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The simultaneous effect of moisture and pyrite on coal spontaneous combustion using CPT and R70 test methods

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Abstract

Among fossil fuels, coal is the most widely used all over the world for generating power and electricity and it is a stable source of energy. Despite all these benefits, coal mining has serious hazards, such as coal spontaneous combustion. There are many factors that influence the tendency for coal to spontaneously combust in coal mines. Pyrite can promote the risk of this phenomenon. This promotion is accelerated by the combination of pyrite and moisture content at the same time. This combination is very rarely discussed in literature. So, in this research, the accelerating effect of reactive pyrite and moisture content on coal spontaneous combustion was measured experimentally using crossing point temperature (CPT) and R70 test methods. For this purpose, a new experimental apparatus was assembled and made in Iran. Reaction rate data obtained from the experimental results showed that pyrite has a twofold action. It first catalyzes the oxidation reaction. Then, in a moist environment, pyrite is itself oxidized, which provides a secondary heat source, and so accelerates the process of coal spontaneous combustion. Since the pyrite oxidation reaction consumes moisture, there is a mutual effect of accelerated heating as less heat is used up in moisture evaporation. The results show that pyrite content can linearly accelerate the coal spontaneous combustion process, while moisture content under 20% increases it, and if the moisture exceeds 20%, the rate of this process is reduced. The results of this research are helpful in the assessment and management of coal spontaneous combustion issues in coal mines.

Keywords:

Coal Spontaneous Combustion, Moisture, Pyrite, CPT Method, R70 Method.

1. Introduction

Fossil fuels contain about 90% of the proven reserves of global energy. Nowadays, coal is a major component and it is the most plentiful and economical fossil fuel including nearly 90% of the fossil fuel energy around the world, which meets growing energy requirements in many countries, and thereby, coal makes up a large portion of economic growth (**Saffari et al., 2013; Thakur et al., 2014; Sereshki et al., 2016; Ghanbari et al., 2018; Medunić et al., 2018**). Over the past 250 years, it has played a vital and fundamental role in the development and stability of the world economy.

Regardless of all its benefits, coal mining is a very intricate system and process. The harsh working conditions and the hazardous environment are the most important factors that affect the coal mining process. The hazards of underground mining are critical parameters, which should be considered in the design and planning phase of coal mining. Some significant hazards in underground coal mining can be summarized as subsidence, outburst, and spontaneous combustion. Therefore, it is necessary to accurately identify the risks involved and to find ways to forecast, prevent, and control them (**Saffari et al., 2017**).

Coal is a burning material, which is applicable to a variety of oxidation scenarios with conditions ranging from the atmospheric temperature to the ignition temperature. "Spontaneous combustion," which is also called "coal self-heating,", "coal self-burning" and "coal self-ignition" events are disasters triggered by both natural and human factors and may occur in underground coal beds, coal gangue dumps, coal piles, and abandoned mines (**Yuan and Smith, 2012; Wang and Chen, 2015**).

This phenomenon is one of the most frequent and one of the most serious natural disasters in the world's coal industry (**Li et al., 1998; Hu and Jiang, 2000; Xian et al., 2001; Xu, 2001; Saffari et al., 2017**). It can cause a series of problems, including personal injury, significant environmental contamination, huge economic loss, and temporary or permanent mine closures, (**Xuyao et al., 2011; Deng et al., 2015; Saffari et al., 2019**).

Many researchers have worked on techniques to determine the mechanism of this phenomenon. The results of their work show that coal spontaneous combustion is an oxidation process that happens without an external heat source and is an extremely complex, dynamic, autoaccelerated physicochemical process, through which

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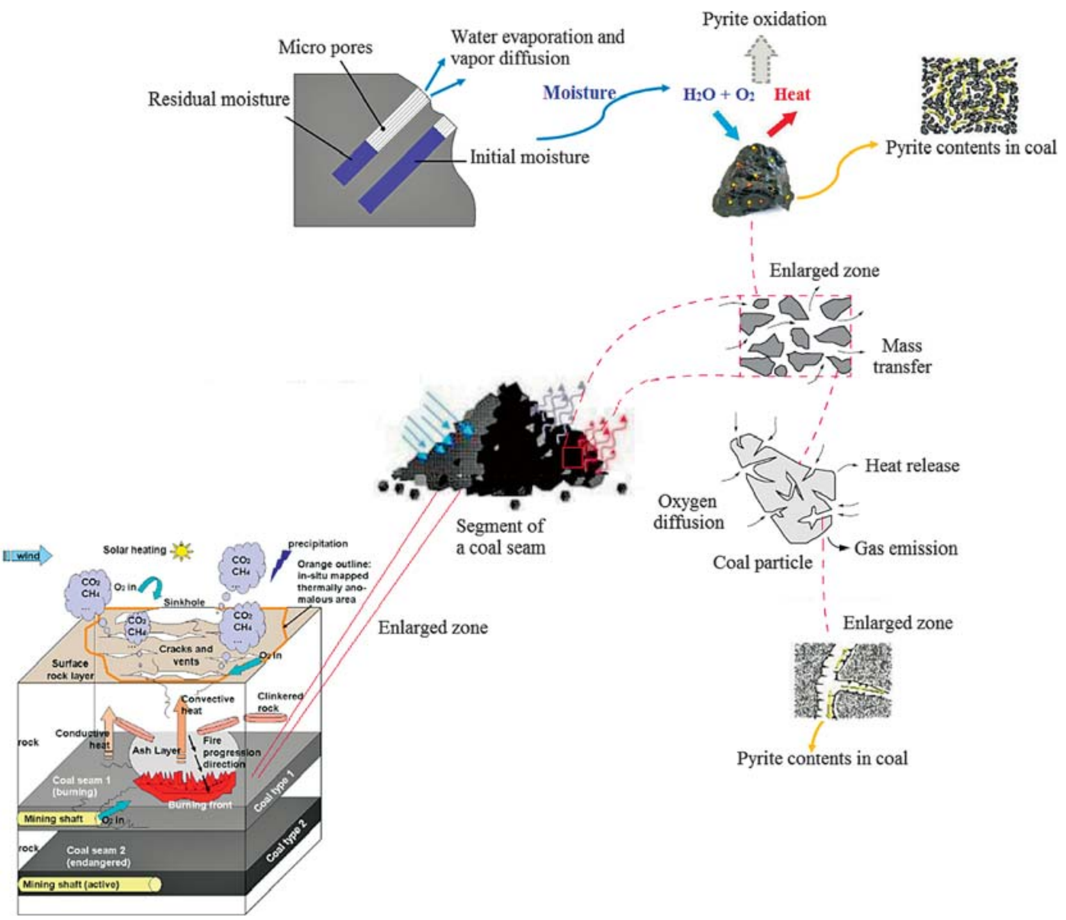

Figure 1: Schematic fundamental phenomena of combination of moisture and pyrite on the coal spontaneous combustion process (modified **after Grewer, 1994; Wang et al., 2003; Dijk et al., 2011**)

corresponding gas products such as O_2, CO, CO_2, and CH_4, are released (**Yuan and Smith, 2013; Deng et al., 2015, 2018**). This reaction changes the internal heat profile of the material, leading to an increase in temperature. This can eventually lead to an open flame and burning of the material (see **Figure 1**) (**Akgun and Arisoy, 1994; Carras and Young, 1994; Ren et al., 1999; Nugroho et al., 2000; Wang et al., 2003; Smith and Glasser, 2005; Beamish and Arisoy, 2008**).

The propensity of coal to spontaneous combustion is an intrinsic property (**Wang et al., 2009**), which is affected by many parameters. This promotion accelerates with the combination of pyrite and moisture at the same time. It acts as an important foundation for coal spontaneous combustion.

The influence of pyrite and moisture on the process of coal spontaneous combustion has been investigated in a number of studies, which are mentioned in **Table 1**, and the percentage of studies done on these parameters is shown in **Figure 2**. Thus, the study of the effects of the pyrite and moisture contents has become increasingly vital in coal spontaneous combustion.

Pyrite is one of the three forms of sulfur, which exist in coal. Pyrites generally show a catalytic effect as their oxidized product accelerates the rate of oxidation of organic compounds present in coal. The pyrite oxidation leads to the formation of ferric ions which catalyzes the reaction, as can be seen in **Equations 1** to **4**. Also, pyrite oxidation results in swelling, which in turn causes the breakage of coal particles, thus increasing the surface area for enhanced oxidation.

Pyrite plays a vital role in coal spontaneous combustion. The specific heat of pyrite is only one-third of that of coal; but with the same heat absorption, the temperature rise of pyrite is three times higher when compared to coal (**Deng et al., 2015**). Moreover, pyrite does not react effectively without the presence of moisture and in a dry state, it does not contribute to the thermal runaway process (**Arisoy and Beamish, 2015**).

Coal with an increasing reactive pyrite content does not reach thermal runaway any faster in a dry state, but in a moist state, it does. So, the key exothermic pyrite reaction takes place with oxygen in the presence of moisture.

Several equations for the reactions of pyrite and oxygen, with the help of moisture are available in literature. For example, the reaction for pyrite oxidation has been given in **Equations 1** to **4**.

Table 1: The most famous and important studies about the effect of moisture and pyrite during coal spontaneous combustion

References	Pyrite	Moisture
Parr & Hilgard, 1925	●	
Stott, 1960		●
Hodges & Hinsley, 1964		●
Nandy et al., 1967		●
Bhattacharyya et al., 1968		●
Guney, 1971		●
Bhattacharyya, 1971		●
Bhattacharyya, 1972		●
Nandy et al., 1972		●
Sondreal & Ellman, 1974		●
Nordon & Bainbridge, 1983		●
Banerjee, 1985		●
Ghosh, 1986	●	
Li & Skinner, 1986		●
Cole et al., 1987	●	
Riley et al., 1987	●	●
Weise et al., 1987	●	
Chandra & Prasad, 1990	●	
Miron et al., 1992	●	
Chen & Stott, 1993		●
Arisoy & Akgun, 1994		●
Barve & Mahadevan, 1994		●
Bhat & Agarwal, 1996		●
Clemens & Matheson, 1996		●
Vance et al., 1996		●
Walters, 1996	●	
Ren et al., 1999		●
Akgun & Essenhigh, 2001		●
Jones, 2001		●
Panigrahi & Saxena, 2001		●
Kadioglu & Varamaz, 2003		●
Kucuk et al., 2003		●
Beamish & Hamilton, 2005		●
Beamish et al., 2005		●
Smith & Glasser, 2005		●
Nelson & Chen, 2007	●	
Singh et al., 2007	●	●
Beamish & Schultz, 2008		●
Beamish & Beamish, 2010		●
Beamish & Beamish, 2011	●	●
Wen, 2011	●	
Beamish & Beamish, 2012		●
Beamish et al., 2012	●	
Sasaki et al., 2014		●
Arisoy & Beamish, 2015	●	●
Beamish & Theiler, 2015		●
Deng et al., 2015	●	
Zhang et al., 2016		●
Zhao et al., 2016		●
Arisoy et al., 2017		●
Beamish & Theiler, 2017	●	●
Yang et al., 2017		●
Wang et al., 2018		●

$$FeS_2 + 8H_2O + \frac{7}{2}O_2 \rightarrow FeSO_4.7H_2O + H_2SO_4$$

$$(\Delta H = -1465.49\text{kJ}) \qquad (1)$$

(Weise et al., 1987)

$$FeS_2 + 8H_2O + 7O_2 \rightarrow FeSO_4.7H_2O + SO_4^{2-} + 2H^+ \qquad (2)$$

(Beamish et al., 2012)

$$FeS_2 + 7O_2 + 16H_2O \rightarrow 2H_2SO_4 + 2FeSO_4.7H_2O + 1321\ \text{kJ} \qquad (3)$$

(Arisoy and Beamish, 2015)

$$FeS_2 \xrightarrow{O_2+H_2O} FeSO_4 \xrightleftharpoons[O_2+H_2O+FeS_2]{O_2+H_2O} Fe_2(SO_4)_3 \qquad (4)$$

(Deng et al., 2015)

The above equations suggest that oxygen and moisture are two prime weathering parameters, which contribute to the pyrite alteration shown and it also leads to the formation of sulphuric acid as a by-product of the alteration process. The presence of moisture doubles the reactivity rate of coal and the presence of pyrite in dispersed form 10 folds the actual reaction rate (**Mahananda, 2014**).

These reactions can occur at low temperatures and additionally, all of the reactions are exothermic reactions. The heat generation from these reactions doubles the amount of heat generated by coal with the same amount of oxygen (**Garcia et al., 1999; Martínez et al., 2009**). So, the presence of pyrite along with moisture is a big promoter for coal spontaneous combustion and can change the propensity of coal towards spontaneous combustion.

Different countries have adopted different methods to assess the propensity of coals to spontaneous combustion in the laboratory for their applicability. The objective of different entities of mining operations is to assess the hazard of spontaneous combustion and its related risk to ensure the safety of miners and machines.

The criteria for the determination of spontaneous combustion should be the purpose of investigation and its use. Mining planners need information regarding the assessment and prediction of spontaneous combustion prior to design whereas operators need information during mining activity or when moving onto new areas or seams. All of these investigations have attempted to characterize coal samples in the laboratory to identify the mechanisms that govern the susceptibility of a particular coal to spontaneously heat under field conditions. The reliability and repeatability of these test results to its applicability or with field conditions/mechanisms play an important role for the selection of testing methods. Similarly, the cost, duration of an experiment and expertise of the user are of the most concern for different testing methods of spontaneous combustion (**Mohalik et al., 2016**). A certain amount of instrumental methods and techniques for the characterization of a coal's propensity to spontaneous combustion exist. Two of the most important methods are the crossing point temperature (CPT) and R70 test methods, which have been used frequently. CPT and R70 test methods are thermal methods used to determine the tendency of coal spontaneous combustion. This rate of the rise in coal temperature during coal oxidation becomes greater under the appropriate conditions. In this study, in order to comprehensively investigate the influence of pyrite along with moisture on coal spontaneous combustion, characteristics on the acceleration of the coal spontaneous combustion process, temperature-programmed experiments were carried out using CPT and R70 test methods.

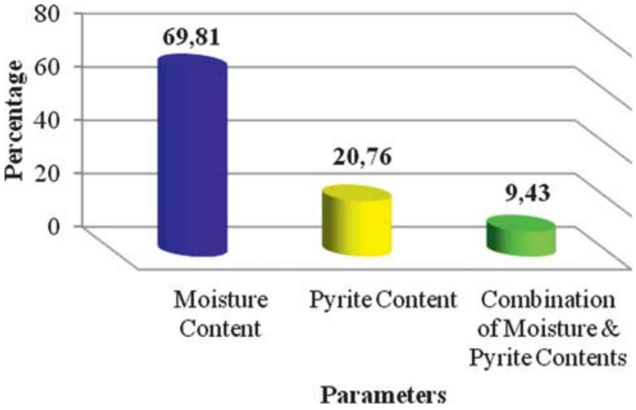

Figure 2: Percentage of studies done on parameters (Moisture, Pyrite and Combination of Moisture and Pyrite) in literature

2. Materials and methods*

2.1. Coal samples and preparation

In this work, six coal samples (from the Eastern Alborz coal mine) were freshly collected directly from the working face of the mine, after removing a coal seam of approximately 25 cm thickness. In order to avoid the possibility of peroxidation in the samples, they were sent to an air sealed plastic container that was completely filled with nitrogen and contained approximately 5 kg of coal, mostly in lump form. The samples were transported to the laboratory in an ice-filled insulated container to avoid peroxidation.

The samples were delivered to the laboratory as rapidly as possible. Upon arrival, the samples were transferred to a freezer for storage until required for testing. In order to minimize unnecessary oxidation, the samples were maintained in lump condition and were kept undisturbed in the laboratory prior to each test. After the test facility was ready, the plastic bag was unwrapped, its surface was removed and its interior core was crushed to obtain the samples. For the CPT test, the coal particles ranging in size from 0.18 mm to 0.38 mm were sieved for the experimental procedure. For the R70 test, the coal with particle in sizes of <212 μm were sieved for the experimental procedure. The CPT and R70 test method required 60 g (±0.01 g) and 150 g (±0.01 g) of crushed coal sample packed in the coal reaction vessel, respectively.

In order to minimize the effects of oxidation on fresh surfaces, coal test samples were ground and prepared just before each trial.

To emphasize the influence of pyrite and moisture on the characteristics of coal spontaneous combustion, samples used in the experiments were mixtures of coal, pyrite and moisture (pyrite and moisture content of coal samples were natural), and the other characteristics were similar, such as, the maceral and pyrite contents in sample No. 6, as shown in **Figure 3**, using a microscopic image.

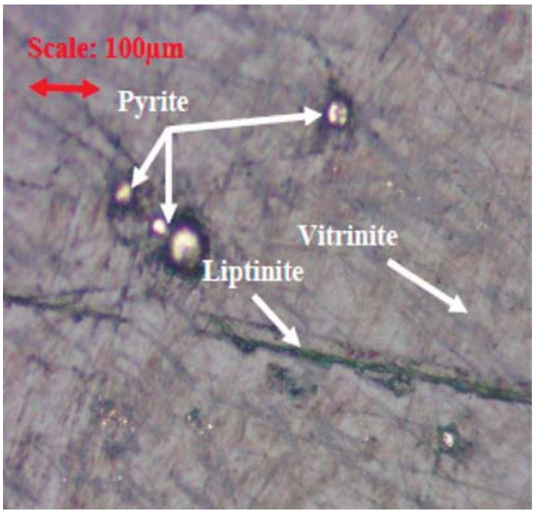

Figure 3: Macerals and minerals found in sample No. 6

2.2. Experimental apparatus and methods

A certain amount of instrumental methods and techniques for the characterization of coal propensity to spontaneous combustion exist. Two of the most widely used methods are CPT and R70 test methods. CPT and R70 test methods are thermal methods used to determine coal spontaneous combustion propensity. For the first time in the world, an experimental apparatus which can measure CPT and R70 test methods in separate tests was made in the Shahrood University of Technology in Iran, and this apparatus was used in this research work.

2.2.1. Crossing point temperature (CPT) method

The temperature at which the coal temperature begins to exceed the surrounding temperature is the so-called crossing point temperature (CPT) (**Xuyao et al., 2011**).

The CPT method is a very important way to reveal the mechanism of coal spontaneous combustion and it is still widely used today. In the experiment, a prepared coal sample is placed in a gauze container and the oven temperature is controlled. The progression of temperature with time in the process of a coal's reaction with air or oxygen and the oven temperature are recorded. When the coal sample temperature equals the linearly ramped oven temperature, the temperature is called CPT, as shown in **Figure 4**. CPT is used as an index to classify the propensity of a coal to spontaneous combustion. (**Chen, 1991**). The CPT method is widely used in India, Turkey, New Zealand South Africa, Poland and China and it has recently been improved (**Mohalik et al., 2016**).

The essence of this method is as follows. The coal sample is placed in a programmed adiabatic oven being heated at a constant rate. The oven is set to run at a constant temperature of 50 °C, while dry air with oxygen is permitted to flow through the coal reaction vessel at a rate of 50 mL/min. The temperature logger is used to continuously monitor the coal temperature and the surrounding temperatures. When the coal temperature reaches 50 °C, the oven is set to increase the temperature at a programmed rate of 1 °C/min while the flow rate of dry air is maintained at 50 mL/min. The experiment ends when the coal temperature is higher than the surrounding programmed adiabatic oven. When the coal sample temperature equals the linearly ramped oven temperature, the temperature is called CPT (**Nugroho et al., 1998; Xuyao et al., 2011**).

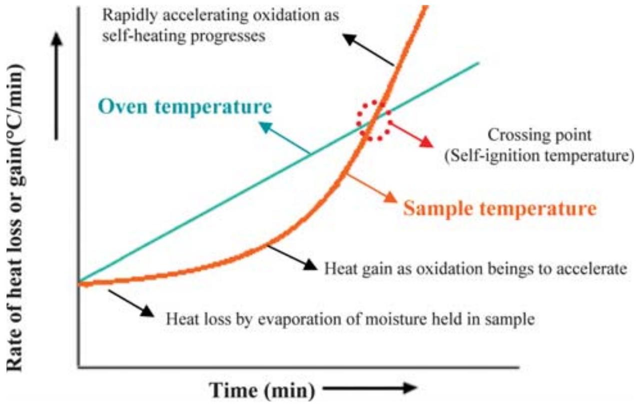

Figure 4: Schematic diagram of crossing point temperature (**Kim, 1995**)

2.2.2. R70 method

The essence of the R70 method is as follows. The ground (with particle sizes of <212 μm) coal sample is dried in the adiabatic oven at a temperature of 105-110 ºC for 16 hours under inert gas flow (nitrogen). Whilst still under nitrogen, the coal is cooled to 40 ºC in the same environment, and after that, it is stored in oxygen-rich air flow at a temperature of 40 ºC. Under the latter conditions, coal oxidation is being initiated and the processes of self-heating are observed. The average rate of the coal heating from 40 to 70 ºC is considered to be the index R70 (ºC/h). The higher value of such an index, the more prone the coal is to spontaneous combustion. Such a method is the most efficient one with respect to simple characterization of coal propensity to oxidation and self-heating (**Beamish et al., 2000, Arisoy and Beamish, 2015**).

2.2.3. Testing system

Figure 5 shows a schematic diagram of the apparatus applied for the low-temperature oxidation of coal. The testing system consists of an experimental apparatus for the simulation of coal oxidation. The instrument consists of the following, as shown in **Figure 6**:

- a temperature-programmed adiabatic oven (a temperature-programmed adiabatic oven can set up relevant parameters related to the range and the rise rate of the temperature, and keep the temperature constant. It is applied to control the temperature of the coal samples, whose temperature ranges from room temperature to 400 °C with a precision of 1 °C in the control);
- an electric heater;
- a fan (used to strengthen and maintain a uniform temperature convection in the oven);
- a coal sample reaction vessel (the coal sample reaction vessel is made of pure aluminum; it has very good thermal conductivity. The coal sample reaction vessel is respectively connected with an inlet for an air supply path, thermocouple for temperature measurement and an outlet for the air outlet path);
- a 15 m gas pre-heating copper tube (Preheating was achieved by passing the air through a copper tube located inside the programmed adiabatic oven);
- thermocouples (thermocouple 1, fixed at the center of the temperature-programmed adiabatic oven, is used to monitor the surrounding temperature while thermocouple 2, positioned in the middle part of the coal reaction vessel, is used to measure the coal sample temperature);
- JUMO Dicon touch, which consists of:
  - a data logger (the temperature changes in the coal sample with time was obtained by a data logging system for later analysis);
  - a micro-controller (the programmed adiabatic oven is set to increase the temperature with a micro-controller);
- a computer;
- a 50 kg N_2 gas cylinder (for preheating of the coal sample for the start test);
- a 50 kg O_2 gas cylinder (the air supply system sends gas into the reaction vessel, and takes gas after the reaction with the coal sample out of the reaction vessel along the exhaust pipe);
- a pressure reducing valve;
- a flow-meter.

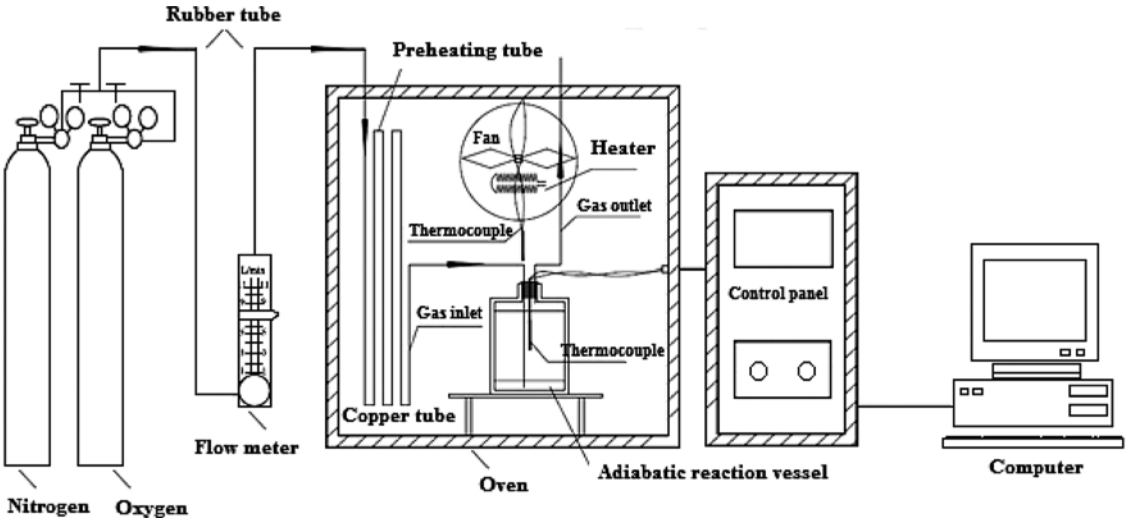

Figure 5: A schematic diagram of the apparatus applied for low-temperature oxidation

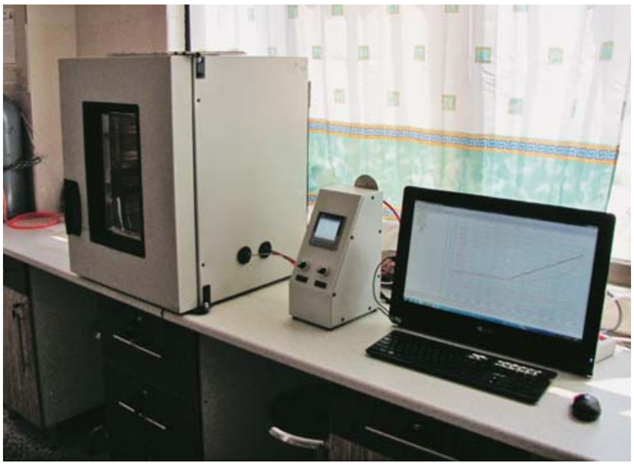

Figure 6: The testing system

Table 2: The coal analytical data for each sample

Sample No.	Moisture Content (%)	Pyrite Content (%)	CPT (°C)	R70 (°C/h)
# 1	0.768	0.11	190	4.27
# 2	1.265	0.20	200	2.50
# 3	13.288	5.30	93	10.20
# 4	32.230	1.2	195	3.95
# 5	35.300	0.00	226	1.85
# 6	19.950	5.7	73	11.25

3. Results and discussion

The curves of CPT and R70 test methods and results for each sample are shown in **Figure 7** and **Figure 8** as

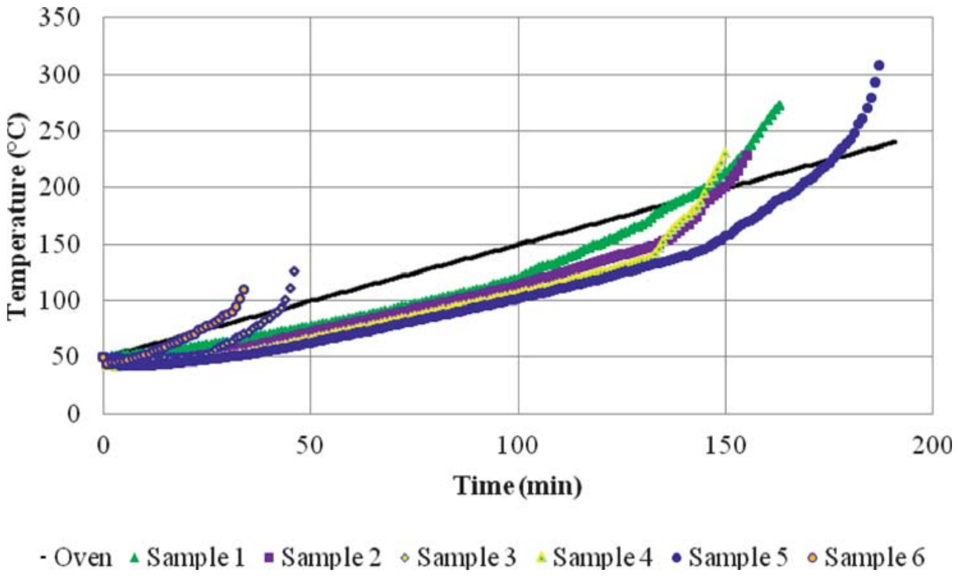

Figure 7: CPT test results for six different coal samples in terms of moisture and pyrite differences

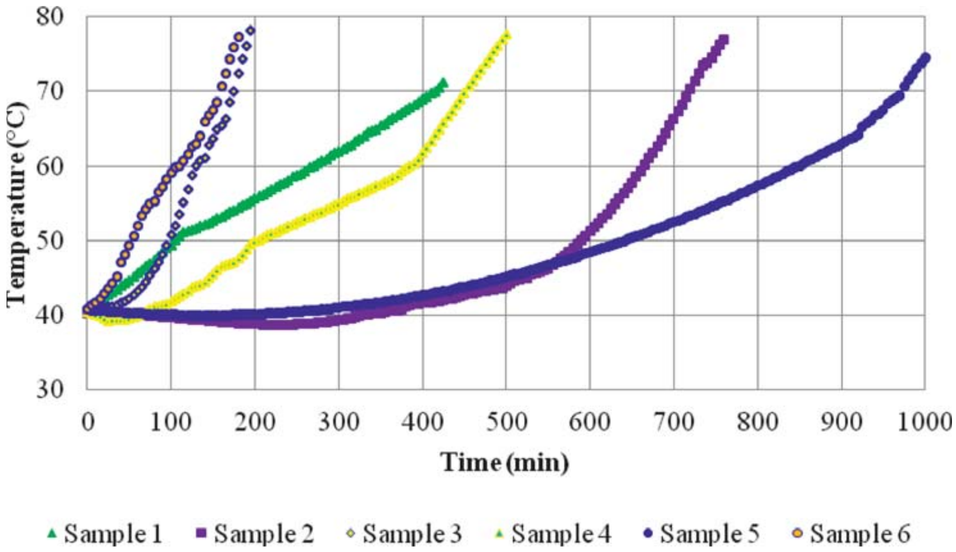

Figure 8: R70 test results for six different coal samples in terms of moisture and pyrite differences

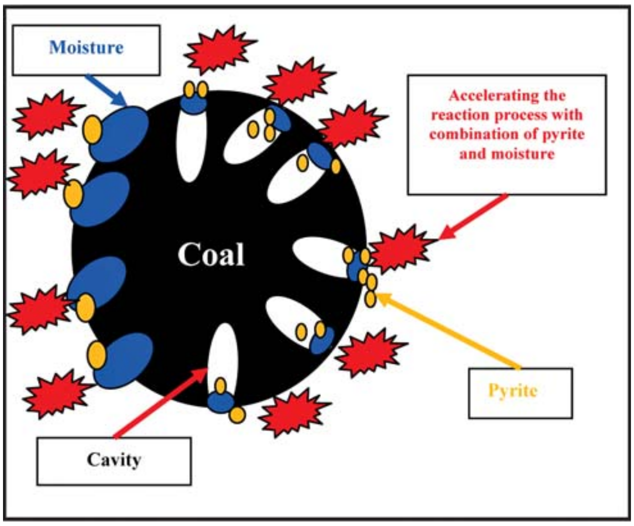

Figure 9: Effect of moisture and pyrite on accelerating the coal spontaneous combustion mechanism

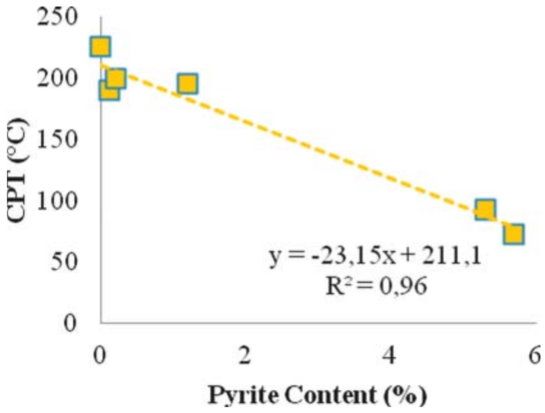

Figure 10: Results of Pyrite Content fit CPT test method

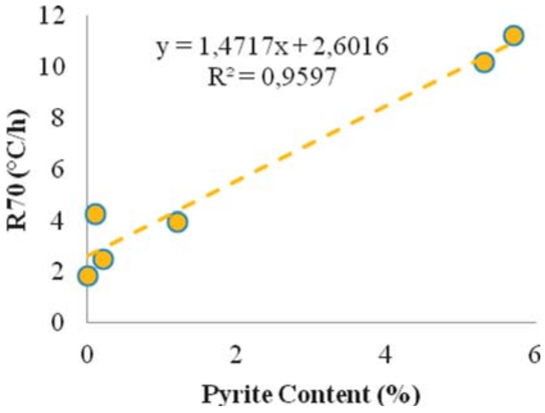

Figure 11: Results of Pyrite Content fit R70 test method

well as **Table 2**. It can be seen that the coal spontaneous combustion curves are different. This difference is due to the amount of moisture and pyrite contents. Their CPT values for samples are between 73 to 226 ºC, and their R70 values for samples are between 1.85 to 11.25 °C/h. Based on the coal moisture and pyrite contents, it could be expected that samples 3 and 6 would have a lower CPT and higher R70 values than the other coal samples. However, the higher pyrite contents and moderate moisture contents of samples 3 and 6 creates a heat effect, compared to the other samples. It must be remembered, that CPT and R70 test for the other samples has a higher and lower value, respectively in comparison to samples 3 and 6. The reason for this difference is low moisture and low pyrite contents in samples 1 and 2, and high moisture and low pyrite contents in samples 4 and 5. In samples 1 and 2, the amount of moisture and pyrite contents is very low and so, the exothermic pyrite oxidation reactions cannot form. In samples 4 and 5, the amount of moisture content is very high, and the heat loss due to moisture evaporation does occur and so, the oxidation mechanism cannot form. As shown in **Figure 9**, the existence of moisture and pyrite contents on coal

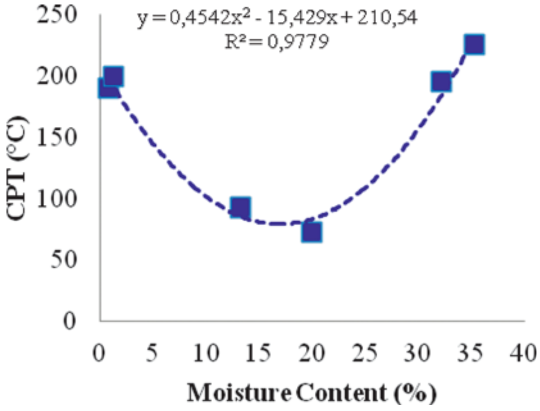

Figure 12: Results of moisture Content fit CPT test method

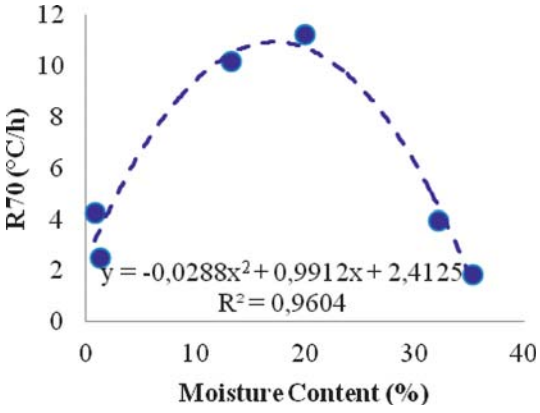

Figure 13: Results of moisture Content fit R70 test method

can accelerate the coal spontaneous combustion mechanism, which is described based on **Equations 1** to **4**.

According to **Equation 2**, the existence of moisture and high pyrite content would produce too much H^+, which means this reaction is strongly exothermic, and it can be seen that the pyrite oxidation reaction is dependent on the presence of both moisture and oxygen (**Beamish et al., 2012**).

Thus, samples 3 and 6 follow these equations and the coal spontaneous combustion rates were increased.

Figure 10 and **Figure 11** illustrate the impact of pyrite content on coal spontaneous combustion at different values of pyrite contents. The results show pyrite content can linearly accelerate the coal spontaneous combustion process, and it has a positive effect on coal spontaneous combustion.

There are clear distinctions between high moisture content and low moisture content on coal spontaneous combustion behaviours. As long as moisture content is under 20%, coal spontaneous combustion rates increase, which is due to its combination with pyrite, and this is an exothermic process, while it can reduce or restrict the coal spontaneous combustion rates in excess of 20%, which is due to heat loss during moisture evaporation (see **Figure 12** and **Figure 13**). So, the existence of moisture in excess of 20%, can absorb the heat and prevent the chemical reactions, which is an endothermic process.

4. Conclusion

The coal spontaneous combustion phenomenon is always a big challenge in coal industries, which is affected by many parameters such as pyrite and moisture contents. Despite extensive research carried out on this topic, the mutual effects of moisture and pyrite contents on the rate of coal spontaneous combustion is still not fully understood as stated in literature review. The adiabatic oxidation methods provide significant information on the process of coal spontaneous combustion under conditions close to those found in nature. The simultaneous effects of moisture and pyrite on coal spontaneous combustion using CPT and R70 test methods have been measured experimentally in this study.

The results of this work show that coal spontaneous combustion promotion accelerates with the combination of pyrite and moisture at the same time, so pyrite and moisture contents play an important role. Clearly, the presence of moisture content for the pyrite oxidation reaction is a major parameter in the overall spontaneous combustion propensity of coals containing reactive pyrite.

It is shown that an increase in pyrite content of coal has a marked effect on coal spontaneous combustion. In this research work, coal samples undergo oxidation most rapidly when the moisture content supply is under about 20%, and it can reduce coal spontaneous combustion in excess of 20%, because when moisture is present in excess of 20%, the heat released by oxidation is used to evaporate the moisture. So, at high moisture levels, there is too much water available and the self-heating is hindered by the heat loss from moisture evaporation. Hence the moisture content has a complex behaviour. These differences have practical implications for coal spontaneous combustion management. An applied result of this finding is that improved coal spontaneous combustion measures including pyrite and moisture contents of coal should be taken into account, because their contents can tremendously promote coal spontaneous combustion.

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SAŽETAK

Simultani učinak vlage i pirita na spontano izgaranje ugljena analiziran metodama TPT i R70

Ugljen je stabilan izvor energije i ujedno najčešće korišteno fosilno gorivo za proizvodnju električne energije u svijetu. Unatoč brojnim koristima, eksploatacija ugljena sa sobom nosi i ozbiljne rizike kao što je spontano izgaranje (samozapaljenje) u rudnicima. Brojni čimbenici utječu na sklonost ugljena samozapaljenju. Pirit može povećati rizik samozapaljenja, a u kombinaciji s vlagom rizik dodatno raste. Takva kombinacija rijetko je opisana u literaturi. Stoga je ovim istraživanjem prikazano ubrzanje učinka reaktivnosti pirita i vlage na samozapaljenje. Oni su izmjereni primjenom točke prijelazne temperature (TPT) i testa R70. U tu svrhu u Iranu je osmišljen i izrađen novi uređaj. Prikazani su eksperimentalni podatci brzine reakcije pri čemu pirit ima dvostruko djelovanje. Najprije katalizira oksidacijsku reakciju, a zatim u vlažnome okruženju i sam oksidira postajući tako sekundarni izvor topline te na taj način ubrzava samozapaljenje ugljena. Budući da oksidacija pirita troši vlagu, postoji zajednički učinak ubrzavanja zagrijavanja, a manje topline troši se na isparavanje vlage. Rezultati su pokazali da sadržaj pirita može linearno ubrzati proces spontanoga izgaranja ugljena, dok ga sadržaj vlage manji od 20 % ubrzava, a veći od 20 % usporava. Rezultati ovoga istraživanja korisni su pri procjeni i upravljanju spontanim izgaranjem ugljena u rudnicima.

Ključne riječi:
spontano izgaranje ugljena, vlaga, pirit, TPT, R70

Authors contribution

Amir Safari (Ph.D. Candidate): initialized the idea, completed literature review and participated in all work stages, such as providing coal samples, running experimental tests and data analysis. **Farhang Sereshki** (Full Professor): executed experimental tests, data analysis and test of its accuracy, and helped with field work. **Mohammad Ataei** (Full Professor): managed the whole process and supervised it from the beginning to the end.