Relationship between Thermal Conductivity and Chemical Structures of Chinese Coals

Qingmin Shi,* Yong Qin,* and Yilin Chen

ABSTRACT: Three different ranks of Chinese coals were investigated on the thermal conductivity and corresponding molecular structure by thermal analyzer, $^{13}$C NMR, and HRTEM techniques. The thermal conductivity of coals measured in room temperature first shows a decrease, then a slight increase, and finally a sharp increase with increasing coalification. Ranging from 30 to 150 °C, increasing the temperature slightly improves the thermal conductivity of coals with varying degrees. Water with a higher thermal conductivity than air contributes to the thermal conductivity of porous coal samples. The value of thermal conductivity is higher along coal bedding planes than when perpendicular to beddings, which indicates the anisotropy of coal thermal conductivity. The anisotropy degree increases with the rank of coals and is affected by clay minerals when coals adsorb water. Molecular structure analysis shows that polycondensed aromatic ring related to lattice vibration contributes to the increase of thermal conductivity. The aliphatic bridges among aromatic clusters ensure the continuity of atom vibrations and contribute to energy transport, but the free-ended side chains have the opposite effect. The relative ordered distributions of lattice fringes of anthracite, which were higher than those of bituminous coal, enhance the anisotropy of thermal conductivity.

1. INTRODUCTION

Coal with low thermal conductivity affects heat transfer in the interior of the Earth and cause underlying rocks to reach higher thermal maturities in a basin with igneous activity. Coals act as thermal insulators in an igneous intrusion region and enhance the geothermal gradients of underlying units. Previous researches show that thermal conductivity decreases with increasing coal rank from lignite to bituminous coals, but it increases for anthracite. The moisture and minerals contribute to the thermal conductivity of a coal except for anthracite. These investigations of thermal conductivity for coals were mostly reported around in the year 2000, whereas further studies are demanded currently due to the new thermal applications in coalbed, such as thermally enhanced gas recovery, coalfield fire estimation, coal mine geothermal utilization, and underground coal gasification.

For coals, the bedding planes are visible and delimited by obvious lithotypes. As references, the investigations on thermal conductivity of laminated metamorphic rocks and clay stones are focused on their anisotropy that affects the thermal regimes of a sedimentary basin. Heat preferentially transports in a direction parallel to the bedding planes of rocks. In the process of diagenetic compaction, the long axis of individual clay particles becomes oriented parallel to bedding planes, resulting in a horizontal matrix conductivity increase and a vertical matrix conductivity decrease. By contrast, sandstones and granite are relatively isotropic and easily influenced by moisture, porosity, and pressure in thermal conductivity.

Their thermal conductivities increase with increasing pressure and moisture content and with decreasing porosity.

Coal has a complex organic macromolecular structure in the microscopic level and a layer structure in the macroscopic level. However, few researchers focus on the chemical structure control and bedding effect of coal on thermal conductivity. In this paper, the variation of thermal conductivity with rank and their anisotropy were investigated. Also, the control mechanism at a molecular level was discussed.

2. EXPERIMENTS

Three tested coals including high-volatile bituminous coal, semi-anthracite, and anthracite were collected from the Shanxi Formation (Early Permian), in Inner Mongolia, and Shanxi Province (the latter two), China. These samples are all bright coals with primary structure. The petrography and proximate analysis data are listed in Table 1.
The test samples were cut into plates with a parameter of 40 mm × 35 mm × 8 mm, and a maximum-size surface perpendicular to coal beddings was polished. The thermal conductivity of coals was measured by a thermal analyzer (CT3200, XIATECH, China) based on the thermal line method. Each kind of coal needs two plates to nip a thermal line/sensor with the polished surface, as shown in Figure 1. The thermal line/sensor is both the thermal source and thermal sensor, which can load a constant heat and improve the temperature of coals. The thermal conductivity of coal is calculated through monitoring the process of the temperature increase by a thermal sensor. The control equation is:

$$\frac{\partial T}{\partial t} = aV^2 T$$

Where $T$ is the temperature, $K$; $t$ is thermal diffusion coefficient of a medium, m$^2$/s; $a = \lambda/\rho C_p$, $\lambda$ is the thermal conductivity of a medium, W/(m·K); $\rho$ and $C_p$ are the medium density and specific heat at constant pressure, respectively.

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Figure 1b shows a linear relationship between $\Delta T$ and $t$ for a sample, which confirms a correct and valid measurement. Heat transports from the thermal line to all around and forms a radiation cylinder (Figure 1a). If the thermal line is perpendicular to coal beddings as displayed in Figure 1a, heat will mainly flow parallel to coal beddings and the corresponding thermal conductivity ($\lambda_{par}$) will be calculated. On the contrary, most heat will transport perpendicular to coal beddings and their thermal conductivity is labeled $\lambda_{perp}$. The anisotropy ratio of thermal conductivity, $A$, is defined as the ratio of $\lambda_{par}$ to $\lambda_{perp}$ namely $A = \lambda_{par}/\lambda_{perp}$.

Coal samples were tested both on dry and water-adsorbed conditions. During measurements, dried coals were first probed at 30, 50, 100, and 150 °C. Then, these coals were put in an isothermal bath with the saturated solution of Na$_2$SO$_4$ for 7 days. The water-adsorbed coals were only measured at 30 °C to avoid the effect of moisture loss over time.

The mineral matters were determined by low temperature ashing and D8 ADVANCE X-ray diffraction made by Bruker. An AVANCE III HD $^{13}$C nuclear magnetic resonance (NMR) made by Bruker, Germany, were used to analyze the molecular structures based on the spectra. Also, a JEM-2100F high-resolution transmission electron microscope (HRTEM) (Bruker, Germany) was used to observe the coal aromatic fringes.

3. RESULTS

3.1. Variation with Coal Rank. Figure 2 shows the relationship between thermal conductivities and coal rank where most of the data plotted are collected from the literature reported by Herrin and Deming, and others are those measured at 30 °C in this work (Table 2). The tendency analyzed here is different from the above literature. It is not neglected that a slow increase stage approximately ranging from 1.0 to 2.3% ($VR_{\text{total}}$) exists in the evolution of thermal conductivity with increasing rank.

Thermal conductivities against both carbon content and $VR_{\text{total}}$ are plotted in Figure 2, which could cover the shortages of each other. The carbon content with a large range before
medium volatile bituminous coals could clearly display the former trend with increasing rank, and the VR \(_{\text{max}}\) could show the latter stage clearer. This tendency is divided into three stages with increasing coal rank. In the first stage, the thermal conductivity decreases due to moisture reducing over the rank until around the boundary between high-volatile bituminous coal and medium-volatile bituminous coal.\(^1\) For the second stage, the subsequent increase is slightly in the range from medium volatile bituminous coal to semi-anthracite. The beginning of a sharp increase of thermal conductivities appears at anthracite, which indicates the third stage of evolution of the thermal conductivity.

Interesting results show that coals in the second stage of thermal conductivity experience the second coalfication jump. In this stage, the oxygen markedly reduces in the process of coalfication accompanied by an especially strong aromatization and ring condensation of the coal molecule.\(^2\) The optical anisotropy of vitrinite appears and rises due to the progressive adjustment of the predominantly aromatic molecule in the bedding plane on the condition of increasing load pressure.

The samples of high-volatile bituminous coal and semi-anthracite in this work distribute at the boundary of these three stages. Their thermal conductivities have a slight difference, which describes the characteristics of the second stage in this study, while the anthracite with a higher value of thermal conductivity belongs to the third stage.

### 3.2. Specific Tendency

Thermal conductivities both parallel and perpendicular to coal beddings were measured and plotted in Figure 3 with the same coordinate system. The samples with adsorbed water are also displayed here in gray background, which distinguish from those dried coals measured at different temperatures. All coals have higher thermal conductivities along bedding planes than when perpendicular to beddings. No matter whether thermal conductivities are parallel to beddings or perpendicular to beddings, the value of anthracite is much higher than both the high-volatile bituminous coal and semi-anthracite on each temperature condition. Also, the value of the semi-anthracite is slightly higher than high-volatile bituminous coal. This trend agrees with the general tendency as summarized in section 3.1.

Coal samples with adsorbed water have higher thermal conductivities than the dry samples at 30 °C, which consists with research conducted by Herrin and Deming.\(^4\) Water with 0.6 W/m·K of thermal conductivity at room temperature is an important factor to affect heat flow in porous rocks.\(^22,24\) It could replace some air that is 0.03 W/m·K at room temperature in pores and improve the thermal conductivity of samples. Therefore, the thermal conductivity of each coal with adsorbed water is improved, while their rank is not changed.

With increasing temperature, coal thermal conductivity slightly improves entirely (Figure 3). In Figure 3b, the thermal conductivities of high-volatile bituminous coal and semi-anthracite are much closer than those parallel to beddings (Figure 3a). The increment of thermal conductivity with temperature appears nonlinear, which reflects different sensitivities of coals to each temperature. An equation is created to evaluate the sensitivity (\(k\)) as follows,

\[
k_i = (\lambda_i - \lambda_0) / [\lambda_0(T_i - T_0)]
\]

where \(k_i\) is the sensitivity coefficient of thermal conductivity at \(T_i\) temperature. \(\lambda_i\) is the thermal conductivity at \(T_i\) temperature. \(\lambda_0\) is the thermal conductivity at \(T_0\) (303.15 K) temperature. The results are listed in Table 3.

The results show that the thermal conductivities of coals perpendicular to beddings are generally more sensitive to temperature than those parallel to beddings (Table 3), especially that of the anthracite. A temperature of 50 °C is the most sensitive for thermal conductivities perpendicular to beddings. Also, the sensitivity shows a decline in a higher temperature. In other words, although temperature can improve the thermal conductivity perpendicular to coal beddings, it can also disturb heat to flow orderly in coals. Compared with the sensitive temperatures of coals parallel to beddings, the high-volatile bituminous coal, semi-anthracite coal, and anthracite have different values, which are 50, 100 and 150 °C, respectively. This is an interesting finding that needs further study in the future. It displays that the sensitivity of anthracite is the highest when heat flow perpendicular to beddings, but lowest when heat flows parallel to beddings. This character also verifies the existence of anisotropy of coal thermal conductivity.

### 3.3. Anisotropy of Thermal Conductivity

In order to investigate the anisotropy of thermal conductivity, an anisotropy ratio (\(A\)) is defined as \(A = \lambda_{\text{perp}} / \lambda_{\text{par}}\). Figure 4 illustrates the anisotropy ratio of coals on different conditions. Among these dried coals, the anisotropy degree of anthracite is the highest following the semi-anthracite. Also, the high-volatile bituminous coal has the lowest anisotropy degree of thermal conductivity. The degrees of anisotropy in orthogonal directions are different and nonlinear with increasing temperature due to the different sensitive temperatures in orthogonal
directions. The temperatures corresponding to the strongest sensitivity of thermal conductivity parallel to beddings are the same with that corresponding to the highest anisotropic degree except anthracite. In fact, the anisotropic degree of the anthracite is also depressed at 150 °C.

Comparing water-adsorbed coals with dried coals (Figure 4), water weaken the anisotropy of thermal conductivity of coals except semi-anthracite due to the different minerals in organic matrix. The phyllosilicate clay is the major composition among inorganic minerals. The content of each kind of clay minerals are listed in Table 4, which shows that the high-volatile bituminous coal and anthracite have similar kinds and content of clay minerals including higher content of illite−smectite mixed layers that will increase greatly in volume when absorbing water. The swelling not only weakens the anisotropy but also drive air out of coals. However, kaolinite has a low shrink−swell capacity and water absorbing capacity but has a high anisotropy of thermal conductivity due to the orientation of platelets that is parallel to beddings. Absorbed water filled in the voids between kaolinite minerals and organic matrix, which improves the capacity of heat transfers through kaolinite from organic matrix (Dao et al.; Bourret et al.18). Therefore, the anisotropy of thermal conductivity of the entire coal is increased when it absorbs water.

4. DISCUSSIONS

Herrin and Deming reported that the thermal conductivity of coals is affected by macro factors such as coal rank, density, and moisture. Essentially, it is controlled by the molecular structure of coals at the microscale for different ranks of coals. Atom vibrations that are called phonons contribute to thermal energy storage and heat transfer in the molecule. Hence, the relationships between the thermal conductivity and chemical-structural properties of coals determined by solid state 13C NMR (solid state 13C nuclear magnetic resonance spectroscopy) and HRTEM (high resolution transmission electron microscopy) techniques are discussed here. These chemical structure parameters of coals have been widely investigated for charactering the macromolecular structure of coals. Partial data from the previous research were collected and combined with the data of thermal conductivity to find a statistical tendency.

4.1. Chemical Structure Control. Figure 5 shows the 13C CP-MAS interrupted 1H decoupling (DD-MAS) NMR spectra of a coal, which is assigned to each functional group according to Solum et al.’s and Okolo et al.’s reports. An aromatic
region $f_b$ (90–240 ppm) and an aliphatic region $f_d$ (0–90 ppm) can be distinguished clearly over chemical shift ranges. A total of 12 parameters relating to the carbon skeletal structure are subdivided from the regions such as the main aromatic ring region $f_a$, bridge head carbon $f_B$, aromatic ring carbon with an attached oxygen $f_P$, aromatic ring carbons with an attached alkyl group $f_S$, methyl groups $f^*_a$, methylene plus methine groups $f^*_H$, aliphatic attachments except side chains, namely aliphatic bridges (A.B.), and side chain. A.B. indicates the proportion of all possible aliphatic bridges, in aliphatic groups.

According to these parameters, lattice parameters of coals are derived by Solum et al. who assumed coal macromolecular fragments consist of an aromatic cluster with side chains and bridging groups that link the condensed aromatic clusters. For a given composition, lattice vibrations make more contributions to heat flow, which result in the thermal conductivity in a crystal being higher than in an amorphous solid. Therefore, we pay more attention on the lattice parameters of crystallitic coals including the mole fraction of aromatic bridgehead carbons $\chi_b$, the number of side chains per cluster S.C., and the proportion of all possible aliphatic bridges in aliphatic carbons A.B. An increase of $\chi_b$ indicates the growth of a condensed aromatic ring in size. $S\ a$, refers to an aliphatic chain attached to an aromatic cluster and assumed to end in a single methyl group. The parameter of A.B. is defined as follows:

$$A.\ B. = (f_a^p + f_a^s - f^*al)/f_d$$

Here, $f_a^p + f_a^s$ is all the attachments including aliphatic bridges and side chain. A.B. indicates the proportion of all possible aliphatic attachments except side chains, namely aliphatic bridges, in aliphatic groups.

The relationships between these three lattice parameters and thermal conductivity of coals is shown in Figure 6. An outlier exits in the each diagram, which is ignored here due to an uncertain reason to be determined. Figure 6a depicts a positive correlation between the mole fraction of aromatic bridgehead carbons and thermal conductivity of coals. This trend indicates that the size growth of polycondensed aromatic ring contributes to heat flow in coals. Polycondensed aromatic rings in a coal are the crystallites that transfer thermal energy and rely on lattice vibrations. This provides an essential reason why a larger size of polycondensed aromatic ring has a higher thermal conductivity.

Heat transfer in coal relies on continuous atom vibrations. Phonon collisions with grain boundary may cause the phonons to scatter and slow down the energy transport. The free-ended side chains (S.C.) impede heat transfer from an aromatic cluster to another one. Figure 6b shows a negative correlation between the number of side chains per cluster and thermal conductivity of coals. In contrast, aliphatic bridges (A.B.) among aromatic clusters ensure the continuity of atom vibrations and contribute to energy transport, as shown in Figure 6c.

For the tested samples in this work, the differences of the lattice parameters between anthracite and semi-anthracite coal are all more than 1.5 times greater than the differences between semi-anthracite coal and bituminous coal (Table 5). This value reflects that the lattice parameters of semi-anthracite are closer to bituminous coal than to anthracite. Therefore, the thermal conductivity of semi-anthracite coal is close to bituminous coal, and anthracite coal has a higher thermal conductivity with a higher value of lattice parameters.

4.2. Lattice Structure Control. Besides the lattice parameters, well-organized aromatic clusters are characteristics of a coal evolution to a crystal of graphite that has a much higher thermal conductivity. Multiple aromatic fringes are extracted from HRTEM digital images after processing by Image Processing Toolkit within Photoshop software (Figure 7). A same size region of image was selected to analyze for the three HRTEM micrographs with the same magnification. The micrographs were obtained on thin and sharp-edged demineralized fine coal particles. The lattice fringes show that all the coals are long-term disordered although with a short-term order (Figure 7). The short-term order of lattice fringes is more evident for the anthracite. Regional stacking generally occurs in all the coal lattice structures (Figure 7), especially in the anthracite.

Generally, molecular-level orientation such as lattice fringes alignment imparts behavioral anisotropy. Mathews et al. assumed that the dominant angle of lattice fringes is aligned.
parallel with the bedding plane of coal. The rose diagrams in Figure 7 illustrate that the lattice fringes of both bituminous coal and semi-anthracite have two clear orientations, which indicate a relative random arrangement of polycondensed aromatic rings. The disordered distribution of lattice fringes weakens the anisotropy of thermal conductivity of the bituminous coal and anthracite coal. On the contrary, the lattice fringes of the anthracite have a concentrated range of angle. These fringes with the approximate angle result in easier phonon transferring along the fringes, whereas scattering perpendicular to fringes are easy due to more grain boundaries.

The number of lattice fringes in selected region are 1012 for the bituminous coal, 993 for the semi-anthracite coal, and 566 for the anthracite. The fringe length indicates the shape of the aromatic ring based on previous studies. Their frequency of occurrence in the lattice fringes population is shown in Figure 8. The bituminous coal and semi-anthracite coal have similar types of aromatic rings with more benzene, naphthalene, and anthracene (especially naphthalene) than the anthracite. However, the other larger aromatic rings are more for the anthracite than for the bituminous coal and semi-anthracite coal. This result is consistent with the former conclusion by $^{13}$C NMR analysis.

5. CONCLUSIONS

The thermal conductivity of three ranks of Chinese coals and their control in molecular scale were studied by thermal analyzer, solid state $^{13}$C NMR, and HRTEM techniques.
aromatic ring improves the thermal conductivity since lattice vibrations contribute to heat flow. The aliphatic bridges (A.B.) among aromatic clusters ensure the continuity of atom vibrations and contribute to energy transport. However, the free-ended side chains (S.C.) impede heat transfer from an aromatic cluster to another one. The disordered distribution of lattice fringes weakens the anisotropy of thermal conductivity of the bituminous coal and anthracite coal.

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**Notes**

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