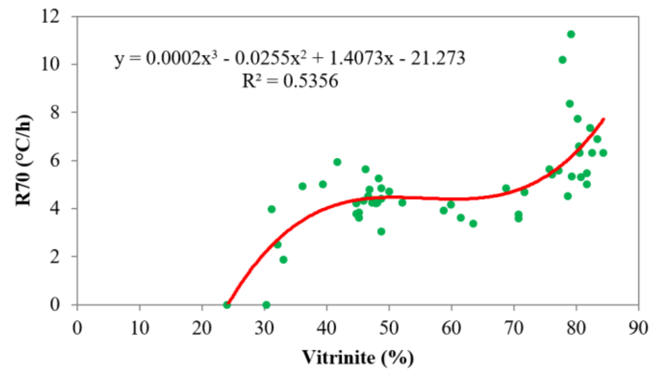

Fig. 8. Integration results of the Vitrinite content comply with the R70 test.

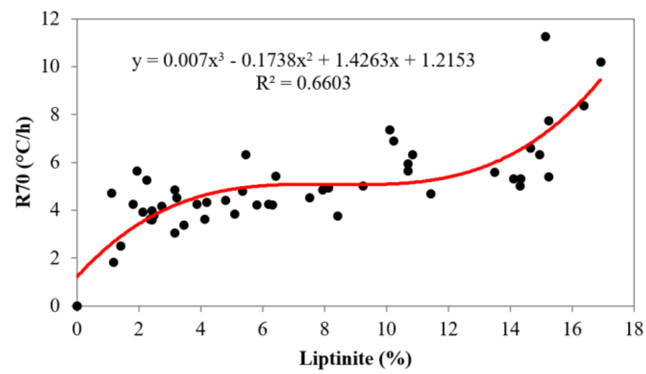

Fig. 9. Integration results of the Liptinite content comply with the R70 test.

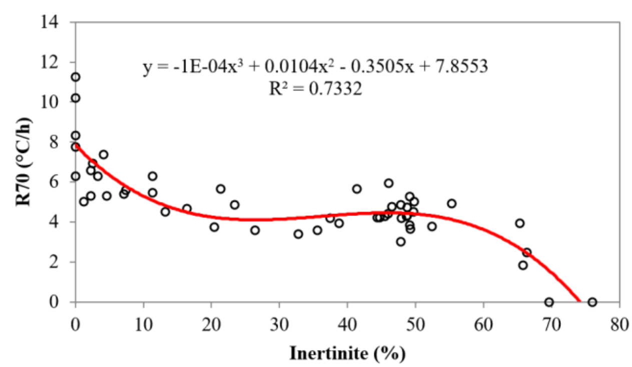

Fig. 10. Integration results of the Inertinite content comply with the R70 test.

4.2. Effects of interactive data on response surface modeling

In order to provide a better understanding of the effect of the maceral content on the tendency spontaneous coal combustion, the 3D surface plots were studied. Besides, the corresponding contour plots represent the x-y plane of the response surface to provide a determination of the effective parameters.

The effects of Vitrinite and Liptinite on R70 at 3D, and residual and its contour plots are represented in Figure 11. As shown in Figure 11, as the Vitrinite and Liptinite increased, the R70 values and the tendency of spontaneous coal combustion increased, and vice versa.

The interaction between Vitrinite and Inertinite on R70, as well as the residual and its contour plots, can be seen in Figure 12. Figure 12 shows that by increasing Vitrinite and decreasing Inertinite, the R70 values increase, and the tendency of spontaneous coal combustion enhances, and vice versa.

The 3D responses of R70, and residual and its contour plots corresponding to Liptinite and Inertinite are given in Figure 13. Figure 13 shows that by increasing Liptinite and decreasing Inertinite, the R70 values are increasing, and the tendency of spontaneous coal combustion is enhanced, and vice versa.

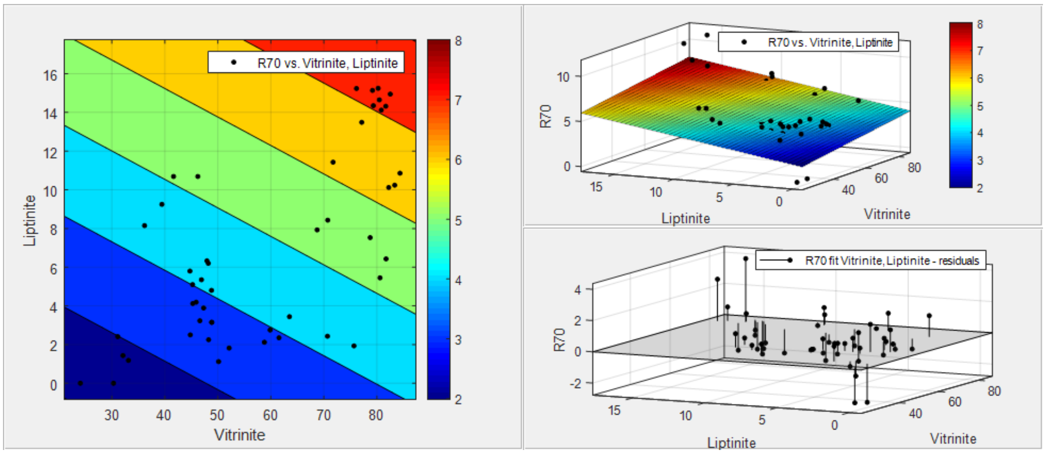

Fig. 11. Contour, surface, and residual plots of R70 versus Vitrinite and Liptinite.

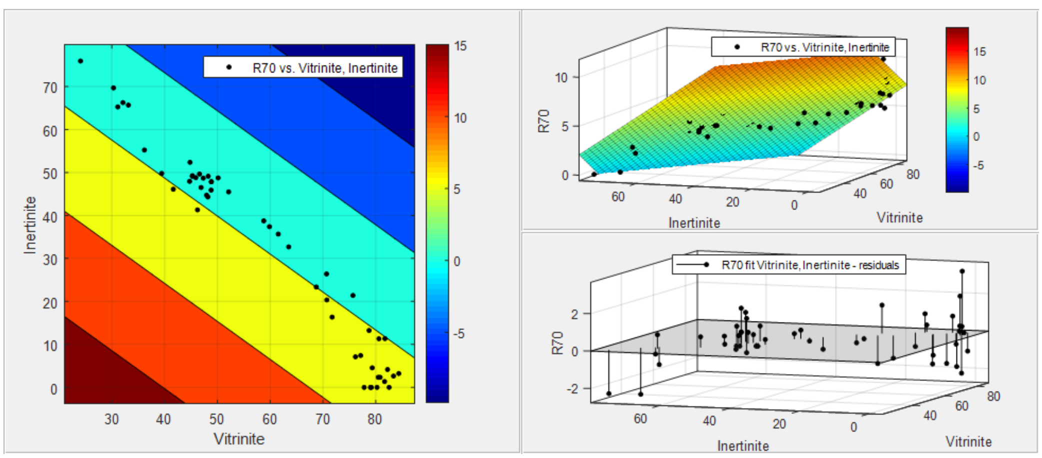

Fig. 12. Contour, surface, and residual plots of R70 versus Vitrinite and Inertinite.

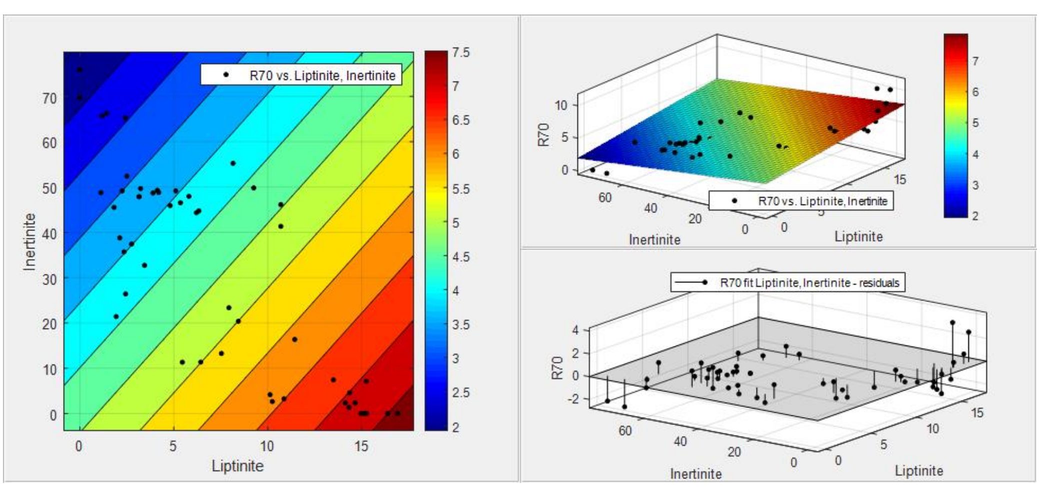

Fig. 13. Contour, surface, and residual plots of R70 versus Liptinite and Inertinite.

5. Conclusions

Spontaneous coal combustion (SCC) is one of the important events in coalfields, which results in the instability of sustainable development. This phenomenon is affected by the intrinsic characteristics of coal. The maceral content in coal has a vital role in this phenomenon. However, there are disagreements among researchers as to the impact of this key parameter on this occurrence.

A comprehensive study in a literature review revealed that the maceral content is a very important factor affecting the occurrence of SCC. Therefore, a comprehensive study was conducted to provide a concluding remark on the effect of this parameter on the tendency of spontaneous coal combustion. In this study, 51 coal samples with different maceral contents were collected from several Iran underground coalfields, and the R70 test was carried out on each coal sample. A multi-variable linear prediction model consisting of three parameters was proposed to evaluate the tendency of spontaneous coal combustion.

The results showed that the maceral content could affect the tendency of spontaneous coal combustion. By increasing Vitrinite and Liptinite contents and decreasing Inertinite, the R70 values increased and the tendency of spontaneous combustion of coal enhanced, and vice versa.

The results showed that there was a definite need to consider the influence of maceral content on spontaneous coal combustion because the maceral content has a complex behavior that should be carefully investigated.

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