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PRELIMINARY RESEARCH ON GRINDABILITY IMPROVEMENT OF COAL BY MICROWAVE PRETREATMENT

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Abstract: Microwave pretreatment is an effective method to promote grindability of coal samples along with moisture reduction. A contrast test was designed to explore a mechanism of grindability improvement process of coal by microwave pretreatment. Coal samples of 2–1 mm were investigated. The moisture content, size distribution and density composition of grinding products of dry coal with thermal drying (DTD), dry coal with microwave pretreatment (DMP) and wet coal with microwave pretreatment (WMP) were measured. A scanning electron microscope analysis showed that DMP had more micron-sized cracks than WMP, meanwhile, WMP had more cracks than DTD after microwave pretreatment and thermal drying. The results of grinding tests showed that yield of -0.125mm size fraction increased with the decrease of moisture content, and was equal to 43.11, 45.03 and 47.09% of DTD, WMP and DMP, respectively, after 15 minutes grinding for the same moisture content. The results showed that cracks caused by dehydration and selective heating characteristics were the main factors responsible for improvement of grindability, however, thermal evaporation of inherent moisture within the coal structure had almost no effect on grindability of coal.

Keyword: *coal, microwave pretreatment, grindability, moisture reduction, liberation*

Introduction

Grinding is generally considered to be the basic but inefficient operation of mineral utilization (Fuerstenau et al., 2004). Mineral composition, fragmentation force and stress intensity are important factors that affect grinding efficiency significantly. Many previous studies devoted to optimize the relationship between energy consumption and size reduction. Various kinds of comminution devices with different breakage mechanisms including ball mill, roll-crusher and high voltage pulses were used to

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study the comminution characteristics of different minerals to improve the efficiency of crushing (Ito et al., 2009; Xiao et al., 2012; Wang et al., 2012).

Microwave radiation is an effective method to reduce the stress intensity and promote fine grinding of ore. Minerals disseminated in an ore have different permittivities, which cause different response characteristics in the microwave field. Microwave heating has an advantage over conventional volumetric heating. It is a selective heating technique that can heat certain phases in a matrix (Samanli, 2004). Mineral matters with different thermal expansion coefficients have various heating rates due to their different dielectric constants and dielectric losses. It causes small cracks and micro-fissure in particles, and then leads the improvement of grindability when microwave is applied (Ruisanchez et al., 2012). Thus, microwave pretreatment has been proved to be an attractive method to improve the efficiency of comminution (Kingman and Rowson, 1998), and it has been widely studied in grinding, extractive metallurgy, dewatering of low-rank coal and flotation of oxidized coal (Kingman et al., 2004; Hong et al., 2009; Sonmez and Giray, 2011; Ge et al., 2013; Xia et al., 2013a; Xia et al., 2013b).

Coal is pulverized into fine particles through grinding to increase specific surface and this process is essential for effective utilization. Previously, several studies have investigated the influence of microwave pretreatment on coal grindability. Most of these studies show that significant increases in coal grindability and reduction of up to 50% in the work index could be achieved after microwave pretreatment (Haque, 1999; Lester and Kingman, 2004; Lester et al., 2005). Microwave pretreatment can significantly improve grindability of coal even at economic inputs that the energy consumption can be reduced by 40% (Lytle et al., 1992; Clark et al., 2000; Sahoo et al., 2011).

Generally, minerals disseminated in coal are mainly composed of clay minerals, quartz, calcite and pyrite. Except pyrite, which is good microwave absorption, the rest minerals and organic matters in coal are insulators that are transparent to microwave radiation. In addition, the moisture molecules in coal are polar, which are also a kind of microwave absorber. Usually the sulfur content in coal, including organic sulfur and inorganic sulfur, is lower than 4%, therefore the assumptions have been proposed that such low content cannot lead to huge improvement of grindability, and hence the moisture in coal plays an important role in improving grindability. Moisture reduction and thermal evaporation of inherent moisture within the coal structure are likely to be the reasonable factors which are responsible for the improvement of grindability. Some papers suggest that the internal pressure of the vapor caused by evaporate contributes to breaking up the ore and more moisture content lead to greater improvement of grindability (Marland et al., 2001; Farag et al., 2012).

The aim of this investigation is to explore reasons for grindability improvement, that is, crack generation, moisture content reduction and thermal evaporation of inherent moisture within the coal structure, in the process of microwave pretreatment. Three types of coal samples that are dry coal with thermal drying (DTD), dry coal with microwave pretreatment (DMP) and wet coal with microwave pretreatment (WMP) were prepared for this study. X-ray diffraction (XRD) and scanning electro microscope (SEM) were used to analyze the mineral composition of coal and find out the changes in the surface morphology of coal, respectively. The size composition and liberation degree of broken products were also measured.

Experimental method and procedure

Materials

A typical coal samples (A_{ad}=28.21%, S_t=3.12%, M_{ad}=7.86%) supported by Huaibei coal preparation plant from China was selected for the study. The mineral composition of coal sample was analyzed by an X-ray diffraction diagram (XRD, Bruker D8 advance, Germany), and results are shown in Fig.1.

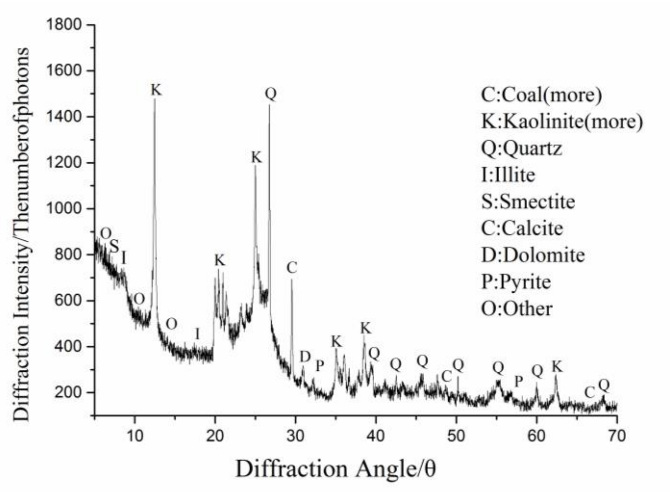

Fig. 1. X-ray spectrum of coal sample

As can be seen from Fig. 1, the main minerals in the coal samples are kaolinite, quartz, illite, smectite, calcite, and small amounts of dolomite and pyrite. Microwave radiation can be transmitted by clay minerals, quartz and calcite, however pyrite with low content is considered to be the best microwave absorber. Furthermore, water within coal structure also has a good ability of absorbing microwave (Marland et al., 2000).

The as-received samples were sized and the size fraction of 2-1 mm was selected as the research object.

The as-received coal had a moisture content of 7.86%. Coal samples with a thickness of 60 mm were processed in the microwave field with different times and cooled to room temperature, and then sealed into sample bag. The samples were labeled dry microwave pretreatment coal (DMP).

To prepare dry coal with thermal drying (DTD), the as-received coal 1000 g sample was split in a rectangular tray with a thickness of 60 mm and was dried in air-blower-driven drying closet for different times of 0, 2, 4, 6, 8 h, and finally sealed in a sample bag after cooling to room temperature.

In order to increase moisture content in the pore structure, the as-received sample was immersed in pure water for 24 h, and then filtered by gravity. The samples that contained more moisture in pore structure with moisture content of 9.86% was dried by hot air with temperature of 40° for 50 minutes to evaporate the surface moisture in a drying oven. Coal samples with a thickness of 60 mm were processed in the microwave field with different times and sealed into a sample bag after cooling to room temperature. Wet coal with microwave pretreatment was labeled WMP.

The energy transfer of pretreatment process of three kinds of coal samples, that is DMP, DTD and WMP, namely the design logic of the test, are shown in Fig. 2. Red stands for high temperature area.

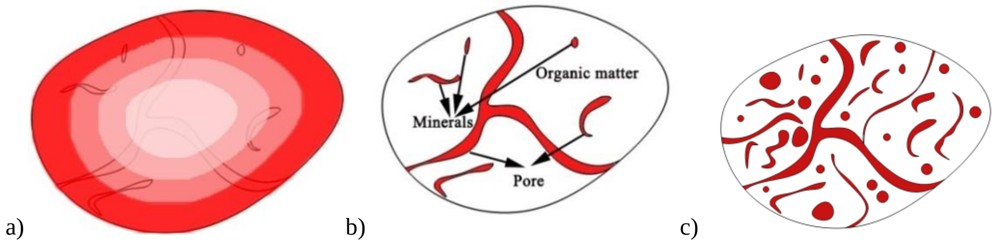

Fig. 2. Energy transfer process under different pretreatment methods: (a) DTD (b) DMP (c) WMP

For DTD, heat transfer is from outside to inside of coal particles, thus, the temperature gradient exists. In the preheating accelerating stage and dehydration process, the temperature of coal surface is always greater than the internal temperature. For DMP, because dielectric constants of moisture and minerals are greater than that of coal, microwave radiation can penetrate coal particles and act directly on the water molecules and other minerals. Therefore, microwave pretreatment reflects the characteristics of selective heating. During dehydration water molecules absorb the microwave energy and the sample is quickly heated. The internal complex pore structure hinders the vapor transmission, which causes the internal pressure of the coal particles to be higher than outside. Thus, the pressure gradient is formed. In theory, the pressure gradient can reduce the stress intensity of coal particles, thus it is helpful to the fine grinding of coal samples. Therefore, WMP was designed to increase the moisture content in the pore structure to verify this possibility.

Microwave pretreatment

A LG microwave oven that operates at 900 W and frequency of 2.45 GHz was used to preprocess the samples. Microwave pretreatment times were determined according to the time mentioned in literature as 0, 30, 60, 120, 180, 240 s. Other tests with longer irradiation periods were not carried out because of safety factor. The pretreatment tests were carried out with 1000 g coal samples. The samples were spread over on a rectangular container with a thickness of approximately 60 mm. The pretreated coal samples were put into hermetic bag after cooling to room temperature.

Moisture content analysis

To measure the total moisture content the coal samples of 50±0.01 g were placed into a preheated blast oven at 105° for 2 h. The weigh was checked every 30 minutes until changes were less than 0.01 g. The total moisture content was calculated by using formula, $M_{ar}=100m_1/m$ where M_{ar} is total moisture content (%), m_i is quantity reduction after measurement (g) and m is the initial quantity of coal samples (g).

Grindability tests

A rod mill, model XMB, was used for grinding. The mill consisted of two steel cylinders with 132 mm in diameter and 150 mm in length, with ten 18 mm in diameter and 144 mm in length steel rod inside each cylinder and steel rod was able to move freely up and down at the desired frequency. The working frequency was 277 rpm. Grinding tests were carried out with 200 g samples inside each cylinder. The power of mill was 370 Wh and the grinding time was 15 minutes.

The size composition of grinding products were analyzed by the standard screening procedure. The results showed that desired dissociation can be achieved when the coal was crushed to below 0.125 mm, therefore, the yield of the -0.125 mm size fraction was used as the evaluation index.

Liberation tests

The float/sink analysis was conducted to evaluate the liberation degree of the grinding products. For products of +0.5 mm, the density solution of 1.30, 1.40, 1.50, 1.60, 1.70, 1.80 $g\cdot cm^{-3}$ with different proportions of $ZnCl_2$. For products of -0.5 mm, the heavy liquid was the mixture of benzene, carbon tetrachloride and tribromomethane. The tests started with the lowest density and proceeded towards the highest density.

Results and discussion

Moisture content analysis

Figure 3 shows that the moisture content decreased with the increase of microwave pretreatment time for both DMP and WMP. It was due to the thermal effect of microwave radiation. The moisture content of DMP was reduced by 4.8 percentage point, and lost 61% of its water, while the moisture content of WMP was reduced by 5 percentage point, and lost 50.7% of its water after 240 s of pretreatment. When microwave pretreatment time was less than 30 s, the moisture content only slightly decreased because the energy was used to increase the temperature of coal particles. When the pretreatment time was 120 s, the moisture content of wet coal was close to the raw coal. For DTD, moisture content reduction showed a good linear relationship with thermal drying time, which was caused by heat-transfer mechanisms. Modelling moisture contents of different drying methods for DMP and WMP is shown in Table 1.

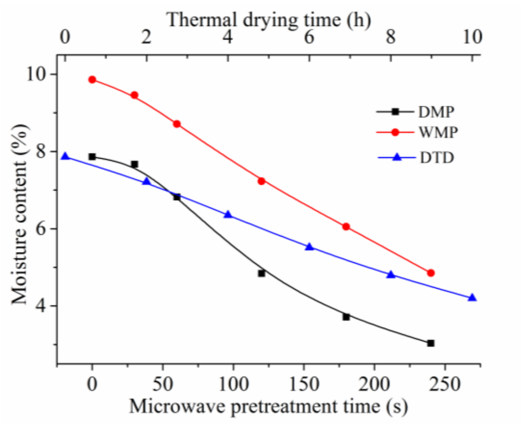

Fig.3. Moisture content of DMP, WMP and DTD

Table 1. Modelling moisture contents of different drying methods

Materials	Fitting models	Model parameters	Statistics	
			Reduced Chi-Sqr	Adj.R-Square
DTD	$y=a+bx$	$a=7.87$ $b=-0.377$		0.9959
DMP	$y=A_2+(A_1-A_2)/(1+(x/x_0)^p)$	$A_1=7.89$ $x_0=1.89$ $A_2=2.19$ $p=2.3$	0.0029	0.99933
WMP	$y=A_2+(A_1-A_2)/(1+(x/x_0)^p)$	$A_1=9.87$ $x_0=10.1$ $A_2=-11.51$ $p=1.27$	0.00978	0.99751

SEM analysis

Scanning Electron Microscope (SEM, FEI Quanta 250, America) was used to analyze the change in the surface topography for three coal types after treatment. Figures 4a, 4b and 4c show the scanning electron micrograph of DMP, WMP and DTD, after pretreatment 120 s, 240 s and 8 h, respectively. The top surface of 20 particles were analyzed for each set of testing.

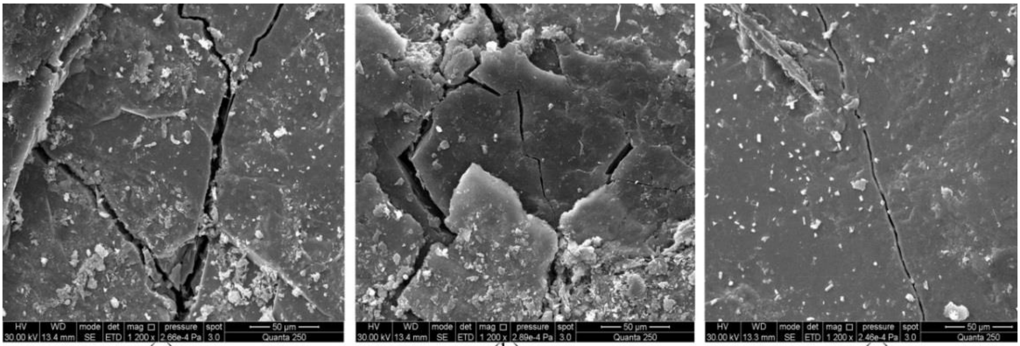

Fig. 4. Scanning electron micrographs of DMP (a), WMP (b) and DTD (c)

The scanning electron micrograph analysis shows that there was a possibility of cracks generation after microwave and thermal drying pretreatment. Massive cracks with relatively large size were observed on 17, 12 and 7 surface particles of DMP (Fig. 4a), WMP (Fig. 4b) and DTD (Fig. 4c), respectively. For comparison, the same tests were conducted for untreated coal, and the result shows that only two cracks were observed. It indicates that the selective heating effect of microwave and moisture content decrease are mainly responsible for crack generation, which is the important reason for improving grindability of coal.

The effect of mineral species on formation of cracks in the microwave field is shown in Fig. 5. As shown in Fig. 5, the mineral species has a significant impact on production of cracks. The white areas are minerals, including clay minerals and pyrite, and the black area is coal. The position of crack is mainly at the interface of coal and minerals. In theory, the temperature difference in this area is the greatest. In addition, the expansion coefficient of coal and mineral particles is different, therefore, this region has the greatest possibility of crack formation.

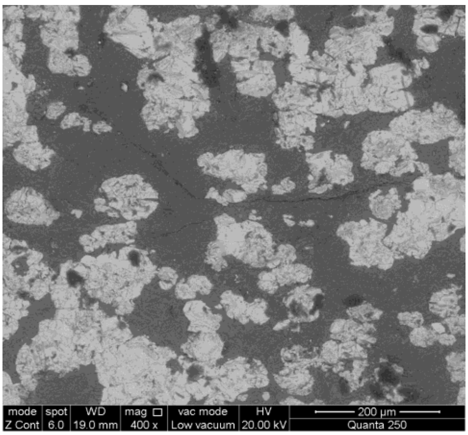

Fig. 5. Back scattering image of coal particles with microwave pretreatment

Size analysis of grinding products

The effect of microwave exposure time and thermal drying time on the yield of particle size -0.125 mm is shown in Fig. 6. As seen, the yield of -0.125 mm increase with the decrease of moisture content for all three types of coals, what indicates that reduction of water content is an important reason for improving grindability of coal samples. The relationship between moisture content and yield of -0.125 mm is shown in Table 2. The linear relationship between these two indicators is significant. Thus, the model relation between the pretreatment time and grinding fineness is obtained and is shown in Table 3.

As shown in Fig. 6, the differences between yields of -0.125 mm particle size of the three types of coal exist when the samples have the same moisture content. It is necessary to note that DTD, DMP and WMP after 8 h, 120 s and 240 s of pretreatment, respectively, have similar moisture contents. Yields of -0.125mm and 2-1 mm particle size of these three coal samples are shown in Fig. 7.

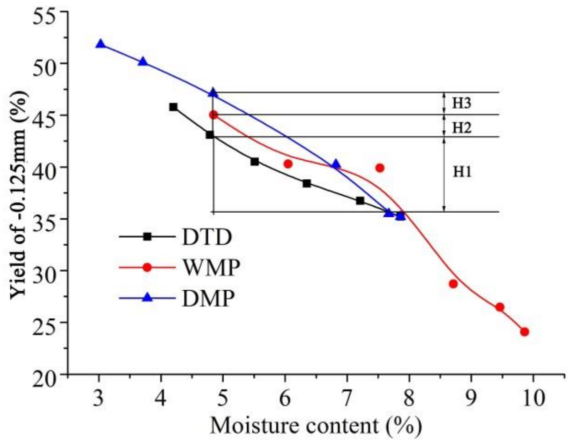

Fig. 6. Influence of moisture content on yield of -0.125 mm parctile size of DMP, WMP and DTD coal types

Table 2. Modelling the effect of moisture contents on yield of -0.125mm size fraction

Materials	Fitting models	Model parameters	Statistics Adj.R-Square
DTD	$z = a + by$	$a = 56.65$ $b = -2.79$	0.97
DMP	$z = a + by$	$a = 63.17$ $b = -3.51$	0.986
WMP	$z = a + by$	$a = 66.73$ $b = -4.22$	0.902

Table 3. Modelling the effect of pretreatment time on grinding fineness

Materials	Fitting models	
DTD	$z = 1.06x + 34.69$	x:h 0<x<10
DMP	$z = 55.45 - \dfrac{19.97}{1 + 0.23x^{2.31}}$	x:min 0<x<5
WMP	$z = 115.3 - \dfrac{90.22}{1 + 0.0537x^{1.27}}$	x:min 0<x<5

H1, H2, H3 marked in Fig. 6 represent the difference of strengthening grinding effect. As shown in Fig. 6, for DTD, H1 represents the effect of water reduction on fine grinding of coal samples. (H2+H3) represents the effect of microwave radiation that does not consider the effect of dewatering i.e., selective heating of microwaves. By using different dielectric constant characteristics of different minerals, the microwave pretreatment technology shows a significant advantage in the grinding efficiency compared with the traditional heating method. The microwave radiation penetrates the whole mineral particles to heat the certain material with a large dielectric constant, and the microwave heating method makes the energy utilization efficiency improved. Numerous cracks with larger size of DMP resulting from microwave radiation can significantly promote fine grinding of coal.

It should be noted that grinding fineness of DMP is greater than that of WMP, which shows that thermal evaporation of moisture in the pores of coal particles cannot significantly promote fine grinding of coal samples. Explanation of this phenomenon is mainly focused on the utilization efficiency of microwave energy. For coal with excess pore moisture, a large amount of microwave energy is used for heating and evaporation the moisture. Consequently, the microwave energy acting on minerals is reduced. The results show that the energy transfer is unreasonable.

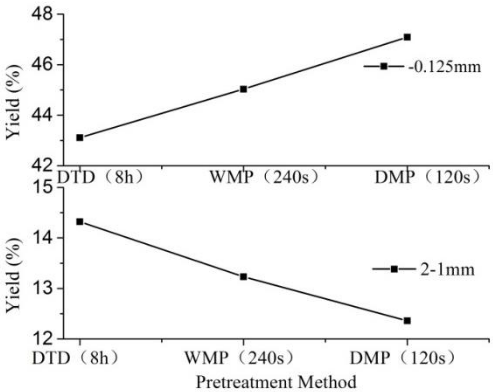

Fig. 7. Yield of -0.125 mm of DTD, WMP and DRY with same moisture content

Liberation of products

The float/sink analyses were performed to evaluate the liberation degree of grinding products of DTD (8 h), DMP (120 s) and WMP (240 s) with same moisture content that followed by 15 minutes grinding. Figure 8 shows that the density distribution of three products is significantly different. Yields of grinding products with density less than 1.4 g·cm^{-3} of DTD, WMP and DMP are 15.06, 16.71 and 19.93%, respectively, which suggest that more clean coal with the low ash content of DMP liberate from middlings.

Figure 9 shows the cumulative yields vs cumulative ash content. It is apparent that clean coal with 10% ash content of grinding products of DMP have the greatest production rate that is 37.27% greater than DTD, meanwhile increased by 9.45% compared with that of WMP. It is clearly that grinding products of DMP achieved the best dissociation. However, grinding products of DTD have the worest liberation degree. It can be explained that: (i) microwave has much more significant influence on fine grinding of coal sample than thermal drying, and (ii) the selective heating characteristic of microwaves can induce cracks at the interface between different minerals and promote dissociation of coal and minerals.

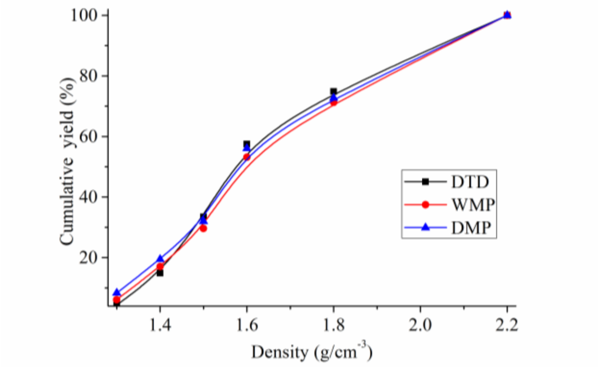

Fig. 8. Cumulative yield vs. density of grinding products

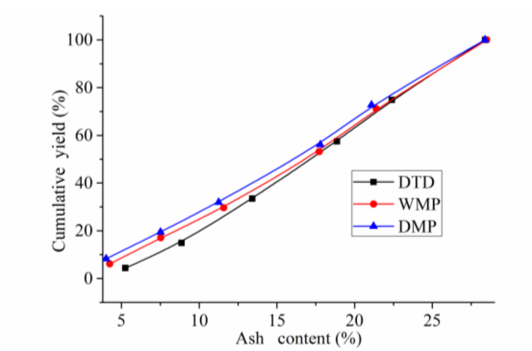

Fig. 9. Cumulative floats curves of grinding products

Conclusions

In this work, the mechanism of grindability improvement process of coal by microwave pretreatment was studied by a contrast test. Moisture reduction and selective heating characteristic of microwaves which can induce generation of cracks in coal particles were the main factors responsible for the improvement of grindability of coal in the microwave field. Thermal expansion of moisture within the pore of coal structure had no effect on grindability of coal.

The SEM analysis showed that DMP had more micron-sized cracks than WMP after microwave pretreatment, meanwhile, WMP had more cracks than DTD. The back scattering image of coal particles with microwave pretreatment showed that the crack position was mainly at the interface of coal and minerals. Grinding of fineness gradually increased with the decrease of the moisture content for all three types of coal. It proved that reduction in the moisture content played an important role in auxiliary grinding processing. In addition, the selective heating of microwave had a significant advantage over traditional heating methods. Thermal evaporation of moisture in the pores of coal particles cannot significantly promote fine grinding of coal samples.

Cracks induced by microwave selective heating can promote mineral dissociation, however, cracks caused by dehydration has no such effect. The yield of clean coal with 10% ash content of grinding products of DMP was 26.52% after 15 minutes grinding, which was 37.27% higher than that of DTD, meanwhile increased by 9.45% than that of WMP.

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