CHEMICAL ENGINEERING TRANSACTIONS
VOL. 46, 2015

A publication of
AIDIC
The Italian Association
of Chemical Engineering
Online at www.aidic.it/cet

Guest Editors: Peiyu Ren, Yancang Li, Huiping Song

ISBN 978-88-95608-37-2; **ISSN** 2283-9216

DOI: 10.3303/CET1546018

Experimental Study on Effects of Moisture on in Relation to Coal Oxidation and Spontaneous Combustion Characteristics of Mengba Coal at High Temperature Environments

Hu Wen, Jun Guo, Yongfei Jin*, Xiaowei Zhai, Kai Wang

School of Energy Engineering,Xi'an University of Science and Technology, Xi'an, 710054, China,
Key Laboratory of Western Mine and Hazard Prevention, Ministry of Education of China, Xi'an, 710054, China.
252038127@qq.com

Mengba coal is mined at a very high temperature and high humidity environment. Normally, the underground temperature is over 40℃, and there are a large amount of water flows out from coal seams. To study the effects of water inflow on the characteristics of coal spontaneously combusted at 40℃, the temperature-programmed experiments were carried out. Five different percentages of water content, which were 3.56%, 5.51%, 7.24%, 9.82% and 11.21%, were tested. Experimental results indicates that the coal samples with 9.82% and 7.24% percentages of moisture content could heat up to 40 ℃ fast, and continue to reach to 43 ℃ due to self-heating at the calorstat. On the basis of the temperature-programmed experiments, it was found that oxygen consumption rate, CO gas generation rate, and exothermic intensity coal samples with different percentages of the moisture content behave similar before the temperature less than 100 ℃. The oxygen consumption rate, CO gas generation rate, and exothermic intensity of coal samples from high to low was in the order of 9.82%, 7.4%, 5.51%, 2,21%, 11.21%, and 3.56% when the temperature was over 100℃. This finding confirmed that the moisture could involve into the complexing reaction of peroxides during low-temperature oxidation process of coal. The experimental results also indicates that the coal samples with 9.82% and 7.24% moisture content is easily inclined to spontaneous combustion and the indicator of the coal spontaneous combustion varies with the content of the moisture.

1. Introduction

As one of the most important reason resulting in fatal disasters, coal spontaneous combustion is a big threat to mining safety and environment pollution of coal mines all over the world. With the increasing complex mining conditions and geological structure, the impacts of underground temperature and water inflow to the spontaneous combustion of mine seam become more prominent. In a high geothermal environment, coal is more inclines to spontaneous combustion due to good conditions of heat storage and oxygen providing, and high capacity of exothermic oxidation (Guo, Xu, Deng, et al., 2001 and Deng, Wang, Zhai, et al, 2014). Compared to the mining conditions and geological structure influence of the moisture is even more complex. Zhang, Wang and Zhong (2007) thought that there could be water film formed on the surface of coal in situation where the moisture is over the saturation of coal. The water film could obstruct the contact between oxygen and coal. At the same time, evaporation could absorb heat to delay the self-heating of coal. A small of amount of water, however, would release wetting heat to promote the self-heating process of coal. From the standpoint of coal physical oxygen uptake inflection point temperature, and DSC "0" value point temperature of Barapukuria Coal Mine in different moisture content, Liu, Jin, Deng, et al. (2013) identified the influence of water on the characteristic temperature. Reich, Snook and Wagenfeld (1992) studied the fractal structure of lignite surface in the wet and dry conditions, separately. He thought that the external water could fill up the gap between particles and thus hinder the contact of coal with oxygen and the vapor pressure could impede the entry of air when the temperature is rising. Meanwhile, the external water would increase the volume and porosity of coal and increase the contact area of coal and oxygen when the water was evaporated. This could effectively promote the occurrence of coal spontaneous combustion. King, Krug and Zepf (1964), Jones and Townend (1949) and Li (1996) stated that the moisture acts as a catalyst during formation process of the

Please cite this article as: Wen H., Guo J., Jin Y.F., Zhai X.W., Wang K., 2015, Experimental study on effects of moisture on in relation to coal oxidation and spontaneous combustion characteristics of mengba coal at high temperature environments, Chemical Engineering Transactions, 46, 103-108 DOI:10.3303/CET1546018

carbon-oxygen-water complex. In the initial stage of coal spontaneous combustion, formation of water-oxygen complex is an exothermic process. The oxygen-water complex is chemical reactant of the coal spontaneous combustion at the late stage.

At present, many scholars have analyzed the influence of ground temperature and moisture on coal spontaneous combustion, separately. However, the study about the role of water in a high temperature is few. Bangladesh Balarpukulia Mine (hereinafter referred to as BCM) is an example. The initial temperature of float coal at BCM is above 40℃. Besides, the amount of water emission is large and could be up to 200 m^3/h. Meanwhile, amount of water emission varies with the arrangement position and the method of working face. The average thickness of coal seam is 40 m, and there are a large amount coal is left in the goat after mining because of coal mining technology. The coal of BCM is easy to spontaneous combustion. However, the spontaneous combustion characteristics are unknown in the high temperature and high water inflow conditions. Therefore, study on the influence of water inflow on coal spontaneous combustion at BCM is of importance.

To study effects of moisture content on the characteristic of coal spontaneous combustion, a series of constant temperature experiments and temperature-programmed experiments (Wen, Zhou, Zhai, et al., 2013. and Jin, Guo, Wen et al., 2015) were carried out. This study can provide some guidance to the predication of the risk of spontaneous combustion at BCM and other similar coal mines with high temperature and humidity.

2. Experiment (Methodology)

2.1 Experimental apparatus

The experimental system, which is made by Xi'an University of Science and Technology. As shown in Fig. 1, the system includes pneumatics, temperature control box and collection and analysis of air sample. The diameter of the homemade container filled with coal sample is 9.5 cm and the height is 25 cm, which is made of steel. The coal sample is put on the steel mesh. The air is preheated before passed into container. The container is placed in a controlled temperature-programmed heating box so that the coal temperature can be measured in real time. The gas sample is collected manually and analyzed by gas chromatography. At last, the variation of gas components can be gotten in the process of temperature- programmed experiment.

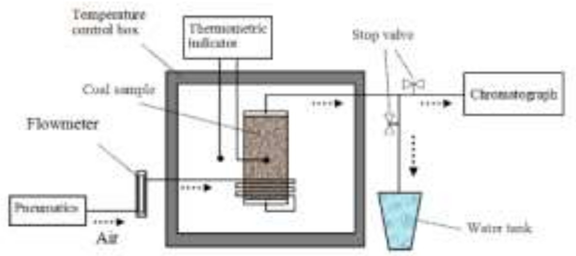

Figure 1: The structure of experiment device

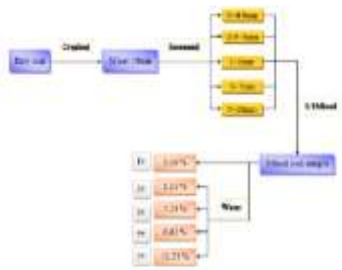

Figure 2: The preparation process of coal sample

2.2 Preparation of coal sample

After peeled off the outer surface, the coal of BCK is crushed in air and classified into five particle size ranges, which are 0-0.9 mm, 0.9-3 mm, 3-5 mm, 5-7 mm, and 7-10 mm. 200 g sample of each size range was collected and mixed together, evenly. Then, the mixed coal sample was splitting into five groups and different amount of water were added into each group. Coal samples with different moisture contents (3.56%, 5.51%, 7.24%,9.82%, and 11.21%) used in this study were prepared and kept in dry, sealed glass bottles prior to use in the experiments. The preparation process of coal sample is shown in Fig. 2.The water content of coal samples are shown in table 1.

Table 1: The water content of coal samples

Sample number	1#	2#	3#	4#	5#
Water content (%)	3.56	5.51	7.24	9.82	11.21

2.3 Experimental method and condition

Five coal samples were pre-heated at 40℃ ambient condition in the temperature-programmed oven. And the temperature and tested gas were collected every 5 min from room temperature until the temperature and gas composition of coal remain stable. Then the temperature- programmed experiment starts. Average particle size and void volume of coal sample are 4.18 mm and 0.545% respectively. The air flow rate is 120 ml/min, and heating rate of oven 0.3℃/min. At the same time, the composition of the gas at different temperature is analyzed with a gas chromatograph.

3. Results and discussion

3.1 Influences of moisture on temperature rising rate of coal at a constant temperature environment

The relationship of the heating rate and temperature of coal is shown in Fig. 3. In general, the heating rate of coal samples decreases with the increase of the temperature. According to the experiment time statistics, the coal sample with 9.82% of moisture content displays the biggest heating rate. Only 35 min is needed for it to reach to 40℃. More importantly, it can continue to reach up to 43.1℃ and kept the temperature constant at that temperature by self-heating. The order of coal samples needing time to reach to 40℃ from low to high is 9.82%, 7.24%, 11.21%, 5.51%, and 3.56%.

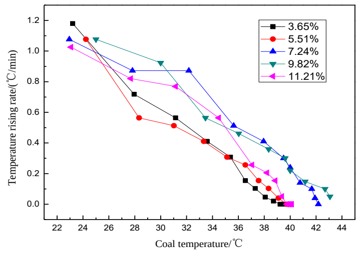

Figure 3: The temperature rising rate of coal samples with different water contents in the constant temperature environment

Since the external water is mainly in the cracks, porosity and surface of coal, in process of temperature rising from room temperature to 40℃, the water of coal when the external environment under constant moisture, can promote heating and generation of peroxide complexes in the process of coal oxidation reaction. At the same time, wetting thermal can be generated and then promote oxidation and heating of coal (Zhang, Wang and Zhong, 2007). In the other hand, there will be liquid film on the surface of coal whose water content is high and steam atmosphere formed by water evaporation can block the contact between oxygen and coal. The heat will be absorbed when the water is evaporated to inhibit the oxidation of coal self-heating. During the experiment, the coal with water contents of 9.82% and 7.24% can reach 40℃ in a short time and can be able to continue to self-heat. It indicates that the two contents of moisture can promote the Barapukuria Coal Mine coal samples to rapidly heat and thermal oxidation.

3.2 Effect of moisture on oxygen consumption rate

In the process of programmed-heating experiment, assuming that the temperature of coal in the oven is evenly and the airflow of import and export is ideal gas, the rate of oxygen consumption of coal can be estimated by analyzing the oxygen concentration of import and export (Xu, 2001) as follows:

$$V_{O_2}^{0}(T) = \frac{Q \cdot C_{O_2}^{0}}{\mathrm{V_m}} \cdot \ln \frac{C_{O_2}}{C_{O_2}^{0}} \qquad (1)$$

Where, $V_{O_2}^{0}(T)$ is the average oxygen consumption rate of coal in the fresh air conditions, mol/ (cm^3·s); Q is supplied air volume, ml: V_m is the volume of coal sample tube, cm^3; C_{O_2} and $C_{O_2}^{0}$ are O_2 concentration of air flow of fresh and export, mol/ cm^3.

Substituting the experimental data into the above formula can get curve of the rate of oxygen consumption with the temperature of coal, as shown in Fig. 4 and Fig. 5.

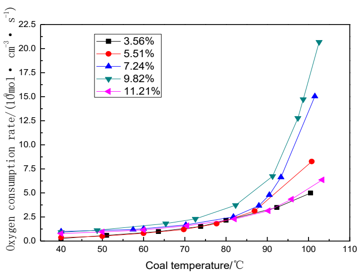

Figure 4: The oxygen consumption rate of coal samples of different water contents during 40~100℃

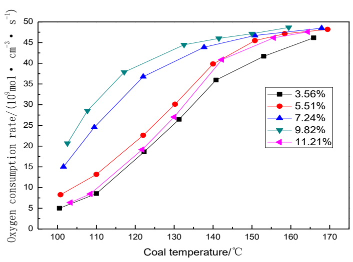

Figure 5: The oxygen consumption rate of coal samples of different water content over 100℃

As shown in Fig. 4 and Fig. 5, the rate of oxygen consumption, in general, increases with the coal temperature rising. Before 90-100℃, the rate of oxygen consumption increases slightly with the water content increasing regardless of moisture contents. The rate of 11.21% and 9.82% for coal is higher than other coals. This is mainly because of the greater water content which can produce more peroxide complex to need more oxygen. With the evaporation of water, more surface of coal is exposed to the increased area of contact between coal and oxygen after 90-100℃. At the same time, a large number of coal peroxide complexes involved in the oxidation reaction and a large water content of coal samples rapidly accelerates the rate of oxygen consumption, especially for coal samples with 9.82% and 7.24% water contents. However, the evaporation of moisture of coal takes away heat. Larger water content can reduce the internal energy of coal. The oxygen consumption rate of coal with water content of 11.21% is lower than other coal sample. The latter rate increases rapidly with the temperature of coal.

3.3 Effect of moisture on CO producing rate

As one of the major products of coal spontaneous combustion, the concentration of CO can characterize the development of coal spontaneous combustion. Fig. 6 shows the CO generation rate at cryogenic stage (40-100℃) for different coal samples, while Fig. 7 displays the results that temperature above 100℃.

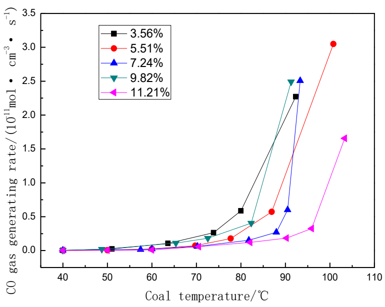

Figure 6: The CO gas generating rate of coal samples of different water content during 40~100℃

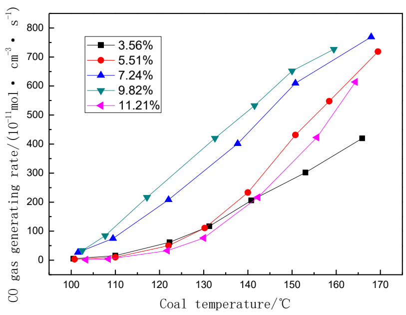

Figure 7: The CO gas generating rate of coal samples of different water content after 100℃

During the heating process, the CO producing rate of coal is related to the water content. With the increase of the temperature, the effects of the water content on the CO producing rate increases. At the early stage where the temperature is below 90 ℃, moisture mainly is involved into the formation of the peroxide complex in the oxidation reaction process and blocks the contact of coal with oxygen. Therefore, coal with higher moisture produces less amount of CO. Due to less external moisture, the coal sample with water content of 3.56% has more opportunities to contact with oxygen promoting adsorption of CO. After 90 ~ 100 ℃, because the hydro peroxide is formed when the peroxide complex is involved in the complex process (Jones, Henderson, Littlefair, 1998. and Deng, Liu, Zhai, 2011), coal sample with high water content generates more CO. However, for excessive moisture, the evaporation delays the coal oxygen compound effects when the water content in coal exceeds 11.21%. From the view of CO gas generation rate, optimum moisture content of coal spontaneous combustion is 9.82% and followed by 7.24% at the high temperature environment.

3.4 Effect of moisture on heating intensity of coal

Heating intensity is a characteristic parameter of coal spontaneous combustion, which could be calculated using chemical bond energy estimation method (Reich, Snook and Wagenfeld, 1992). The actual heating intensity is between the maximum and minimum heat intensity and has the same trend. The maximum heating intensity was selected as an indicator to compare heating intensity of coal samples in this paper. The maximum heat intensity curve of coal samples with different water contents for BCM is shown in Fig. 8.

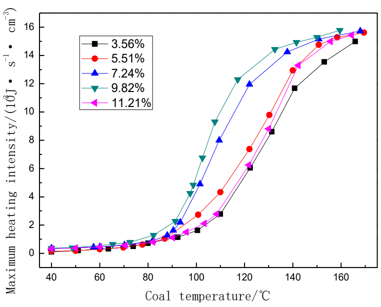

Figure 8: The maximum exothermal intensity of coal samples of different water content

As seen from Fig. 8, the variation of the maximum coal heating intensity and oxygen consumption rate are basically the same. At the constant temperature stage, coal samples of high moisture content accumulate peroxide complex. At the late stage of the low-temperature oxidation, oxygen consumption rate is high and the heat intensity increases with the increasing of the water content. Due to the effect of evaporation heat, heating intensity of coal with moisture content of 11.21% decreases after 90 ℃.Nevertheless, the effect of coal samples with water content of 9.82% and 7.24% is relatively small. In general, it promotes the oxidation and coal spontaneous combustion.

3.5 Effects of the moisture on the Index Gas

R = $\Delta CO / \Delta O_2$ was selected as coal spontaneous combustion index parameter, the variation of indicators for coal samples with different water content in heating oxidation process is studied based on the experimental data. The variation of R values in different temperature phase for coal samples with different moisture contents is shown in Fig. 9 and 10.

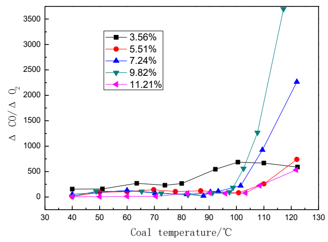

Figure 9: The $\Delta CO/\Delta O_2$ value of coal samples of different water content during 40~100 ℃

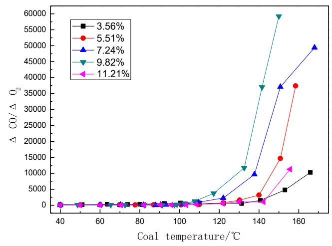

Figure 10: The $\Delta CO/\Delta O_2$ value of coal samples of different water content during the whole experiment

As it can be seen in Fig. 9 and Fig. 10, the indicators are different in different temperature for coal samples with different water contents. At the low temperature phase, difference of R value is small, CO generation is large and oxygen consumption is small when the water content was 3.56% in the coal samples. Therefore, R is relatively large. But after 100℃, oxidation of coal with water content of 9.82% and 7.24% increased and CO generation increases. Therefore, R value increases rapidly. These phenomena indicate that coal spontaneous combustion index parameters should be properly selected according to the size of the water for different water inflow of face.

4. Conclusions

1) Isothermal oxidation experiment results indicate that the coal samples with water contents of 9.82% and 7.24% can quickly reach 40℃ and can self-heat to 43℃ among tested five coal samples.
2) Before 90-100℃, the generation rate of CO decreases with the increase of water content, while the oxygen consumption rate and heat intensity rises when the water content increases. After 100℃, the order of coal spontaneous combustion characteristic parameters from high to low is 9.82%, 7.24%, 5.51%, 11.21, and 3.56%. It verifies that the moisture is mainly involved into the formation reaction of oxidation complex in the process of oxidation of coal at low temperature, and provides material basis for the latter oxidation.
3) According to the variation of spontaneous combustion characteristic parameters, such as the rate of oxygen consumption, CO, CO_2 generation rate and heat intensity, the coal of BCM with water contents of 9.82% and 7.24% has higher risk of spontaneous combustion.
4) Due to the high temperature at underground, BCM should selected proper parameters to predict the characteristics of the coal spontaneous combustion according to the water inflow and moisture characteristics of different working faces.

Acknowledgements

This paper is supported by the following funds: China postdoctoral science foundation (2014M552466); the national natural science foundation of China (51404195); Key project of national natural science foundation of China (51134019); the national natural science foundation of Shaanxi province (2014JM7276). Thanks sincerely.

References

Deng J., Wang K., & Zhai X.W., 2014, Research on the Effects of High Temperature Environment on Oxidation and Spontaneous Combustion Characteristics. *Safety in Coal Mines*. 45(3), 13-15. DOI: 10.13347 /j.cnki.mkaq.2014. 03.004.

Deng J., Liu W.Y., & Zhai X.W., 2011, Research on the Effects of Moisture on Oxidation and Spontaneous Combustion Properties of Mengba Coal Mine. Safety in Coal Mines. 42(11), 15-19. DOI: 10.13347/j.cnki.mkaq.2011.11.018.

Guo X.M., Xu J.C., & Deng J., 2001, Analyzing the effect of ground temperature on spontaneous combustion of coal. Journal of China Coal Society. 26(2), 160-162. DOI: 10.13225/j.cnki.jccs.2001.02.013.

Jones J. C., Henderson K. P., & Littlefair J., 1998, Kinetics parameters of oxidation of coals by heat-release measurement and their relevance to self-heating tests. Fuel. 77(2), 19222.

Jin Y.F., Guo J., Wen H., Liu W.Y., Wang K., Ma X.F., 2015, Experimental study on the high temperature lean oxygen oxidation combustion characteristic parameters of coal spontaneous combustion. Journal of China Coal Society, 40(3), 596-602. DOI: 10.13225 /j. cnki. jccs. 2014. 0626.

Jones R. E., Townend D. T. A., 1949, Oxidation of coal. Journal of Society Chemistry Industry.68 (2), 193-197.

King J., Krug D., Zepf D., 1964, The role of oxygen complex in oxidation of carbonaceous compounds. Soc Chem Industry Trans Farad Soc. 42(4), 297-299.

Liu W.Y., Jin Y.F., & Deng J., 2013, Experimental Study on Influence of Moisture on Coal's Characteristic Temperature in Mengba Mine. Ming Safety & Environmental Protection. 40(4), 1-3.

Li Z.H., 1996, Mechanism of Free Radical Reaetions in Spontaneous Combustion of Coal. Journal of China University Mining & Technology. 25(3), 111-114.

Reich M. H., Snook I. K., & Wagenfeld H. K., 1992, A fractal interpretation of the effect of drying on the pore structure of Victorian brown coal. Fuel, 71(6), 669 -672. DOI: 10.1016/0016-2361(92)90170-S.

Wen H., Zhou Y., Zhai X.W., 2013, Experimental Study of Coal Oxidation Process in Constant Temperature Condition. Safety in Coal Mines. 44(10), 1-3. DOI: 10.13347/j.cnki.mkaq.2013.10.013

Xu J.C., 2001, Determination Theory of Coal Spontaneous Combustion Zone. Beijing. China Coal Industry Publishing House. 63-64.

Zhang Y.T., Wang D.X., & Zhong X.X., 2007, Study on Influence of Water on Low-temperature Oxidation of Coal. Safety in Coal Mines. 396(11), 1-4. DOI: 10.13347/j.cnki.mkaq.2007.11.011.