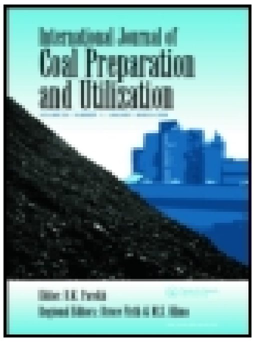

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Evaluation effect of macerals petrographic and pyrite contents on spontaneous coal combustion in Tabas Parvadeh and Eastern Alborz coal mines in Iran

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ABSTRACT
Among the fossil fuels, coal is the most widely used one all over the world for different purposes and is a stable source of energy. Coal mining has serious hazards such as spontaneous combustion. Many factors can influence the tendency for occurrence of this phenomenon in coal mines and coal storages, one of which is the petrographic characteristics (maceral and pyrite contents). The petrographic characteristics are directly affected by the growth of coalification and origin of coal forming. In this research work, firstly, the maceral and pyrite contents in coal samples were identified in two various mines in Iran (Tabas Parvadeh and Eastern Alborz coal mines). Then the spontaneous coal combustion propensity of each sample was measured using the Crossing Point Temperature (CPT) test method. The aim for carrying out this research work was to measure the effect of the maceral and pyrite contents on the grade of spontaneous coal combustion propensity. It was found that the pyrite contents and the vitrinite, liptinite, and inertinite mean levels in the Tabas Parvadeh and Eastern Alborz coal mines were (2.08%, 53.30%, 9.91%, 34.71%) and (1.14%, 53.06%, 6.39%, 39.41%), respectively. The mean CPT value was 148.43°C in the Tabas Parvadeh coal mines and 175.86°C in the Eastern Alborz coal mines. By examining the results of the experiments and comparing the CPT values, it was found that with an increase in the liptinite and pyrite contents and a decrease in the inertinite content, the Tabas Parvadeh coal mines tend to have more tendency for spontaneous coal combustion in comparison with the Eastern Alborz coal mines.

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KEYWORDS
Petrographic characteristics; spontaneous coal combustion; tabas parvadeh coal mines; eastern alborz coal mines; crossing point temperature

Introduction

Fossil fuels consist of nearly 90% of the proved reserves of the global energy. Today, coal is the major component and economical fossil fuel including nearly 90% of the fossil fuel energy around the world (Lang and Fu-Bao 2010; Thakur 2016; Thakur, Schatzel, and Aminian 2014), and is a vital wealth for a foreseeable future (Taheri et al. 2017).

In coal mines, especially the underground ones, various problems such as roof unstableness, water incursion, coal dust, subsidence, outburst, and spontaneous combustion have appeared, hindering an efficient and safe underground mining operation.

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The coarse working status and the perilous environment are the most important elements that affect a coal mining system. Ergo, it is mandatory to precisely analyze the risks involved and to find the plan to estimate, stop, and control those (Ghanbari et al. 2018; Saffari et al. 2013, 2017; Vaziri, Khademi Hamidi, and Sayadi 2018) such as spontaneous coal combustion.

Oxidation of coal is the main heat cause for the spontaneous combustion phenomenon (Wang, Dlugogorski, and Kennedy 2003; Yuan and Smith 2009; Zhou et al. 2017). This process is a principal safety point in coal mining and coal storages (Beamish, Barakat, and George 2001; Beamish and Blazak 2005; Carras and Young 1994; Singh et al. 2007; Song and Kuenzer 2014; Xue et al. 2010). This phenomenon has a menace to beneficial energy resources (Song and Kuenzer 2014), which is an environmental, economic, and social threat in all stages of coal mining (Beamish 2005; Dijk et al. 2011; Finkelman 2004). Thus, this occurrence is an urgent problem that should be solved (Lang and Fu-Bao 2010). Thus, a better conception about the propensity of this event could greatly help a coal mining process.

Coal is regarded as a two-elemental system consisting of organic substantial (maceral) and inorganic substantial (mineral) (Dalla Torre, Mählmann, and Ernst 1997; Gürdal 2008; Liu et al. 2005; Rice 1993; Robeck and Huo 2016; Vassileva and Vassilev 2006).

Maceral is the main material that can be used to consider the nature of coal (type and grade) and find out its capabilities for various uses (Dai et al. 2010). It is a non-crystalline organic material, and its composition may vary widely. In terms of macerals, coals are arranged as vitrinite, liptinite, and inertinite. The properties of coal rely on the type and level of maceral present in coal (Cui and Bustin 2006). Pyrite is one of the three forms of sulfur that exist in coal. Pyrites show catalytic reaction as their oxidized products speed-up the value of oxidation of the organic composition that exists in coal. Pyrite oxidation starts for the creation of ferric ions, which catalyzes the reaction. Thus, the level of tendency for spontaneous coal combustion depends on various factors, one of which is the maceral petrographic and pyrite contents.

Spontaneous combustion occurs without an external heat source, which is a result of a number of miscellaneous exothermic reactions (Beamish and Blazak 2005; Beamish and Hamilton 2005); the intrinsic coal characteristics such as the petrographic characteristics have been systematically specified to have a big influence on this phenomenon (Saffari et al. 2013, 2017).

This process turn the inside heat index of the coal, causing an increase in the temperature and leading to an open fire and burning of the coal (Akgün and Arisoy 1994; Beamish and Arisoy 2008; Carras and Young 1994; Nugroho, McIntosh, and Gibbs 2000; Ren, Edwards, and Clarke 1999; Smith and Glasser 2005; Wang, Dlugogorski, and Kennedy 2003).

The maceral petrographic and pyrite contents of coal are important parameters involved in the tendency for spontaneous coal combustion. Previous surveys on the effect of macerals on the tendency for spontaneous coal combustion have achieved recognition of macerals to classify coal types. In previous research works, there is a little study where the effect of macerals on the tendency for spontaneous coal combustion has been examined. Nevertheless, the effect of macerals on the level of spontaneous coal combustion has not been tested, which is of great significance for coal miners. Hence, in this research work, the effect of the maceral and pyrite contents on spontaneous coal

Table 1. The age and rank of coal samples.

Case Studies	Age of the coals	Coal rank
Tabas Parvadeh coal mine	Mesozoic (Jurassic)	Bituminous
Eastern Alborz coal mine	Mesozoic (Jurassic)	Bituminous

combustion was carried out. For this purpose, the coal samples were obtained from the Tabas Parvadeh and Eastern Alborz coal mines in Iran.

As mentioned by Thomas and Thomas (2002), the coal deposits of Iran are Mesozoic (Jurassic) in age, with some Lignites of Paleogene-Neogene age. The Jurassic coals are bituminous with high ash and sulfur contents, and have coking properties. All are strongly tectonized with seam thicknesses ranging from 1 m to 4 m. The coal supplies local needs and the metallurgical industry. Principal coalfields are located at Alborz in the North and at Tabas in the central Iran. So, the age and rank of coal samples are mentioned in Table 1.

The samples were tested for tendency for spontaneous coal combustion using the Crossing Point Temperature (CPT) test method, described in Section 3.

Firstly, the collected samples were analyzed to distinguish the types and composition of the maceral and pyrite contents. Then, the same samples were subjected to tendency for spontaneous coal combustion. The results obtained for the effects of regression analysis of maceral types and its composition on spontaneous coal combustion propensity are presented in this research work.

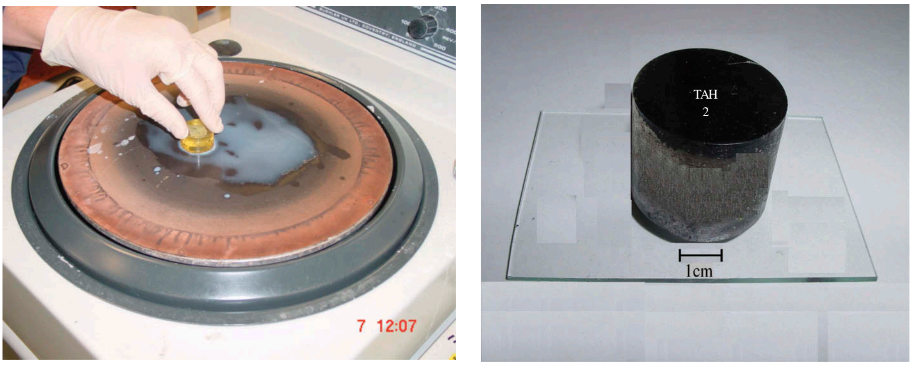
Maceral and Pyrite Content Determinations

Firstly, 14 coal samples were collected from two coalfields in Iran. Seven of these coal samples were collected from Tabas Parvadeh and the rest were gathered from Eastern Alborz. Then these coal samples were sent to the laboratory for the maceral and pyrite composition analysis, as described in Section 2.1 and 2.2. The petrographic examination was done on the sized fraction.

Sample preparation

The coal samples to be used for the petrographic study were made into polished cylinders approximately 20 mm in diameter using plastic moulds. Initially, the coal samples were initially crushed to particle sizes between approximately 100μmand 0.7 mm. A 0.7 mm sieve was used to separate out large fractions. The oversize particles were re-crushed until all the coal particles were <0.7 mm. A riffle was used to aid in the collection of sufficient representative samples to half fill the rubber mould. A cold setting polyester resin, consisting of 98% Astic resin and 2% hardener was mixed and added to fill the mould. The specimen and resin mixtures in the moulds were thoroughly stirred with a paper clip so that all coal grains were in direct contact with resin. The specimens were then placed in a special vacuum chamber at 70 kPa (absolute) for 2 min to remove air bubbles. Once the specimens were mixed and all air bubbles expelled, they were then left to set for 24 h.

The final step in sample preparation was specimen polishing carried out on a rotating lapping machine (Fig. 1). It was important to produce a highly polished face on the



a) A sample polished on a lapping machine **b) Finished sample**

Figure 1. Polished sample on lapping machine. a) A sample polished on a lapping machine. b) Finished sample.

specimens to allow the collection of accurate reflectance data and to aid in maceral identification.

A coarse carborundum paper was initially used to remove sharp edges from one face of the specimen to allow for easy handling. After this, 240 grit carborundum paper was used to polish the opposite face manually. After several strokes, the polished face was rotated 90 degrees and polishing continued.

The specimen face was periodically flushed and cleaned with soap and water, and then alcohol was sprayed on the face and it was dried with an automatic heater. This process was repeated until a smooth and shiny face was created. The aim of this exercise was to successively polish the specimen to greater degrees of smoothness, so as to remove surface scratches and to create a surface of maximum quality free from surface roughness, using 400, 600 and 1200 grit papers successively and pouring "Diamond Polishing Solvent".

Microscopy

A Leitz MPV-2 microscope was used for maceral analysis. Reflected white light and fluorescence mode illumination were used with 32x and 50x oil immersion objectives giving a total magnification of approximately 400x to 500x. Figure 2 shows a general view of the Leitz orthoplan microscope and the Leitz MPV-2 photometer.

Point count analysis utilizes a swift automatic point counter and mechanical stage. The composition of the specimens was examined under oil immersion with the microscope at a total magnification of 320x. A mechanical stage was used to move the specimens under the objective lens in a regular manner.

Point counts were thus carried out with a regular east west (or column) spacing of 0.6 mm and a north south (or row) spacing also of 0.6 mm. These spacing were selected so that the likelihood of counting any one particular grain (maximum size of 0.7 mm) twice was very low. The objective lens had a small cross-hair inscribed in its center. Each time the lens was moved to a new position the coal component under the cross-hair was identified and recorded. Where the movement controller moved the objective lens and

Figure 2. A general view of Orthoplan maceral microscope with MPV-2 photometer.

cross-hair over the specimen resin, the point was ignored and the lens moved to the next point.

A minimum of 500 points were counted for each specimen in this manner. Care was taken to ensure that point readings were taken at the end of the last row (the row in which the 500th point was recorded), so as not to create a composition bias due to grain sedimentation in the resin. Each coal component was classified into one of the following:

- Vitrinite (VIT)
- Inertinite (INT)
- Liptinite (LIP)
- Pyrite (PYR)

The maceral and pyrite compositions were recognized in each sample based on (Nugroho, McIntosh, and Gibbs 1998) (Australian Standard AS 2856.2–1998 2013), as shown in Figs. 3 and 4, using microscopic images, and listed in Tables 2 and 3. Figure 5 shows the mean values for the maceral and pyrite compositions of the samples.

The results obtained show that the vitrinite levels are approximately the same in both case studies. Also, the results show that the liptinite and pyrite contents in the Tabas Parvadeh coal mine samples are greater than those in the Eastern Alborz coal mine samples. On the other hand, the inertinite level was higher in the Eastern Alborz coal mine samples. The outcomes of this research work are in line with the studies carried out by Sereshki, Vaezian, and Saffari (2016) and Taheri et al. (2017).

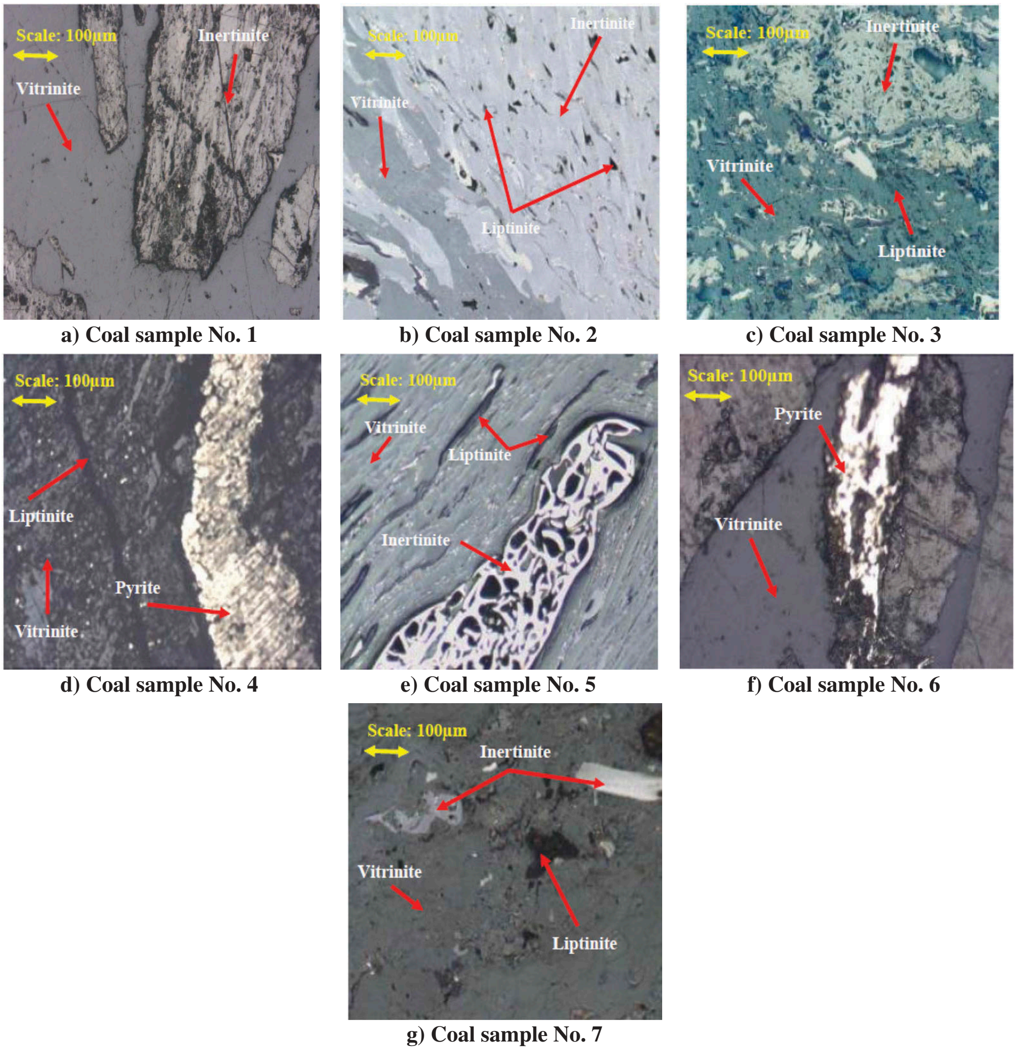


Figure 3. Macerals and minerals found in Tabas Parvadeh coal mine samples. a) Coal sample No. 1. b) Coal sample No. 2. c) Coal sample No. 3. d) Coal sample No. 4. e) Coal sample No. 5. f) Coal sample No. 6. g) Coal sample No. 7.

Tendency for Spontaneous Coal Combustion

Coal Samples and Their Preparation

Fourteen coal samples (from the Tabas Parvadeh and Eastern Alborz coal mines in Iran) were freshly collected directly from the worked face of the mines.

The conditions of the collected samples were studied to avoid the possibility of peroxidation and the amount of sample required for each experiment was determined in accordance with the patterns provided by Xuyao et al. (2011) and Zhang et al. (2016).

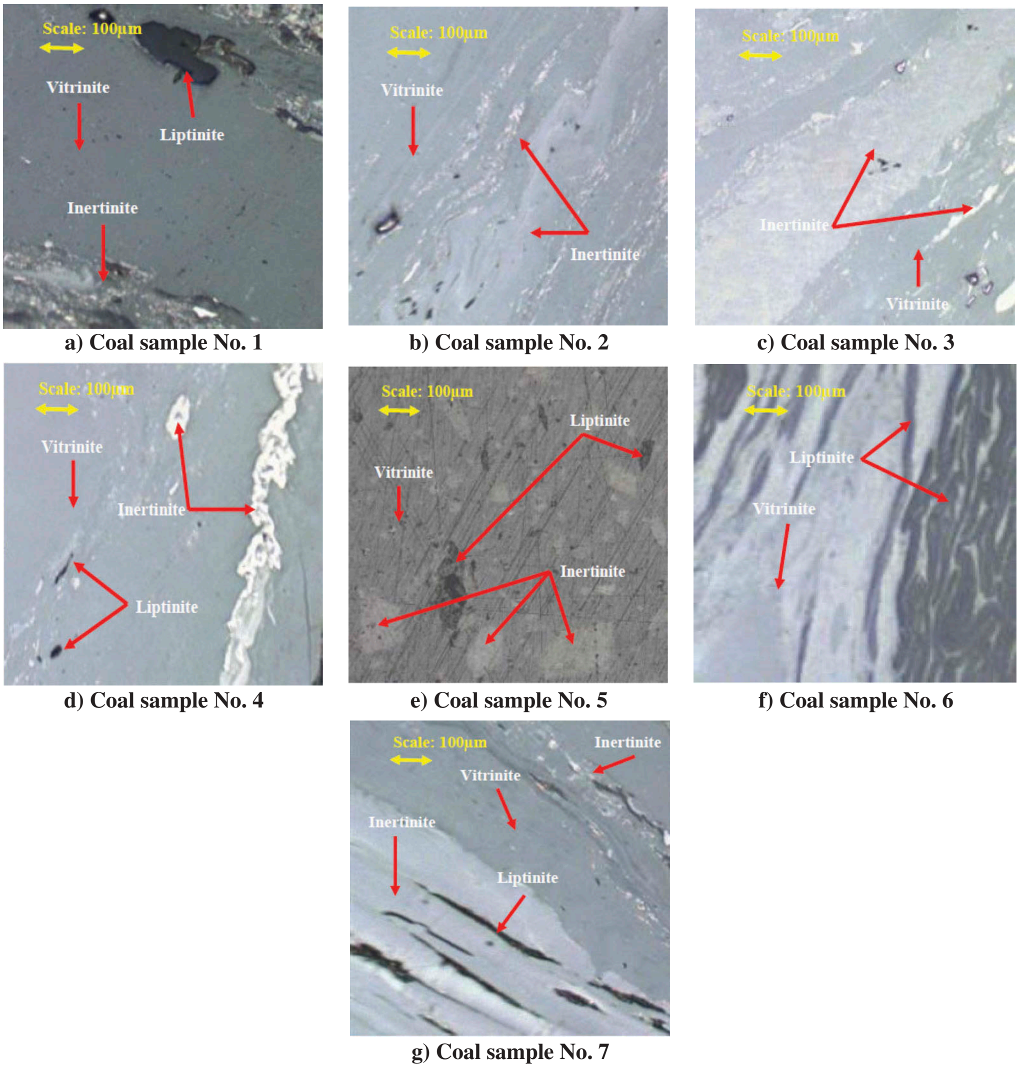


Figure 4. Macerals and minerals found in Eastern Alborz coal mine samples. a) Coal sample No. 1. b) Coal sample No. 2. c) Coal sample No. 3. d) Coal sample No. 4. e) Coal sample No. 5. f) Coal sample No. 6. g) Coal sample No. 7.

Table 2. Maceral and pyrite composition analysis in Tabas Parvadeh coal mine samples.

Sample No.	1	2	3	4	5	6	7	Mean value
Pyrite content (%)	0.11	0.45	1.78	4.52	1.53	4.66	1.51	2.08
Vitrinite (%)	47.31	36.17	46.22	80.23	44.73	78.97	39.44	53.30
Liptinite (%)	3.89	8.15	10.69	15.25	5.8	16.37	9.25	9.91
Inertinite (%)	48.69	55.23	41.31	0	47.94	0	49.8	34.71
CPT (°C)	190	180	146	105	165	100	153	148.43

Table 3. Maceral and pyrite composition analysis in Eastern Alborz coal mine samples.

Sample No.	1	2	3	4	5	6	7	Mean value
Pyrite content (%)	1.11	1.36	0.52	1.96	1.23	1.78	0	1.14
Vitrinite (%)	45.9	45.26	46.6	77.13	46.9	79.32	30.31	53.06
Liptinite (%)	4.19	4.12	3.23	13.49	5.35	14.35	0	6.39
Inertinite (%)	48.8	49.26	49.65	7.42	46.52	4.55	69.69	39.41
CPT (°C)	174	160	186	135	168	138	270	175.86

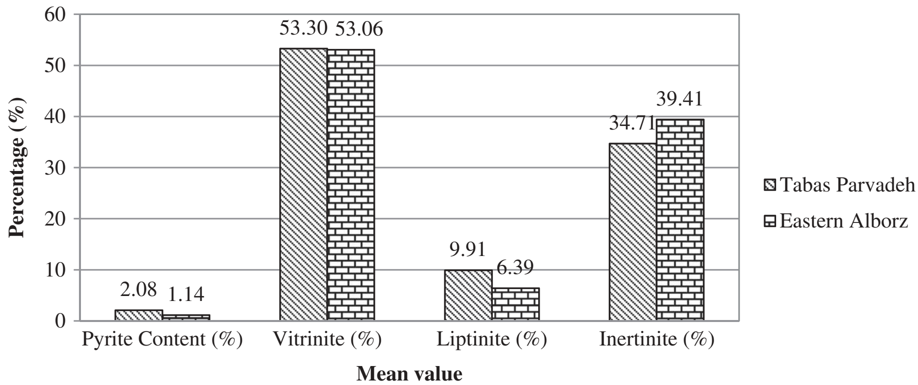


Figure 5. Comparison between maceral and pyrite compositions of Tabas Parvadeh and Eastern Alborz coal mine samples.

Experimental Apparatus and Method

CPT Method

The essence of this method is as what follows. The coal sample was fixed in a reactor available in an oven, and was heated with a constant rate. The programmed adiabatic oven was set to run at a fixed temperature of 50°C, while dry air with oxygen was passed to flow through the coal reaction vessel at a rate of 50 mL/min. The temperature logger was used to log the coal and the surrounding temperature. When the coal temperature reached 50°C, the programmed adiabatic oven was set to rise the temperature at a programmed rate of 1°C/min, while the flow rate of dry air was maintained at 50 mL/min. The experiment was ended when the coal temperature was higher than the surrounding programmed adiabatic oven, and when the coal sample temperature equaled the linearly ramped oven temperature, it was called CPT (Chen 1991; Mohalik, Lester, and Lowndes 2016; Nugroho, McIntosh, and Gibbs 1998; Wang et al. 2009; Xuyao et al. 2011), as shown in Fig. 6.

Testing System

Figure 7 shows a simplified diagram for the set utilized for the oxidation of coal. Figure 8 shows a schematic sample container (bomb) of the apparatus. The instrument is shown in Fig. 9; it was made in Shahrood University of Technology in Iran (Faculty of Mining, Petroleum and Geophysics Engineering).

- Temperature-programmed adiabatic oven (used to control the temperature of the coal sample, whose temperature ranged from the room temperature to 400°C with a precision of 1°C in the control)

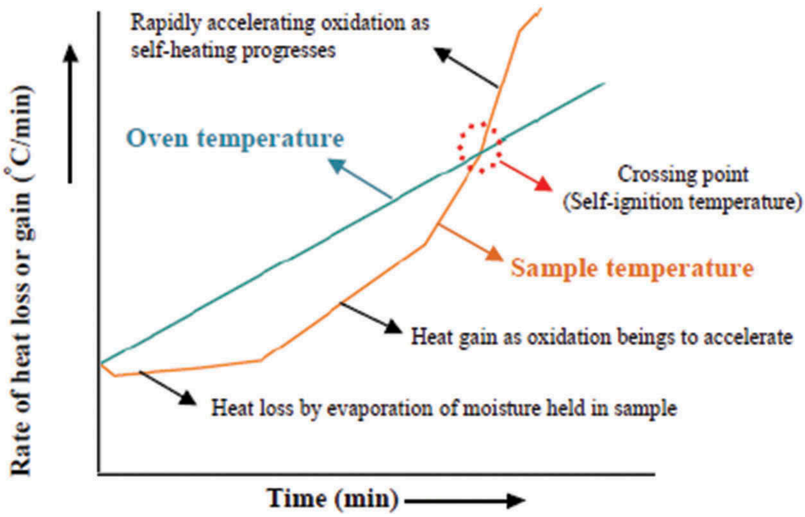


Figure 6. Schematic diagram of CPT (Kim 1995).

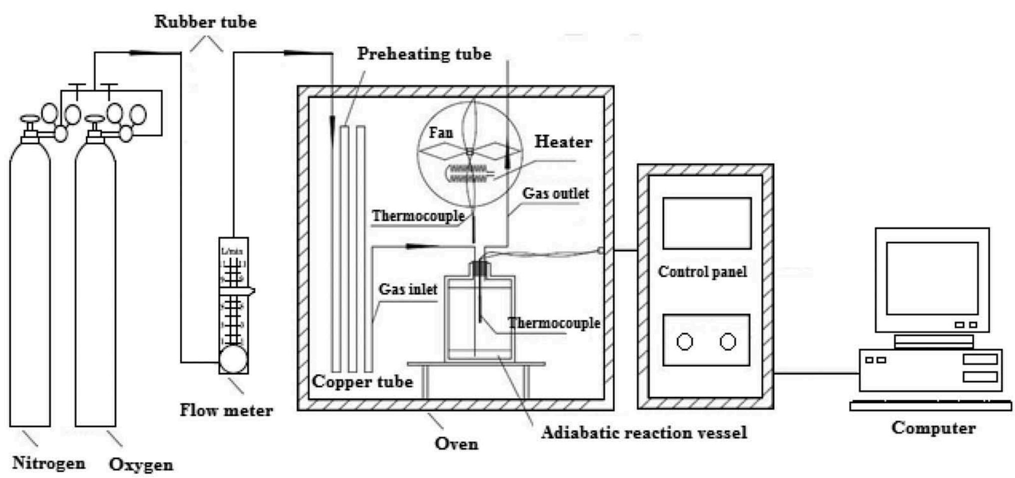


Figure 7. A graphic diagram for the set applied for the coal oxidation.

- Electric heater
- Fan
- Sample container (bomb) (made of pure aluminum and is, respectively, connected to an inlet for air supply path, a thermocouple used for temperature measurement, and an outlet for the air outlet path) (Fig. 8)
- 15 m gas pre-heating copper tube
- Two thermocouples (thermocouple #1, used to monitor the oven temperature, and thermocouple #2, used to measure the coal sample temperature)
- JUMO Dicon touch (Control panel), consisting of:
- Data logger (to records the temperature changes in the coal sample with time)

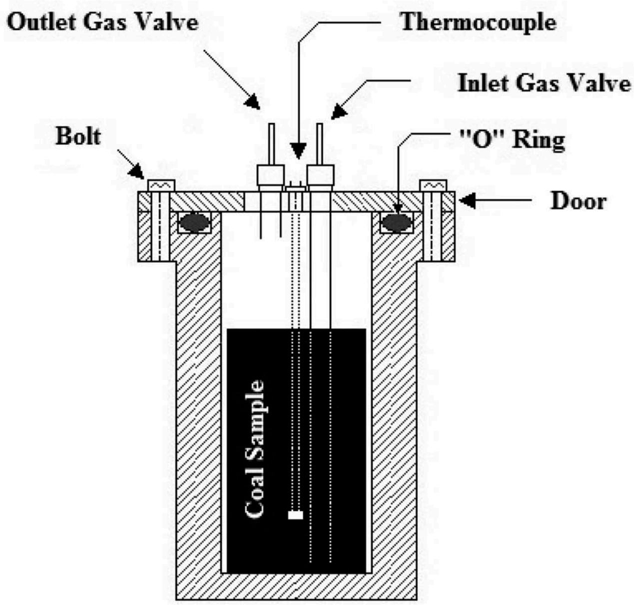


Figure 8. Sample container (bomb).

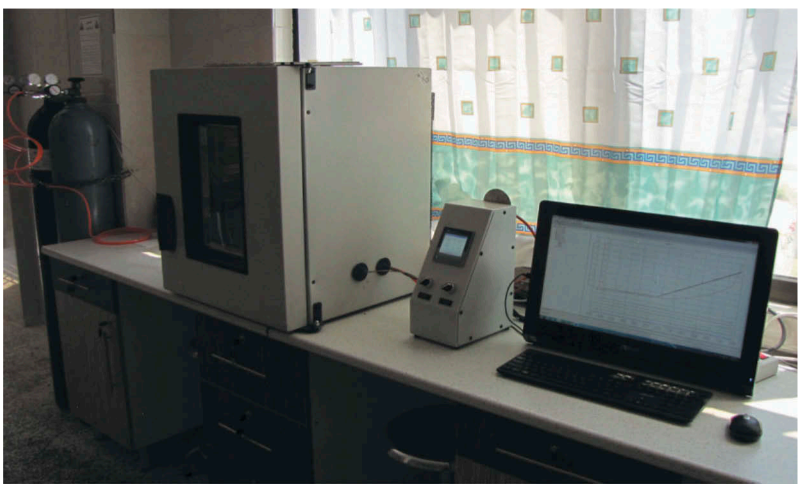


Figure 9. The testing system.

- Micro-controller (the programmed adiabatic oven was set to increase the temperature with a micro-controller)
- Computer
- 50 kg O_2 gas cylinder (air supply system)
- 50 kg N_2 gas cylinder (for pre-heating coal sample to 50°C to start the test)
- Pressure reducing valve
- Flow-meter

CPT Test Method for Coal Samples

The coal samples were tested with the CPT experimental set-up. Figures 10 and 11 show the curves for reactivities of the coal samples in the Tabas Parvadeh and Eastern Alborz coal mines using the CPT test method, respectively. Also, the CPT test results for the coal samples are given in Tables 2 and 3 (end rows). Figure 12 shows the results of a comparison between the mean CPT values for the samples collected from the two understudied coalfields.

Figures 10–12 show that the Tabas Parvadeh coal mine samples have a higher potential for spontaneous combustion in comparison with the Eastern Alborz coal mine samples. These values and ratings are generally consistent with the coal petrographic differences between the samples (described in Section 4).

According to the analysis results, the Tabas Parvadeh coal mines have higher levels of liptinite and pyrite contents and lower levels of inertinite than the coal samples in Eastern Alborz.

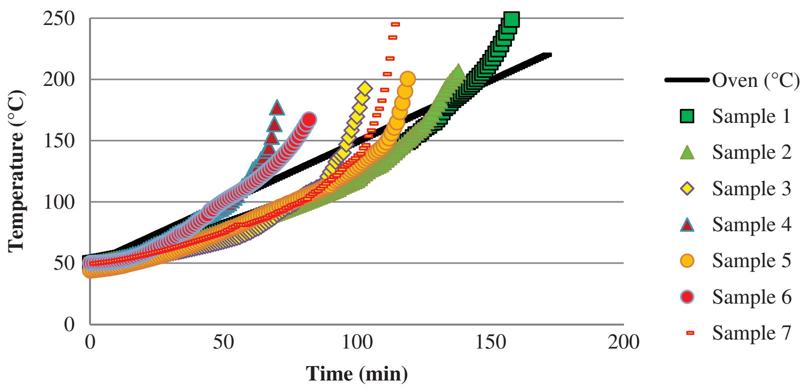


Figure 10. CPT test results for Tabas Parvadeh coal mine samples.

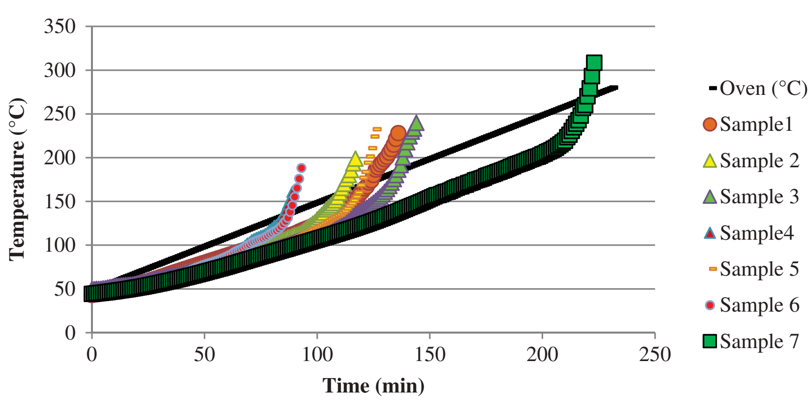


Figure 11. CPT test results for Eastern Alborz coal mine samples.

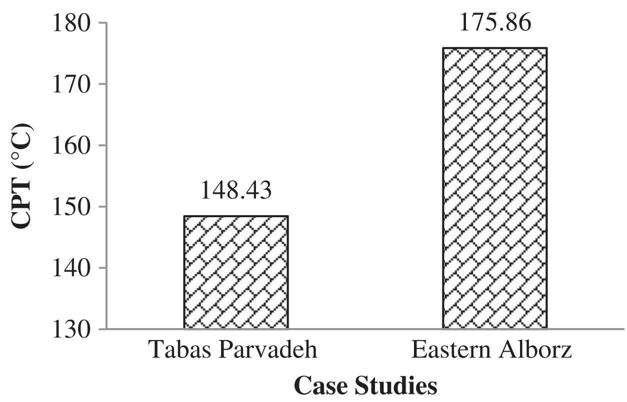


Figure 12. Comparison between CPT test results for Tabas Parvadeh and Eastern Alborz coal mine samples.

Effect of Coal Petrography on Spontaneous Coal Combustion in Case Studies

It is meaningful to have a full comprehension of the relationship between coal petrography and spontaneous coal combustion propensity for detecting this phenomenon. Coal petrography can affect this process; some of the characteristics accelerate this process and the others reduce it. Therefore, in this section, based on the relationship regression between the characteristics and the CPT method, these relationships were described and their roles were determined. Figure 13–20 show the relations between the CPT values and the coal petrography using the regression analysis.

The results obtained show that the percentage pyrite content (Figs. 13 and 14), vitrinite (Figs. 15 and 16), and liptinite (Figs. 17 and 18) has an influence on the CPT values, and spontaneous coal combustion is accelerated with an increase in these characteristics. In addition, inertinite (Figs. 19 and 20) has an inverse relationship with the spontaneous coal combustion tendency, and coal spontaneous combustion decreases with an increase in this parameter.

In Fig. 15–20, the results of the vitrinite, liptinite, and inertinite fit CPT test method are given, respectively. These figures show that the spontaneous samples have higher

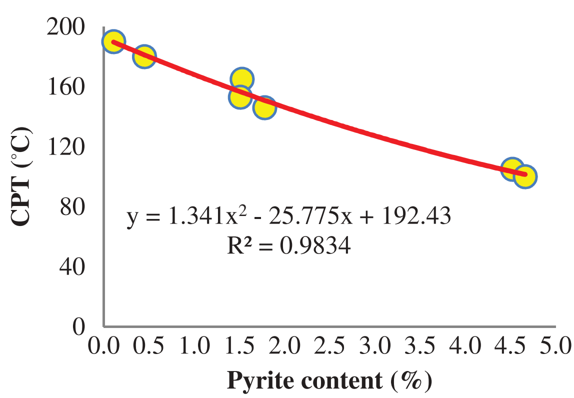


Figure 13. Results of pyrite content fit CPT test method for Tabas Parvadeh coal mine samples.

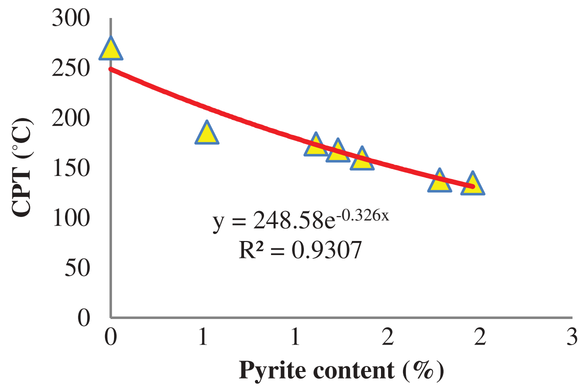


Figure 14. Results of pyrite content fit CPT test method for Eastern Alborz coal mine samples.

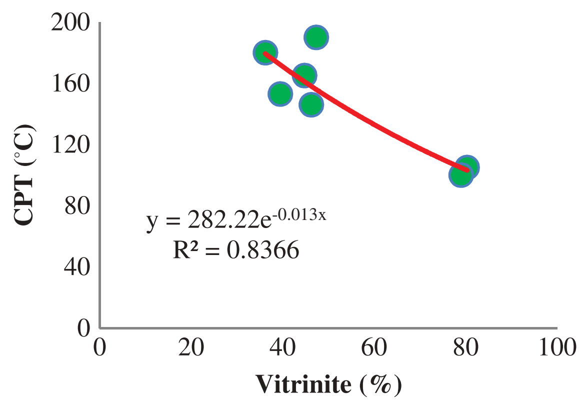


Figure 15. Results of vitrinite fit CPT test method for Tabas Parvadeh coal mine samples.

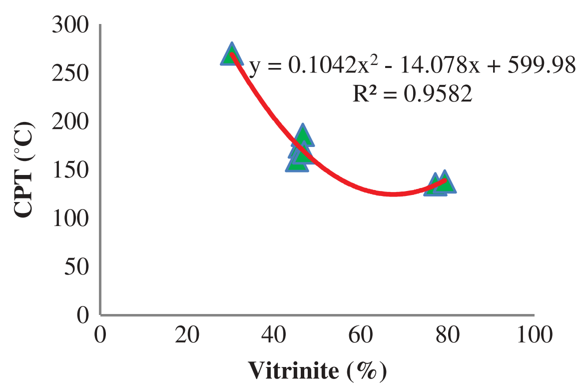


Figure 16. Results of vitrinite fit CPT test method for Eastern Alborz coal mine samples.

percentages of vitrinite and liptinite than the non-spontaneous sample. The non-spontaneous coal samples have a higher percentage of inertinite than the spontaneous one.

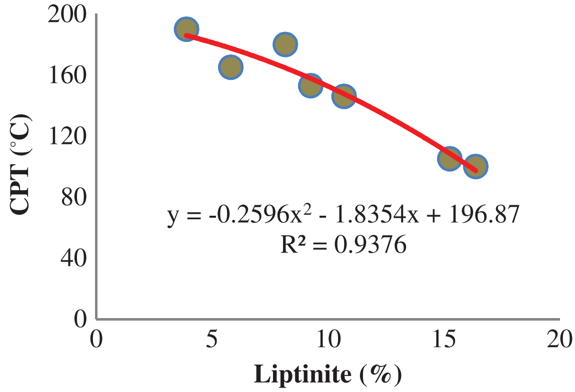


Figure 17. Results of liptinite fit CPT test method for Tabas Parvadeh coal mine samples.

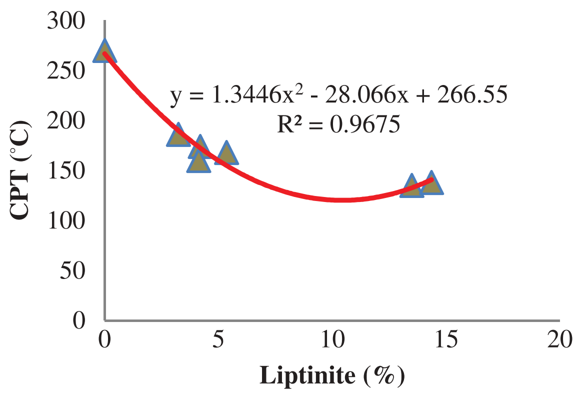


Figure 18. Results of liptinite fit CPT test method for Eastern Alborz coal mine samples.

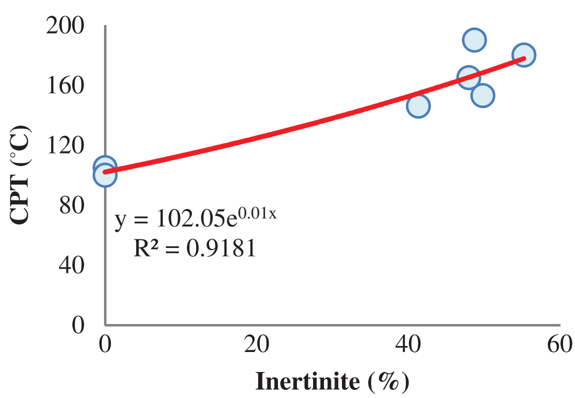


Figure 19. Results of inertinite fit CPT test method for Tabas Parvadeh coal mine samples.

Oxidation incurs the non-aromatic sections of the coal molecules. These forms are common in lipnite and vitrinite, especially in the low ranks (Falcon 1986). The highly reactive parts in coals, especially the lower ranks, are identified by high proportions of

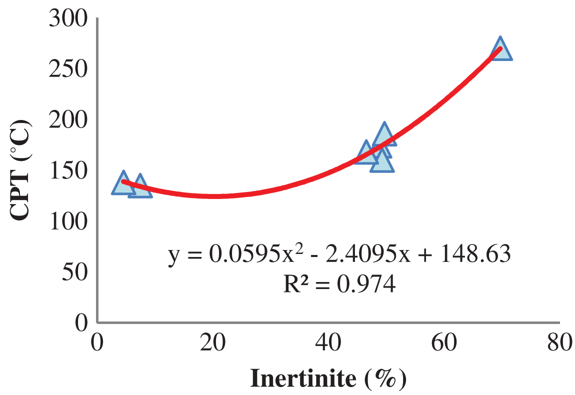


Figure 20. Results of inertinite fit CPT test method for Eastern Alborz coal mine samples.

vitrinite and liptinite. Vitrinite maintains more non-aromatic (and therefore, potentially reactive) parts in its molecular structure than inertinite. Oxygen is, therefore, adsorbed onto the molecular structure more abundantly per unit area and with lower activation energies, and thus leading to easier, and possibly more rapid, forms of oxidation at lower temperatures in earlier stages than inertinite of the same rank (Falcon and Snyman 1986).

As shown in Fig. 13–20, the range of CPT values in the Tabas Parvadeh coal mine samples are lower than those in the Eastern Alborz coal mine samples, and have a sharper slope of variation. Thus, the Tabas Parvadeh coal mine samples have a higher potential of spontaneous combustion in comparison with the Eastern Alborz coal mine samples. These values and ratings are generally consistent with the coal petrographic differences between the samples, mentioned above.

Conclusions

An organized experimental study was carried out for finding the spontaneous coal combustion tendency by collecting coal samples from two coal fields in Iran. The spontaneous coal combustion tendency was related to the coal petrography, resulting in the following main conclusions:

- The spontaneous coal combustion tendency was influenced by the pyrite, vitrinite, and liptinite contents, and had an influence on the CPT values; spontaneous coal combustion was accelerated with an increase in these characteristics.
- The inertinite content had an inverse relationship with the spontaneous coal combustion tendency, and spontaneous coal combustion decreases with an increase in this characteristic.
- The liptinite and pyrite content levels in the Tabas Parvadeh coal mine samples were greater than those in the Eastern Alborz coal mine samples.
- The inertinite level was higher in the Eastern Alborz coal mine samples.

- The mean CPT value was 148.43°C in the Tabas Parvadeh coal mines and 175.86°C in the Eastern Alborz coal mines. Thus, the Tabas Parvadeh coal mine samples had a higher potential for spontaneous combustion.
- The CPT value trend in the Tabas Parvadeh coal mines had a sharper slope variation.
- The increase in the liptinite and pyrite contents and a decrease in the inertinite content in the Tabas Parvadeh coal mines have contributed to the tendency for spontaneous coal combustion.

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