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Examination of the role of moisture content on the spontaneous combustion of coal (SCC)

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Abstract

The percentage of moisture content is one of coal's inherent characteristics, which has a crucial aspect in the occurrence of the spontaneous combustion of coal (SCC) and has long been an attractive issue for analysis among scientists. A complete understanding of this phenomenon helps experts in the prediction and prevention of its occurrence. For this reason, the present work predicts this phenomenon based on moisture content. Thus, a final conclusion about the effect of this parameter was determined. Fifty-one coal samples with different percentages of moisture content were collected from underground coal fields in Iran for the practice training and testing of crossing point temperature (CPT) and R70 test methods. Then, the method of regression analysis was used for modelling and predicting the SCC tendency. The results showed that coal samples were oxidized most rapidly when the moisture content supply was under 20%, and it can reduce the prevalence of the SCC in situations when the moisture was in excess of 20% because the heat released by oxidation is used to evaporate the moisture. For validation and testing, 13 coal samples of another coalfield were collected and CPT and R70 test methods were carried out. The results of the test methods were compared through regression equations. The results obtained from the models indicate a good appropriate prediction in terms of the determination of the SCC tendency by regression analysis.

Keywords:
pollution, spontaneous combustion of coal (SCC), moisture content, crossing point temperature (CPT), R70.

1. Introduction

The Spontaneous Combustion of Coal (SCC) is a universal hazard in coal industries. In this process, coal exposed to the air reacts with oxygen, gets rapidly oxidized, and starts an open flame. This process can appear anywhere in all divisions of coal mining. Environmental components are extremely affected after the occurrence of the SCC. This phenomenon affects the direct failure of equipment, loss of coal reserves, releases noxious gases, heat, smoke, dust, and causes serious environmental, economic, and social threats (**Saffari et al., 2013; 2017; 2019a**).

Due to coal's intrinsic characteristics, which include moisture content, the phenomenon has occurred in coal mines. (**Xuyao et al., 2011**). The moisture content has an important role in this phenomenon and has remained unclear despite the fact that it has been studied over a long period of time by researchers. The moisture content of coal influences coal oxidation, and there are opposite effects of moisture content on the SCC (**Bhat and Agarwal, 1996; Liang and Wang, 2003; Xu et al., 2013**). By some, the presence of moisture content has been considered as absolutely required before the SCC will take place; others have said that moisture merely facilitates or accelerates the self-heating that would take place without it; while still others claim that even a small amount of moisture will prevent the SCC.

Researchers have already handled research on how moisture content affects the SCC liability, and a summary of their results is shown in **Table 1**.

Table 1, shows how moisture plays a different role in coal oxidation, but the results of these studies are contradictory and moisture content has a complex influence on this phenomenon. So a comprehensive study for the evaluation of the effect of moisture content on this phenomenon is mandatory. Therefore, in this work, 51 coal samples with different percentages of moisture content were collected from underground coal fields in Iran (Tabas Parvadeh coal mine, Eastern Alborz coal mine, and Kerman coal mine) for the practice training and testing of crossing point temperature (CPT) and R70 test methods. These tests were carried out for each of the coal samples, and the results of general regression analysis equations were compared to the moisture content and these indexes. Then for the validation and testing of these equations, 13 coal samples from the Central Alborz coal mine, which is one of the coal fields in Iran, were collected and CPT and R70 test methods for each

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Table 1: Different comments about the effect of moisture content on the SCC propensity

Reference	Moisture content accelerates the SCC	Moisture content decelerates the SCC
Jones and Townend (1949)	●	
Berkowitz (1951)	●	
Yohe (1958)	●	
Hodges and Hinsley (1964)	●	
King et al. (1964)	●	
Nandy et al. (1967)	●	
Bhattacharyya et al. (1968)	●	
Banerjee et al. (1970)		●
Bhattacharyya (1972)		●
Sondreal and Ellman (1974)	●	
Hodges et al. (1976)	●	
Armstrong (1979)		●
Schmal et al. (1985)		●
Brooks and Glasser (1986)		●
Li and Skinner (1986)		●
Buckmaster and Kudynska (1992)	●	
Kudynska and Buckmaster (1992)	●	
Reich et al. (1992)		●
Chen and Stott (1993)	●	
Arisoy and Akgün (1994)		●
Bhat and Agarwal (1996)	●	
Clemens and Matheson (1996)	●	
Krishnaswamy et al. (1996)		●
Vance et al. (1996)	●	
Ceglarska-Stefanska et al. (1998)	●	●
Gong et al. (1999)	●	
Kawatra and Hess (1999)	●	
Ren et al. (1999)		●
Akgun and Essenhigh (2001)		●
Kaymakci and Didari (2001)	●	
Kadioğlu and Varamaz (2003)	●	
Küçük et al. (2003)	●	
Wang et al. (2003)	●	●
Beamish and Hamilton (2005)		●
Beamish et al. (2005)		●
Pone et al. (2007)	●	●
Singh et al. (2007)	●	
Majumder et al. (2008)	●	
Li et al. (2009)	●	
Wang et al. (2009)	●	
Beamish and Beamish (2010)	●	
Beamish and Beamish (2011)	●	
Saffari et al. (2013)	●	
Wang et al. (2013)	●	●
Xu et al. (2013)	●	●
Panigrahi and Ray (2014)	●	
Choudhury et al. (2016)		●
Onifade and Genc (2018)	●	
Wang et al. (2018)	●	
Wu et al. (2018)		●
Saffari et al. (2019b)	●	●

of the coal samples were carried out, and the results of the test methods were compared with regression equations.

2. Materials and methods

2.1. Coal samples and Laboratory studies

The crossing point temperature (CPT) and R70 test methods are used successfully for the evaluation of the SCC propensity. In this study of datasets, 64 coal samples were collected and laboratory tests (moisture content, CPT, and R70 test methods) were conducted.

Coal samples were freshly collected directly from the worked face of the mine. The samples were collected and transferred directly to the laboratory to avoid the possibility of peroxidation, along with the required amount of samples for each experiment, all in accordance with the methods provided by **Xuyao et al. (2011)**, and **Zhang et al. (2016)**.

After removing a coal seam of approximately 25 cm thickness to avoid the possibility of peroxidation, the samples were sealed in airtight plastic bags, immediately after collection to avoid oxidation, which was completely filled with nitrogen and contained approximately 5 kg of coal, mostly in lump form. An airtight bag is produced using aluminum-coated polyester, which allows for the maximum reduction of oxygen permeability. Each sample bag was clearly labelled with a chosen number. The samples were delivered to the laboratory as rapidly as possible in an ice-filled insulated container to avoid their peroxidation. Upon arrival, they were stored and prepared under laboratory conditions (room temperature). 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. In order to minimize the effects of oxidation on fresh surfaces, a coal test was carried out on the samples just before each run.

For the CPT test, coal particles ranging from 0.18 mm to 0.38 mm were sieved to provide experimental procedure. For the R70 test, coal with particle sizes of <212 μm was sieved to provide experimental procedure. The CPT and R70 test method require 60 g (±0.01 g) and 150 g (±0.01 g) of crushed coal samples, which were packed into the coal reaction vessel, respectively. In order to minimize the effects of oxidation, samples were ground and prepared just before each run.

Samples were extracted from several coal fields in Iran. Fifty-one coal samples intended for training were collected from the Tabas Parvadeh coal mine, the Eastern Alborz coal mine, and the Kerman coal mine. General equations were applied to these samples in order to evaluate the relationship between moisture content and CPT, and R70 indexes. Then for the validation of these

Figure 1: Locations of Case studies (**http://earth.google.com/, 2018**)

Table 2: Basic descriptive statistics of the moisture content for this study

Statistic	Value	Percentile	Value
Sample Size	51	Min	0.244
Range	35.056	5%	0.5724
Mean	5.01	10%	1.0696
Variance	59.514	25% (Q1)	1.36
Std. Deviation	7.7146	50% (Median)	2.612
Coef. of Variation	1.5398	75% (Q3)	4.344
Std. Error	1.0803	90%	12.647
Skewness	2.9957	95%	30.808
Excess Kurtosis	8.4756	Max	35.3

equations, 13 coal samples were collected from the Central Alborz coal mine for testing. The Central Alborz coal mine was also one of the coal fields in Iran where samples were not only collected but CPT and R70 test methods for each of the coal samples were carried out, and the results of the test methods were compared with regression equations.

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 content 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. The principal coalfields are located at Alborz (Eastern Alborz and Central Alborz) in the North and at Kerman and Tabas in central Iran (**Thomas and Thomas, 2002**).

Figure 1 shows the locations of the case studies. Basic descriptive statistics of the moisture content for this study are given in **Table 2**.

2.2. *Experimental apparatus and methods*

2.2.1. Crossing point temperature (CPT) method

The essence of this method is as follows. A coal sample is placed in a reactor in an oven which is being heated at a constant rate. The programmed adiabatic oven was set to run at a fixed temperature of 50 °C while dry air with oxygen (oxygen concentration was 99.999%) was passed to flow through the coal reaction vessel at a rate of 50 mL/min. The temperature logger was used to continuously monitor the coal and surrounding temperatures. When the coal temperature achieved 50 °C, the programmed adiabatic oven was set to raise 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 ended when the coal temperature was higher than the surrounding programmed adiabatic oven and when the coal sample temperature equalled the linearly ramped oven temperature. This temperature is called the crossing point temperature (CPT) (**Nugroho et al., 1998; Wang et al., 2009; Xuyao et al., 2011; Mohalik et al., 2016**), as shown in **Figure 2**.

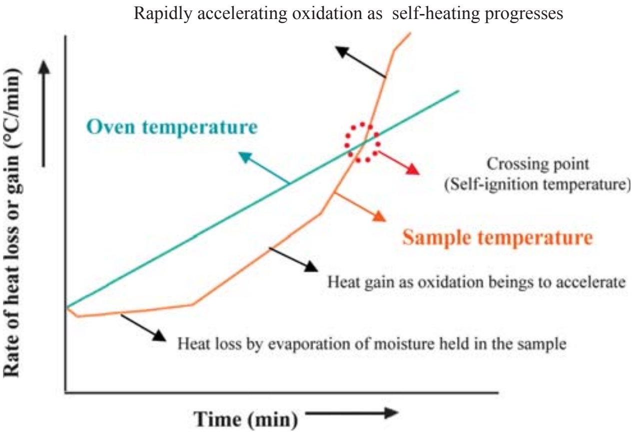

Figure 2: Schematic diagram of crossing point temperature (**Kim, 1995**)

2.2.2. R70 method

The essence of the R70 method is as follows. A ground coal sample (with particle sizes of <212 µm) is dried in an adiabatic oven at a temperature of 105-110 ºC for 16 hours under inert gas flow (nitrogen). Whilst still under nitrogen flow (nitrogen concentration was 99.999%), the coal is cooled to 40 ºC in the same environment, and after that, it is stored in oxygen-rich air flow (oxygen concentration was 99.999%) at a temperature of 40 ºC. Under the latter conditions, coal oxidation is initiated and processes of self-heating are 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 such an index, the more prone the coal is to spontaneously combust. Such a method is the most efficient one with respect to the simple characterization of a coal's propen-

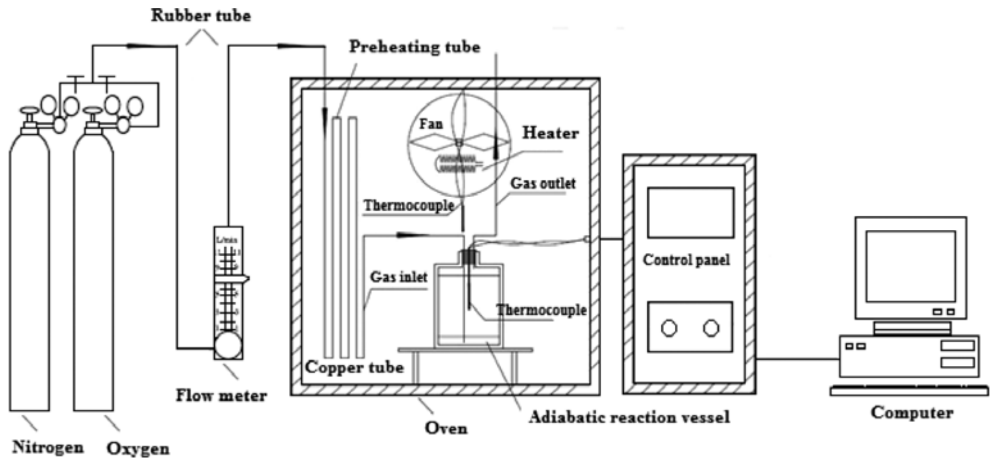

Figure 3: A schematic diagram of the apparatus applied for low-temperature oxidation

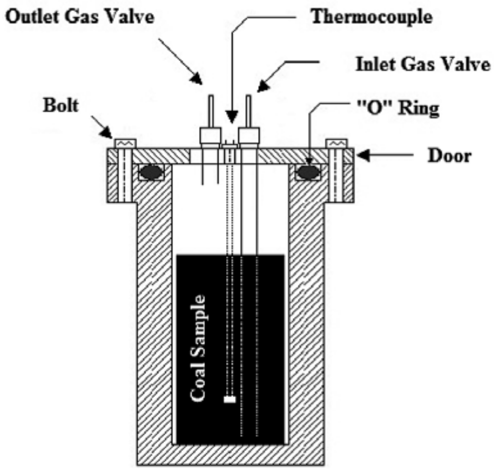

Figure 4: Sample container (bomb)

Figure 5: The testing system

sity to oxidation and self-heating (**Beamish et al., 2000, Arisoy and Beamish, 2015**).

2.2.3. Testing system

Figure 3 shows a simplified diagram of the set utilized for the oxidation of coal. **Figure 4** shows a schematic sample container (bomb) of the apparatus. The apparatus consists of the following, as shown in **Figure 5**, and was made in the Shahrood University of Technology in Iran (Faculty of Mining, Petroleum & Geophysics Engineering).

- a temperature-programmed adiabatic oven;
- an electric heater;
- a fan;
- a sample container (bomb) (made of pure aluminum and is respectively connected with an inlet for an air supply path, thermocouple for temperature measurement and an outlet for the air outlet path) (see **Figure 4**);
- a 15 m gas preheating 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), which consists of:
  - a data logger (for recording temperature changes in the coal sample over time);
  - a micro controller (the programmed adiabatic oven was set to increase the temperature with a micro controller);

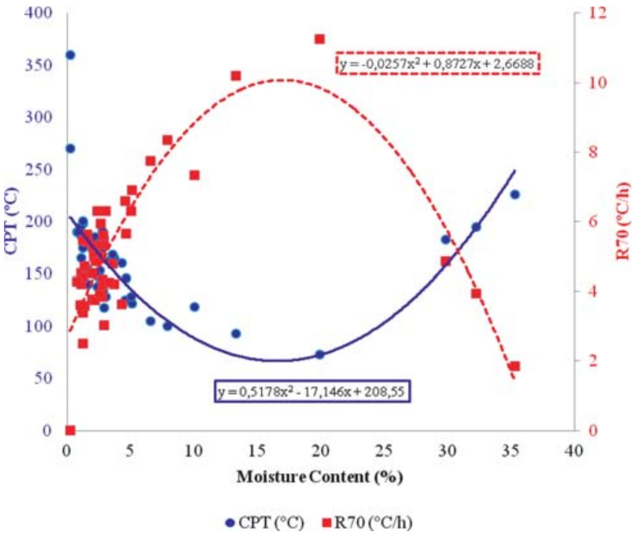

Figure 6: Integration results of moisture content fit R70 and CPT test methods

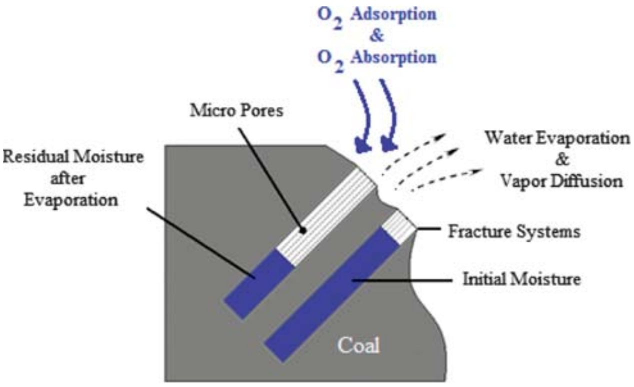

Figure 7: Water evaporation, O_2 adsorption, and O_2 absorption into micro-pores of coal, and acceleration of the SCC tendency

- a computer;
- a 50 kg O_2 gas cylinder (the air supply system);
- a 50 kg N_2 gas cylinder (for preheating of the coal sample to 50ºC to start the test);
- a pressure reducing valve;
- a flow meter.

3. Modelling and discussion

This section provides an optimal model for predicting the SCC tendency based on regression analysis. For modelling in this research, after conducting the experimental tests, a set of data including 64 experimental tests was collected, among which 51 test samples are selected as training data and the remaining 13 samples were se-

Table 3: Measured and predicted CPT values by the proposed regression model for testing datasets

Sample No.	Moisture Content (%)	Measured CPT (°C)	Predicted CPT (°C)	Measured R70 (°C/h)	Predicted R70 (°C/h)
1	1.233	180	188.20	3.03	3.71
2	1.435	180	185.01	4.52	3.87
3	0.918	170	193.25	3.80	3.45
4	3.297	150	157.65	5.45	5.27
5	1.830	180	178.91	4.12	4.18
6	0.470	175	200.61	2.15	3.07
7	1.100	160	190.32	3.64	3.60
8	0.990	210	192.09	2.30	3.51
9	3.700	140	152.19	6.10	5.55
10	1.318	195	186.85	2.98	3.77
11	2.005	165	176.25	3.22	4.32
12	13.435	85	71.74	10.15	9.75
13	30.125	175	161.94	4.90	5.64

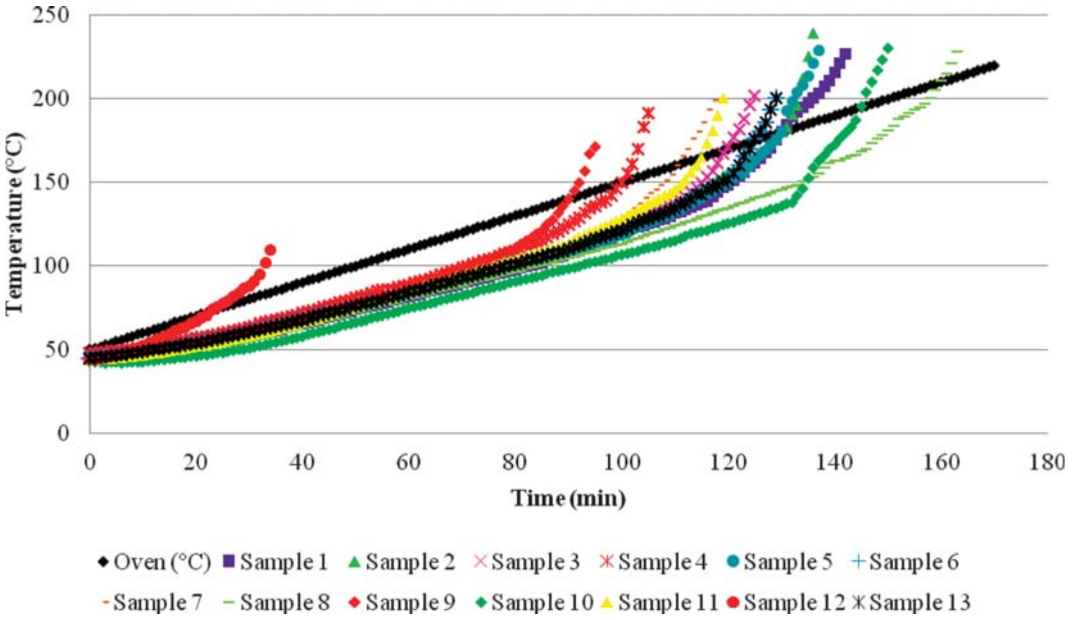

Figure 8: CPT test results for the Central Alborz coal mine samples (test data)

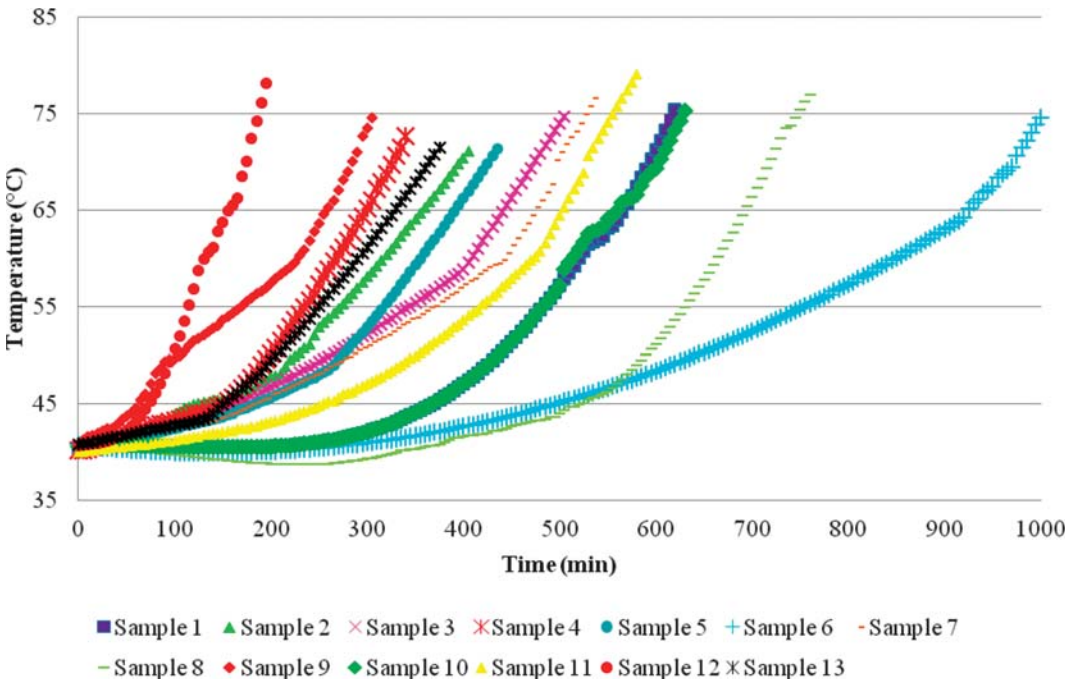

Figure 9: R70 test results for the Central Alborz coal mine samples (test data)

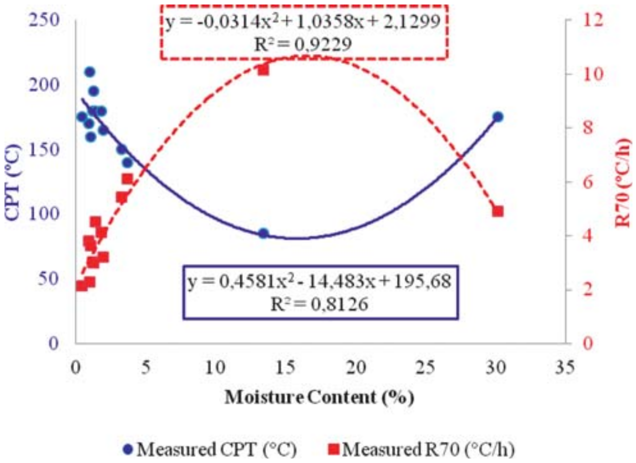

Figure 10: Results for the measured values of moisture content fit R70 and CPT test methods in the Central Alborz coal mine

lected as the test data. Additionally, as mentioned above, moisture content is considered as the input data and the R70 and crossing point temperature (CPT) test methods are the output parameters of the SCC tendency model.

The results of moisture content fit R70 and CPT test methods in Tabas Parvadeh, Eastern Alborz, and Kerman coal mines which were used for training data are shown in **Figure 6**, and regression analysis is given in **Equations 1** and **2**.

$$R_{70} = -0.0257M^2 + 0.8727M + 2.6688 \quad (1)$$

$$CPT = 0.5178M^2 - 17.146M + 208.55 \quad (2)$$

Where M is moisture content.

As shown in **Figure 6**, the SCC status shows two different points due to the moisture content of coal. Generally, the maximum propensity of SCC increases steadily for low-medium moisture content (about less than 20%) in coal. However, with high moisture content (in excess of 20%) in coal, the temperature increases rapidly at the beginning and then evaporation commences, thus the temperature approaches a steady state value. In this phase, heat liberation due to the chemical reaction and heat dissipation due to evaporation and other transfer mechanisms are approximately equal, so the propensity of the SCC is decreased. Also, the existence of moisture can obstruct oxygen from accessing the coal's surface. Hence, moisture in coal can play an inhibiting role in the oxidation process. Therefore, in this level, a safety issue develops due to the very high moisture content of coals.

It is considered that another inhibiting effect of moisture in high percentages is that the moisture condensed in pore channels blocks the oxygen diffusion toward the reaction sites in pore walls (**Wang et al., 2003**). Once oxygen molecules are accessible to almost all reaction sites in coal pores, moisture will play an active role in coal oxidation because it is necessary for the chemisorption reactions and chain reactions (**Clemens et al., 1991; Chen and Stott, 1993; Wang et al., 2002; Xu et al., 2013**). Thus, moisture's ability of heat-absorbing helps to cool down the coal's temperature. In addition, the water film formed isolates oxygen from coal and inhibits its spontaneous combustion (**Yang et al., 2017**). As shown in **Figure 7**, when high percentages of moisture exist in coal pores of coal, the moisture doesn't allow oxygen to react with coal, but when the moisture evaporates, the porosity of coal increases, and the oxygen is adsorbed into the micro-pores of coal, thus the temperature of coal increases, as well as the SCC tendency.

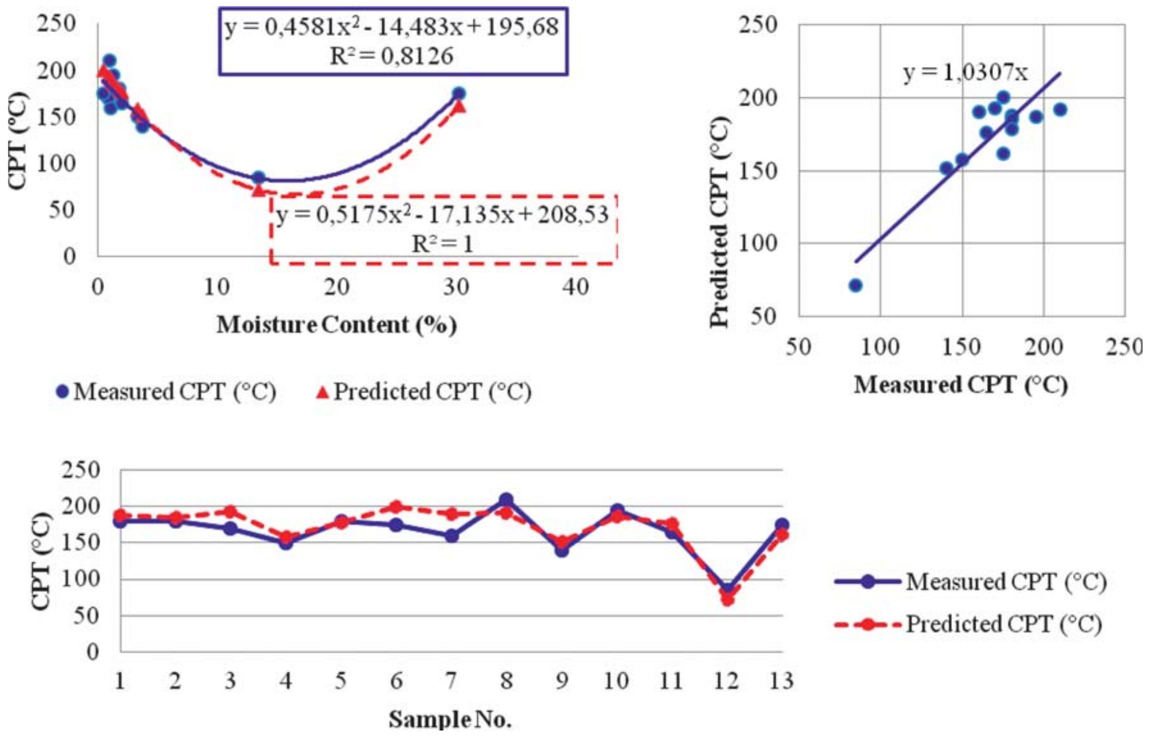

Figure 11: Comparison of the measured and the predicted values of the CPT test method for the test data

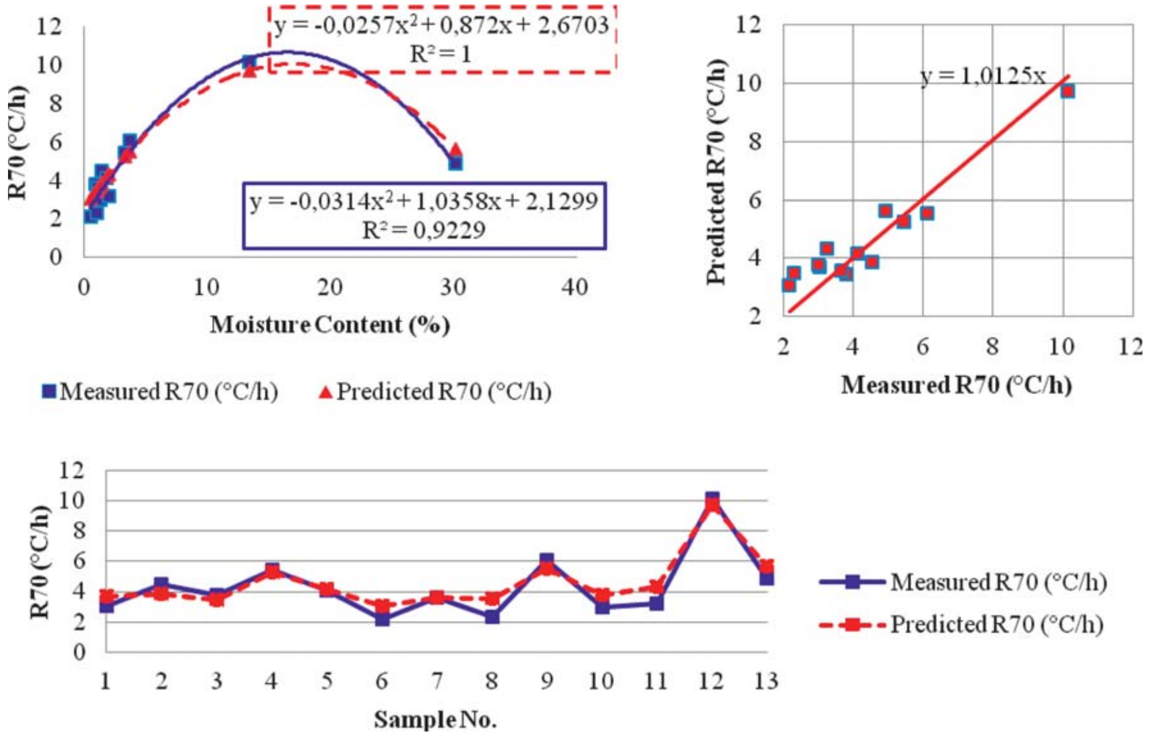

Figure 12: Comparison of the measured and the predicted values of the R70 test method for the test data

4. Validation of the proposed models

In this section, the performance of the developed prediction models was evaluated using the 13 collected coal samples from the Central Alborz coal mine in Iran. Crossing point temperature (CPT) and R70 test methods for each of the coal samples were carried out, and the results of the test methods are given in **Table 3** and **Figures 8** to **10**. Then based on the presented models in **Equations 1-2**, the CPT and R70 test methods for test-

Table 4: Statistical tests used to compare fitness estimates

Statistical parameter	Equation	Description
Root Mean Square Error (RMSE)	$RMSE = \sqrt{\frac{1}{n} \times \sum_{i=1}^{n} \left(Y_{Meas} - Y_{Esti}\right)^2}$	A smaller value shows a better fit (Ideal =0).
Variance Accounted For (VAF)	$VAF = \left[1 - \frac{\mathrm{var}\left(Y_{Meas} - Y_{Esti}\right)}{\mathrm{var}(Y_{Meas})}\right] \times 100$	A bigger value shows a better fit (Ideal=100).
Mean Absolute Percentage Error (MAPE)	$MAPE = \frac{1}{n} \sum_{i=1}^{n} \left\lvert \frac{Y_{Meas} - Y_{Esti}}{Y_{Meas}} \right\rvert \times 100$	A smaller value shows a better fit (Ideal =0).
Mean Absolute Error (MAE)	$MAE = \frac{1}{n} \times \left[\sum_{i=1}^{n} \left\lvert Y_{Meas} - Y_{Esti} \right\rvert\right]$	A smaller value shows a better fit (Ideal =0).
Variance Absolute Error (VARE)	$VARE = \mathrm{var}\left(\left\lvert Y_{Meas} - Y_{Esti} \right\rvert\right)$	A smaller value shows a better fit (Ideal =0).
In the above relations, n is the number of data; Y_{meas} and Y_{Esti} are measured and estimated values; *var* is the variances of the measured and predicted values.		

Table 5: The statistical tests for the proposed regression models

	Statistical				
Models	RMSE	VAF	MAPE	MAE	VARE
R70	0.69	89.92	18.16	0.59	0.0000
CPT	15.88	73.81	8.58	13.61	0.0017

ing data were predicted and the predicted values were compared with the measured values. **Table 3** and **Figures 11** to **12**, show this comparison between the predicted CPT and R70 using the proposed regression models with the actual CPT and R70 values.

In this work, the adequacy of the prediction model was also evaluated in terms of the coefficient of determination (R^2). **Figures 11** and **12**, show the relationship between the measured and predicted values, with a good coefficient of determination, obtained from the prediction models. A value for R^2 close to one shows a good fit of the prediction model, and a value close to zero presents a poor fit. As can be seen, the coefficient of determination was 1 for CPT and R70.

The criteria of the root mean square error (RMSE), the proportion of variance accounted for (VAF), the mean absolute percentage error (MAPE), the mean absolute error (MAE), and the variance absolute error (VARE) were used to evaluate the performance of models. These criteria are expressed in **Table 4**.

Furthermore, a prediction model is accepted as excellent when RMSE, MAPE, MAE, and VARE are equal to zero and VAF is 100%. The statistical values for the mentioned criteria, obtained from the proposed regression model, were calculated in **Table 5**. The statistical values for the mentioned criteria indicated an acceptable performance of the proposed model. Therefore, the output of these models can be considered as a preliminary estimation of the SCC propensity based on moisture content, and show the proper accuracy of these modelling.

5. Conclusions

After a comprehensive study of the literature review, it's clear that the percentage of moisture content is a very important factor in the occurrence of the SCC phenomenon. Therefore, for a final conclusion about the effect of this parameter on the SCC tendency, a comprehensive study was done. In this study, 51 coal samples with different percentages of moisture content for training data of overall underground coal fields in Iran were collected and crossing point temperature (CPT) and R70 test methods for each of the coal samples were carried out. Then, 13 coal samples for testing from another coal field in Iran were selected and used for validation of the regression equations. Coal samples were oxidized in an adiabatic oven in the "as received" state. The results show that moisture content can affect the SCC in two ways, which are, as the important discoveries arising out of this study, summarized below:

1) The presence of a low level of moisture in coals catalyses the iron, so this reaction accelerates. The results show that a coal sample oxidizes most rapidly when the moisture content supply is less than about 20%. So, moisture content (under 20%) in coal has a great influence on the heat released during coal oxidation at low temperatures. On the other hand, when the process of wetting is followed by evaporation, the coal pores are swelled and then cleared. With the pores cleared, a larger surface area is exposed for the coal to react with oxygen and this advances the process of SCC.

2) Moisture content in excess of 20% can reduce the SCC, 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 the moisture evaporation. As the moisture evaporates, the evaporation cools the surface temperature of the coal and this can delay or even stop the SCC process.

3) The results show that there is an explicit need for an assessment of the influence of moisture content in the SCC because moisture content has a complex behaviour which should be carefully analyzed.

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SAŽETAK

Ispitivanje uloge sadržaja vlage na spontano izgaranje ugljena

Sadržaj vlage jedna je od bitnih značajki ugljena koja ima presudan utjecaj pri spontanome izgaranju ugljena i dugo je atraktivna problematika brojnim znanstvenicima. Potpuno razumijevanje te pojave pomaže stručnjacima pri njezinu predviđanju i sprječavanju. Stoga je prikazano predviđanje te pojave temeljem sadržaja vlage, koja je iznimno bitna varijabla pri pojavi ovoga procesa. Prikazan je zaključak o učinku ovoga parametra. Prikupljen je 51 uzorak ugljena s različitim sadržajem vlage iz podzemnih rudnika ugljena Irana i provedene su metode ispitivanja točke prijelazne temperature (TPT) i R70 za svaki od uzoraka. Modeliranje i predviđanje tendencije spontanoga zapaljenja ugljena provedeno je uporabom regresijske analize. Rezultati pokazuju kako ugljen najbrže oksidira pri sadržaju vlage manjem od 20 % i da se spontano zapaljenje može smanjiti pri vlažnosti većoj od 20 %. Kod vrijednosti vlage od 20 % toplina koja se oslobađa oksidacijom troši se na isparavanje vlage. Validacija i testiranje provedeni su na 13 uzoraka ugljena pri čemu su na svakome od njih provedene metode TPT i R70, a rezultati uspoređeni regresijom. Regresijska analiza pokazala je kako se potencijalno samozapaljenje ugljena može dobro predvidjeti.

Ključne riječi:
onečišćenje, spontano izgaranje ugljena, sadržaj vlage, prijelazna temperatura, R70

Authors contribution

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