Evolution of Pore Structure during Pressurized Dewatering and Effects on Moisture Readsorption of Lignite

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ABSTRACT: In this study, pressurized method was used to dry lignite at moderate temperature to change its pore structure but preserve its oxygen-containing functional groups. The effects of drying conditions (time, pressure, and temperature) on equilibrium moisture content (EMC) and pore structure of dewatered coals were investigated, and the correlations between pore structure and EMC were also evaluated. The pore structure parameters of raw coal and dewatered coals were measured by nitrogen adsorption experiments. The EMC of dewatered coals was obtained by gravimetric method. The results indicated that the porous structure of dewatered coal was jointly affected by three factors (drying time, pressure, and temperature) in the initial pressurized drying stage. The drying process exhibited obvious effect in the initial stage of drying lignite. Destruction of pores under pressure was prevented due to the water present in these pores. To further improve the pore structures of dehydrated coals obtained by high-pressure treatment, the temperature was increased to above 140 °C under 3 MPa; thus, a large number of macropores were evolved into mesopores. Furthermore, the experiments on water reabsorption by dewatered coals indicated that the EMC (0.15–0.18) of dehydrated coal was the lowest when the pressure was 3 MPa, temperature was 140–160 °C, and the time required was 30 min. The moisture reabsorption contents of dehydrated coals were found to be positively correlated with its pore volume at high relative humidity. When the relative humidity was below 20%, they were related to specific surface areas or oxygen-containing functional groups. Therefore, pressure in the process of drying lignite was the main factor influencing the pore structure and the water reabsorption of dewatered coals, and the drying temperature was dominant under the pressurizing conditions.

1. INTRODUCTION

Lignite is a primary and important energy source that constitutes significant supplies for both energy and chemical feedstock because of its abundance, easy access, and low mining cost.¹ It has been estimated that nearly half of the world’s total coal reserves consists of lignite.² However, its high moisture content (25–65%) results in its low heat value and utilization efficiency and high transportation cost.³,⁴ Most lignite is exclusively used as fuel in power plants built near coal mines or as raw material in the chemical industry. Nonetheless, the use of lignite in practical applications has several disadvantages, including low energy efficiency, environmental pollution, and high costs.⁵

The high moisture content of lignite is mainly determined by its porous structure and the presence of abundant oxygen-containing functional groups in it structure.⁶,⁷ Reduction in the moisture content of lignite is extremely important before its industrial applications. Therefore, lignite needs to be dried for its effective utilization. Park et al.⁸ and Pusat et al.⁹ confirmed that the removal of moisture from coal particles increased with the increase in the drying gas temperature and gas velocity. Internal heat and mass were found to transfer better through smaller particles because of their larger surface area. Zhang et al.¹⁰ studied a new dewatering process, namely, vibration mechanical thermal expression (VMTE) process. The water content in lignite was found to decrease with increase in temperature, time, pressure, and vibration. The dewatering process of VMTE is better than the mechanical thermal expression (MTE) process. During the dewatering process of lignite, its physicochemical structure was affected obviously by different drying conditions. Based on previous studies,¹¹,¹² changes in pore structure of lignite include the collapse of pore structure caused by shrinkage stress; opening, closing, and cross-linking of pores; and thermal decomposition of organic macromolecular structures at high temperature.¹³ Moreover, according to literature reports, in the drying process, the pore structure shrank with the evaporation of moisture, and the shrinkage was found to be irreversible.¹⁴,¹⁵ Evans¹⁶ reported that pore structure collapsed a little at the beginning of drying; smaller pore structure was destroyed when more than 80% of water content was removed; and finally, pore structure did not change when more than 90% of water content

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was removed in the drying process of Yallourn lignite. Zhang et al.\textsuperscript{17} studied the changing rules of the course of lignite thermally upgraded and found that specific surface area and most probable pore size decreased with increasing drying temperature and time. Bergins et al.\textsuperscript{18,19} studied three different types of lignite by MTE and concluded that the volume of macropores decreased and that of mesopores increased with increasing temperature. Lester et al.\textsuperscript{20} reported that rapid diffusion of water vapor in lignite was the main reason for the collapse of the pore structure. For the water reabsorption behavior of lignite, Tahmasebi et al.\textsuperscript{21} reported that physical and chemical structure (pore structure and oxygen-containing functional groups) of lignite exhibited important influence on water reabsorption. Notably, the oxygen-containing functional groups (the carboxyl groups, in particular) of lignite play an important role in water adsorption, and the hydrogen bond formed between the oxygen-containing functional groups and water contributes significantly to the water adsorption process.\textsuperscript{17,22,23} Some researchers\textsuperscript{24,25} studied water readsorption mechanism of lignite and found that water was readsorbed as a monolayer on the surface of dewatered coal (DC) through hydrogen bond, followed by multilayer adsorption by readsorption on the monolayer. It has been concluded that the correlation between oxygen-containing functional groups of lignite and water readsorption behavior of lignite has been validated effectively; however, the correlation between pore structure and water readsorption behavior of lignite has not been reported in relevant literatures under the condition that oxygen-containing functional groups of lignite remain unchanged. Not only does the pore structure of lignite change at high temperature but also its oxygen-containing functional groups exhibit variation. Therefore, the pressurized method was used to dry lignite at moderate temperature to preserve the oxygen-containing functional groups in this study. The effects of drying conditions (time, pressure, and temperature) on equilibrium moisture content (EMC) and pore structure of dewatered coals were investigated, and the correlations between pore structure and EMC were also evaluated.

2. RESULTS AND DISCUSSION

2.1. Drying Behavior of Lignite under Different Operating Conditions. Figure 1 exhibits the effects of drying time and pressure on the drying efficiency (DE) of lignite at the temperature of 110 °C. Figure 1a demonstrates that the DE increases with increasing drying time. When drying pressure is 0.1 MPa at 110 °C, the DE with a drying time of 60 min is 4.5 times with that of 10 min. However, the DE increased gently when the drying time was more than 30 min under other pressures. Figure 1b exhibits that with the increase in drying pressure to 3 MPa, the DE sharply increases. When the drying times were 60, 45, 30, 20, and 10 min at 110 °C, the DE of dewatered coal obtained under 3 MPa was 1.06, 1.25, 1.44, 1.67, and 2.07 times with that under 0.1 MPa pressure, respectively. When drying pressure increases from 3 to 10 MPa, DE presents a significant downward trend, thus moisture could not easily escape from lignite when the coal was densified under higher pressure. The thickness of the samples prepared under different pressures (1, 3, 5, 7, and 10 MPa) was 9.22, 8.80, 8.42, 7.00, and 6.21 mm; and their specific volume was 407.7, 389.2, 372.3, 309.6, and 274.6 mm\textsuperscript{3} g\textsuperscript{−1}, respectively. It could be considered that the effect of drying pressure on the removal of water from lignite was obvious, and it varied under different conditions.
The DE of dewatering coal was the highest under 3 MPa, and the influences of temperature and time on DE could not be objectively investigated. So in Figure 2, the effect of temperature and time on DE was investigated at 5 MPa instead of 3 MPa. Effects of drying time and temperature on the DE of lignite at the pressure of 5 MPa are shown in Figure 2. Figure 2b shows that drying temperature has obvious influence on DE. When drying temperature exceeds 110 °C, rapid variations in DE are observed. The higher the temperature, the higher the DE of lignite. When temperature is above 100 °C, the state of water is rapidly transited from liquid to vapor. Thus, when drying temperature is below 110 °C, only the surface moisture of lignite is slowly removed; however, at above 110 °C, part of inherent moisture is also removed in vapor state. When the drying temperature is 50, 80, 110, 140, and 160 °C at 5 MPa, respectively, the DE of dewatered coal obtained at 60 min (drying time) is 1.76, 1.24, 1.14, 1.05, and 1.03 times with that at 10 min, respectively. When the drying time is 10, 20, 30, 45, and 60 min, respectively, the DE of dewatered coal at 160 °C (drying temperature) is 4.39, 3.29, 2.78, 2.66, and 2.58 times that at 50 °C, respectively. Compared to drying temperature, drying time had smaller effect on DE under pressurized condition, which was significantly different from that under room pressure (0.1 MPa in Figure 1a). The water in lignite generally undergoes two processes including evaporation of surface water and removal of internal water during drying. The experimental results showed that pressure affected the initial stage of lignite drying and temperature might eliminate the decrease in DE by high pressure, which should be closely related to the pore structure of lignite.

Figure 3 shows infrared spectrograms of raw coal (RC) and different coals obtained under different dewatering temperatures. The peaks corresponding to oxygen-containing functional groups are located at 3500–3000 and 1650–1600 cm⁻¹. The oxygen-containing functional groups of the samples (RC, DC110, DC140, and DC160) hardly exhibited any variation. When the drying temperature was below 160 °C, the oxygen-containing functional groups of the samples were preserved. Thus, it could be concluded that there was no release of volatiles in the process of pressurized dewatering. Therefore, volatile substances did not affect the DE of lignite during drying.

2.2. Evolution of Pore Structural Parameters of Lignite under Different Drying Conditions. 2.2.1. Effects of Drying Time on Pore Structure Parameter of Lignite. Table 1 lists pore structural parameters of raw lignite and dewatered coals obtained at different drying times. The pore structural parameters decreased gradually with increasing drying time, and they were significantly lower than those of raw lignite. Pore shrunk and swelled into a gel-like structure with the removal or absorption of water in coal. As mentioned above, water in coal was removed faster at the beginning of drying of lignite, which completely corresponds to the changes in pore structure. Compared to raw coal, the DCP3T110t10 dewatered coal obtained from raw lignite was jointly affected by three factors (drying time, pressure, and temperature). In contrast, dewatered coals dried for more than 10 min were affected by only one factor (drying time) compared to dewatered coals obtained at 10 min of drying. Isothermal adsorption/desorption curves of raw coal and dewatered coals under different drying times are shown in Figure 4. Clearly, little change is observed in p/p0 < 0.1, which indicates that operating conditions did not affect the micropores. The macropores were destroyed in lignite and adsorption/desorption quantities generally decreased according to p/p0 > 0.9. The types of pore structures were cylindrical bore or flaky and did not lead to any change in lignite-based hysteresis of the isothermal adsorption/desorption curve.

2.2.2. Effects of Drying Pressure on Pore Structural Parameters of Lignite. Table 2 summarizes pore structural parameters of raw lignite and dewatered coal under different drying pressures. The pore structural parameters of dewatered coals first decreased and then increased with increasing pressure and were also significantly lower than those of raw lignite. This was not the same variation trend as that of DE vs pressure. This phenomenon illustrated that the destruction of the pore structure was affected by two factors including pressurization and removal of water. However, the transition points of two factor actions were different, the former was 5 MPa and the latter was 3 MPa. Thus, it could be considered that destruction of the pore structure not only exhibited a relationship with removal of

![Figure 3. Infrared spectograms of raw coal and different dewatered coals obtained under different dewatering temperatures.](image)

![Figure 4. Isothermal adsorption/desorption curves of the raw coal and dewatered coals obtained under different drying times.](image)

| sample          | specific surface area (m² g⁻¹ db) | pore volume (cm³ g⁻¹ db) | average pore diameter (nm) |
|-----------------|----------------------------------|-------------------------|---------------------------|
| RC              | 7.02                             | 0.028                   | 15.89                     |
| DCP3T110t10     | 5.32                             | 0.014                   | 10.43                     |
| DCP3T110t20     | 4.65                             | 0.012                   | 10.34                     |
| DCP3T110t30     | 4.43                             | 0.012                   | 10.47                     |
| DCP3T110t45     | 4.15                             | 0.011                   | 11.04                     |
destroyed in lignite when pore diameter. Figure 5 shows the isothermal adsorption/desorption curves of raw coal and dewatered coal. Although pore structures were destroyed. Moreover, water in macropores was removed and then compressed and even broken; however, this was not observed in case of smaller pores. Therefore, this process mainly influenced the specific surface area, pore volume, and average pore diameter. Figure 5 shows the isothermal adsorption/desorption curves of raw lignite and dewatered coals under different drying pressures.

| sample   | specific surface area (m² g⁻¹ db) | pore volume (cm³ g⁻¹ db) | average pore diameter (nm) |
|----------|-----------------------------------|-------------------------|---------------------------|
| RC       | 7.02                              | 0.028                   | 15.89                     |
| DCP,T₁₅₀T₁₀  | 5.89                           | 0.018                   | 12.14                     |
| DCP,T₁₃₀T₁₀  | 5.32                           | 0.014                   | 10.43                     |
| DCP,T₁₁₀T₁₀  | 5.25                           | 0.012                   | 9.52                      |
| DCP,T₁₀₀T₁₀  | 5.59                           | 0.015                   | 11.18                     |
| DCP,T₉₀T₁₀   | 5.40                           | 0.013                   | 10.42                     |

Figure 5. Isothermal adsorption/desorption curves of raw lignite and dewatered coals under different drying pressures.

Water in pores prevented them from being destroyed. The isothermal adsorption/desorption curves of raw coal and dewatered coals under different drying temperatures are shown in Figure 6. Clearly, adsorption quantity gradually reduces for $p/p₀ < 0.1$ when temperature is above 110 °C, which indicates that operating conditions affect the micropores and decrease their volume. Moreover, macropores were also destroyed in lignite with $p/p₀ > 0.9$. At the same time, the types of pore structures were cylindrical bore or flaky and did not exhibit any change in lignite-based hysteresis of isothermal adsorption/desorption curve.

2.3. Effects of Operating Conditions on Equilibrium Moisture Content of Dewatered Coal. The equilibrium moisture content (EMC) of dewatered coal at 30 °C and under relative humidity of 95% was measured, and the results are presented in Figure 7. Both drying time and pressure reduce the EMC of dewatered coal; however, EMC changes little when drying time is above 30 min or drying pressure is above 3 MPa. This phenomenon indicates that excessive increase in drying time and pressure is not a good choice. Comparative analysis of the two factors reveals that the effect of drying pressure is more powerful on properties of lignite.

Figure 7 shows the effects of drying time and temperature on EMC of the dewatered coal at 30 °C and under relative humidity of 95%. Figure 8a demonstrates that drying time has little effect on EMC. On the other hand, the EMC of dewatered coal decreases obviously with increasing drying temperature. Comparison of these three factors indicates that the pressure in the process of drying lignite is the main factor influencing the water reabsorption of dewatered coal, and drying temperature is dominant under the pressurizing condition.

| sample   | specific surface area (m² g⁻¹ db) | pore volume (cm³ g⁻¹ db) | average pore diameter (nm) |
|----------|-----------------------------------|-------------------------|---------------------------|
| RC       | 7.02                              | 0.028                   | 15.89                     |
| DCP,T₁₀₀T₅₀  | 6.56                           | 0.026                   | 14.36                     |
| DCP,T₁₀₀T₃₀  | 5.95                           | 0.015                   | 8.59                      |
| DCP,T₁₀₀T₁₀  | 5.27                           | 0.015                   | 10.01                     |
| DCP,T₁₀₀T₅₀  | 3.69                           | 0.012                   | 11.39                     |
| DCP,T₁₀₀T₁₀  | 3.53                           | 0.013                   | 13.01                     |

Figure 6. Isothermal adsorption/desorption curves of the raw lignite and dewatered coals at different drying temperatures.
Figure 9 shows EMC results of raw coal and dewatered coals obtained under different drying conditions (time, pressure, and temperature), at 30 °C, and under relative humidity of 95%. The EMC values of dewatered coals first decreased and then changed slightly with increasing drying time and pressure.

Water in lignite was removed and pore structure was destroyed when pressure impacted raw lignite in the drying process. Open pores were transformed into close pores when drying pressure was increased excessively, which restrained pore volume change and water readsorption of dewatered coal. For the EMC of dewatered coal obtained under different temperatures, it does not change obviously with the increase of drying temperature from 50 to 80 °C. The EMC of dewatered coal obtained under different pressure conditions was almost the same. The EMC of dewatered coal obtained above 110 °C reduced faster. The phenomenon should be related to water types and porous structure of the dewatered coal. There are three forms of water, namely, free water, water bound in pores (capillary water), and water bound to surface functional groups (bound water: multilayer water and monolayer water). The first stage involves the removal of free water. The second stage is the desorption of capillary water. The third and fourth stages of dewatering correspond to the loss of molecular (multilayer and monolayer) water.26 Free water was mainly removed at lower drying temperature, whereas multilayer water even monolayer water could be removed at higher drying temperatures. Consequently, lower drying temperature and pressure are beneficial to the water readsorption of dewatered coal.

In other words, the porous structure of dewatered coal obtained under different drying conditions is closely related to its water readsorption behavior. The correlations between the pore volume of dewatered coal and different drying conditions are shown in Figure 10. The pore volume of lignite decreases with increase of drying time, temperature, and pressure. The effect of drying temperature on pore volume of lignite is the most significant; the pore volume of dehydrated coal prepared at 160 °C is half that of the sample at 50 °C. The pore volume of dehydrated coal prepared under 10 MPa is only 0.28 times that of the sample obtained under 1 MPa. Drying time has little effect on pore volume. The pore volume of dewatered coal prepared under 5 MPa and 50 °C is the largest among all samples. With the increase of drying temperature, the pore volume of lignite decreases sharply. Combination of the results of Figures 7 and 8 indicate that pore volume is one of the important physical parameters for the water readsorption capacity of dewatered coal.
coal. The more pore volume of dewatered coal decreases, the less water is reabsorbed on it. The dewatered lignite undergoes water readsorption on multimolecular layers by hydrogen bond, which is also related to its large pore volume. Larger pore volume leads to more moisture content and formation of more hydrogen bonds, which result in more content of reabsorbed water of dewatered coal. The EMC (0.15) of dewatered coal DCPTT160T160 obtained under optimum drying conditions is the lowest. The pore volume of raw lignite is largest among all samples; therefore, the EMC of raw lignite (0.34) is the highest. The EMC of raw lignite is more than twice the EMC of dewatered coal (60 min, 5 MPa, 160 °C).

2.4. Relationships between Pore Structural Parameters and Moisture Readsorption Content of Dewatered Coal. Figure 11 shows the relationship between total pore volume ($V_{total}$), mesopore volume ($V_{meso}$), and specific surface area and EMC or moisture readsorption content of dewatered coals. Figure 11a exhibits a linear relation between EMC of dewatered coals and total pore volume at 30 °C and under relative humidity of 97%. Figure 11b demonstrates a linear relation between moisture readsorption content of dewatered coal and mesopore volume at 30 °C and under different relative humidity (RH = 21–75%). The adsorption of water molecule on micropore is initially monolayer adsorption, and thus the water readsorption capacity of dewatered coal at the initial stage should be related to specific surface area. Figure 11c shows a linear relation between moisture readsorption content of dewatered coal and specific surface area at 30 °C and under relative humidity of 11.3%, which exhibits that the occurrence of the monolayer water readsorption at a relative humidity of 11% is mainly affected by specific surface area.
To verify the validity of the rules mentioned above, raw coal was placed under high temperature to remove the oxygen-containing functional groups, and then this sample pretreated at high temperature was used as typical example for detecting the effects of pore structural changes. Figure 12 shows infrared spectrograms of different coals obtained under different temperatures. The peaks corresponding to oxygen-containing functional groups affecting water readsorption are located at 3500–3000 and 1650–1600 cm⁻¹. The spectrograms show the presence of few oxygen-containing functional groups when temperature was above 600 °C. The peak corresponding to infrared absorption of ash is located in the range of 3730–3600 cm⁻¹. The oxygen-containing functional groups of demineralized coal are less than that of raw coal. Thus, 600 and 700 °C were selected as proximate temperatures to conduct coal treatment. Previous researchers have reported that ash content shows a relationship with water readsorption.

Table 4 presents that kind of lignite has a higher ash content (A₉ = 27.78%). Table 5 presents that kind of lignite has a higher ash content (A₉ = 27.78%). Therefore, raw coal was subjected to ash removal treatment using hydrochloric acid and hydrofluoric acid, and then it was calcined at 700 °C. As a result, the treated raw coal showed very little ash content (A₉ = 1.34%), and the infrared peak of ash (3730–3600 cm⁻¹) disappeared.

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Table 4. Pore Structural Parameters of Different Dried Samples Obtained under High Temperature

| Samples          | Specific surface area (m² g⁻¹ db) | Pore volume (cm³ g⁻¹ db) | Average pore diameter (nm) |
|------------------|-----------------------------------|--------------------------|---------------------------|
| DCP₀₁T₆₀₀₅₀      | 61.71                             | 0.066                    | 4.011                     |
| DCP₀₁T₇₀₀₅₀      | 134.86                            | 0.102                    | 2.927                     |
| DCP₀₁T₇₀₀₅₀A₆₅₀  | 258.45                            | 0.155                    | 2.397                     |

Table 4. Pore Structural Parameters of Different Dried Samples Obtained under High Temperature

area and total pore volume of the three samples is as follows: DCP₀₁T₇₀₀₅₀A₆₅₀ > DCP₀₁T₆₀₀₅₀ > DCP₀₁T₆₀₀₅₀. Figure 13 shows relationship between pore structural parameters and moisture readsorption content of dewatered coals. Water readsorption capacity in coal was positively correlated with total pore volume and specific surface area when the coal was placed under a higher-relative-humidity (p/p₀ > 0.38) environment. The order of EMC of the three samples is as follows: DCP₀₁T₇₀₀₅₀A₆₅₀ > DCP₀₁T₆₀₀₅₀ > DCP₀₁T₆₀₀₅₀. Although DCP₀₁T₆₀₀₅₀ contains the most oxygen-containing functional groups among these samples, its water readsorption capacity is independent at the relative humidity of p/p₀ > 0.38. In the lower relative humidity environment of p/p₀ < 0.38, the moisture readsorption content order of the three samples is DCP₀₁T₆₀₀₅₀ > DCP₀₁T₇₀₀₅₀ > DCP₀₁T₇₀₀₅₀A₆₅₀, which shows that EMC is positively correlated with the quantities of oxygen-containing functional groups in the samples and independent of their pore structures. When the drying temperature was below 160 °C, it did not affect the total oxygen-containing functional groups in coals. Therefore, water readsorption content in coal was found to be related to specific surface area when coal was placed in a lower-relative-humidity environment. Above all, water readsorption content in coal is related to the total pore volume when coal is placed in a higher-relative-humidity environment; however, in a lower-relative-humidity environment, water readsorption content in coal is related to specific surface area. Water readsorption content in coal is not related to ash content in the samples.

3. CONCLUSIONS

A new drying method (involving preservation of oxygen-containing functional groups), namely, pressurized dewatering, was used for lignite drying for the first time in this study. The pressure influenced the initial stage of drying lignite and the effect on the removal of internal water from lignite was obvious. The increase of drying temperature might eliminate the decrease in drying efficiency (DE) under high pressure. The porous structures of dewatered coals were jointly affected by three factors (drying time, pressure, and temperature) for less than 10 min of drying under pressurized drying, and dewatered coals dried for more than 10 min were only affected by one factor (drying time). Water in pores could prevent the pores of lignite from being destroyed under pressure; as a result, porous structural parameters of dehydrated coals obtained under higher pressure were large and their equilibrium moisture contents (EMCs: 0.23–0.30) were relatively high. However, the internal water of lignite could be effectively removed when temperature was elevated to above 140 °C, and therefore it had significant effect on pore structure and could decrease the EMC of dehydrated coals under higher pressure. Consequently, the experimental results indicated that the EMC (0.15–0.18) of dehydrated coal was the lowest when the pressure was 3 MPa, the temperature was 140–160 °C, and the time was 30 min. By correlating the moisture readsorption contents of dehydrated coals with their pore structures at different relative humidity, the results showed that the moisture readsorption contents of dehydrated coals were positively correlated with their pore volume...
at high humidity, and they were related to specific surface areas or oxygen-containing functional groups in a lower-relative-humidity environment. Over all, pressure during the drying process of lignite was the main factor affecting the pore structure and the water reabsorption of dewatered coal, and temperature played a dominant role under the pressurizing conditions.

4. EXPERIMENTAL SECTION

4.1. Coal Sample and Its Preparation. Lignite from Inner Mongolia in China was selected for this study. The raw coal (RC) was ground and sieved to the particle sizes of $0.150 - 0.178$ mm under N$_2$ atmosphere prior to its use. If the sample size is larger than 0.178 mm, the coal sample will be very hard pressurized to be a 13 mm diameter wafer, whereas if the sample size is smaller than 0.15 mm, the moisture content would decrease obviously during grinding. So, the sample was kept in a tightly closed container. The proximate and ultimate analysis results of the coal sample are listed in Table 5, wherein the full moisture content ($M_t$) was measured at 105 °C in a nitrogen oven according to the Chinese national standard GB/TB 212-2008.

4.2. Dewatering of Lignite. Figure 14 shows the schematic illustration of a hydraulic press equipment used for drying raw coal under N$_2$ atmosphere. During drying, the pressure and temperature controlled using the pressure and temperature controllers varied from 0.1 to 10 MPa and 50 to 160 °C, respectively, to prevent the change of oxygen-containing functional groups. The samples obtained under different drying conditions are represented as DCP$_{x}T_{y}t_{z}$. For example, DCP$_{1}T_{50}t_{20}$ indicates a dehydrated coal obtained under the drying conditions of 1 MPa, 50 °C, and 20 min. The drying efficiency (DE) was calculated by using eq 1 as follows

$$DE = \frac{m_{0} - m_{f}}{m_{0} \times M_{t}}$$

(1)

where $m_{0}$ is the initial weight of the coal sample (g) and $m_{f}$ is the weight of the coal sample after drying (g).

4.3. Measurement of Equilibrium Moisture Content. The moisture reabsorption behaviors of raw coal (RC) and dewatered coal (DC) samples were measured using a constant temperature and humidity equipment at 30 °C under different relative humidity ranging from 6.4 to 97%. During the measurement, the samples were weighed in glass dishes and then placed into the equipment for 72 h to reach the moisture adsorption equilibrium. The equilibrium moisture content (EMC, g g$^{-1}$ db) was calculated by using eq 2 as follows

$$EMC = \frac{m_{a} - m_{d}}{m_{d}}$$

(2)

where $m_{a}$ is the weight of the coal sample after moisture reabsorption (g) and $m_{d}$ is the weight of the coal sample after drying according to the Chinese national standard GB/TB 212-2008 (g).

Equilibrium adsorption time under relative humidity of 75% is shown in Figure 15, which exhibits that three dewatered coal samples obtained under ordinary pressure and different drying temperatures can reach water reabsorption equilibrium after 10 h. As a result, 72 h were enough for different samples to reach water reabsorption equilibrium.

4.4. Analysis of Pore Structure. The pore structural parameters of raw coal and dewatered coals were measured using a N$_2$ adsorption analyzer (JW-BK122W, China) at $-196$ °C. Before the experiments, the coal samples were degassed at a relatively low temperature of 50 °C for 24 h under vacuum to avoid obvious changes of pore structure caused by the pretreatment process. The pore size distribution and specific surface area of the samples were calculated, respectively, by using the Barrett–Joyner–Halenda and the Brunauer–Emmett–Teller equations$^{31,32}$ according to adsorption isotherm.

Table 5. Proximate and Ultimate Analyses of Raw Coal$^{a}$

| proximate analysis/wt % | ultimate analysis/wt %, daf |
|------------------------|----------------------------|
| $M_t$ | $A_d$ | $V_{ad}$ | C | H | O$^b$ | N | S |
| 26.5 | 27.78 | 44.32 | 70.69 | 2.42 | 24.31 | 0.96 | 1.62 |

$^a$A$_d$ is air-dried basis; daf is dry and ash-free basis. $^b$By difference.

Figure 14. Schematic illustration of the drying equipment.

Figure 15. Determination of moisture reabsorption equilibrium time of dewatered coal.

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