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The characteristics of methane adsorption capacity and behavior of tectonic coal

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The research of methane adsorption on tectonic coal is an important content to gas disaster prevention and coalbed methane (CBM) exploration in outburst coal seams. Many projects of methane adsorption capacity and behavior of tectonic coal, such as adsorption difference between tectonic coal and its untectonic coal, adsorption evaluation of tectonic coal, factors for adsorption capacity of tectonic coal, gas-solid coupling feature in tectonic coal and supercritical adsorption phenomena of tectonic coal, were carried out by scientists. Combined with a long-term study on organic matter structure and methane adsorption of tectonic coal, the author summarized recentyears' researches on adsorption capacity and behavior of tectonic coalmethane at home and abroad from the dispute of adsorption ability determination, the thermodynamic characterization of methane adsorption capacity, and the methane adsorption behavior of quantum chemical calculation of the adsorption and the gas content calculation based on loss compensation, respectively. It is believed that the coal structure controls the methane adsorption capacity and behavior characteristics of different tectonic coals, and from the perspective of thermodynamics, the adsorption capacity of different types of coals can be better distinguished. In the future, a more scientific and complete quantum chemical calculation of methane adsorption by tectonic coal and a compensation method based on instantaneous emission loss should be established, so as to better reveal the methane adsorption behavior of tectonic coal and the mechanism of coal and gas outburst. The research has a reference to fine research of coal adsorption and CBM exploration practices.

KEYWORDS

tectonic coal, methane adsorption, adsorption capacity, adsorption behavior, structure-property relationship, gas content

Introduction

The theoretical research and engineering practice of gas geology attach great importance to the research on tectonic coal (Aguado and Nicieza, 2007; Wold et al., 2007; Jiang et al., 2009; Yan et al., 2013; Zhang et al., 2021; Lin et al., 2022; Tu et al., 2022): On the one hand, coal and gas outburst is the result of the combination of tectonic coal, gas and tectonic actio. The "Regulations on Prevention and Control of Coal and Gas Outbursts" formulated by the State Administration of Work Safety of China even has listed tectonic coal as the first index for predicting the risk of coal seam outbursts; on the other hand, the low strength, low permeability and complex gas-solid coupling relationship of tectonic coal make coalbed methane (CBM) extraction more difficult. For example, the gas geology community often takes the prediction of tectonic coal as the basis for predicting coal and gas outburst, and discusses why tectonic coal has the risk of outburst from the aspects of coal structure and mechanical properties (Cao et al., 2003); when drilling a seam with developing tectonic coal, the instantaneous gas emission is large, making it difficult to accurately measure the gas content, which in turn affects the accuracy of gas emission prediction seriously. Perhaps it was because of the existence of tectonic coal, an associated "product" that shared the same parent material with normal coal (in this paper, the "untectonic coal" which is not affected by structural deformation is used to represent "normal coal"), when predecessors studied the characteristics of gas adsorption of coal, they paid special attention to the gas adsorption of different types of destruction coals (different types of tectonic coals). Ju et al. (2004) divided tectonic coal into three deformation sequences: brittle deformation, brittleductile and ductile deformation according to the tectonic deformation mechanism system. Jiang et al. (2016) considered that the brittle series of tectonic coal, cataclastic coal, mortar coal, granulitic coal and schistose coal were favorable target layers for CBM development, while Mylonitic coal of ductile deformation was considered to be a high-incidence area of coal and gas outbursts. The special pore and fissure structure of tectonic coal determined the coalbed methane (gas) adsorption capacity and behavior characteristics different from that of untectonic coal, and generally believed that the characteristics of structural fragmentation, large specific surface area and large molecular gap made tectonic coals have stronger adsorption capacity for methane (Qu et al., 2015). However, there are also reports that there is no significant difference in methane adsorption capacity between tectonic coal and co-existing untectonic coal, or the increase in the degree of damage does not improve the methane adsorption capacity of coal (Wang et al., 1993; Dong., 2019).

At present, researchers mostly use experimental methods to study the gas adsorption capacity and behavior characteristics of coal. The experimental method is to reflect the adsorption characteristics of coal samples from a macro perspective, the control variates method is often used to study the influence of a certain factor (such as temperature, pressure, etc.) on the gas adsorption capacity of coal, and the single-layer adsorption model (Langmuir adsorption model) and the multi-layer adsorption model (BET model) are mainly used for calculation. Scholars have done a lot of experimental research on the methane adsorption characteristics of tectonic coal. Zhang et al. (2007) and Jiang et al. (2009) believed that in the process of tectonic coal deformation, in addition to the evolution of chemical structure and coal macromolecular structure to order and high aromatization, many low molecular compounds and gaseous hydrocarbons (gas) were produced, which further improved the gas-bearing performance of the tectonic coal distribution area. Ju et al. (2008) carried out isothermal adsorption and desorption experiments on untectonic coal and tectonic coal under equilibrium water conditions, and found that due to differences in pore structure and chemical structure, the adsorption and desorption mechanism of tectonic coal was different from that of untectonic structural coal. Pan et al. (2012) believed that the adsorption of coalbed methane was related to the deformation structure of tectonic coal at different temperatures and pressures, and the methane adsorption of coal decreased with the increase of temperature. At a certain temperature, the adsorption of methane by different structural deformed coals was also different. For example, at a constant temperature of 30°C, the adsorption capacity of strong brittleductile deformed coal was significantly higher than that of weakly brittle deformed coal and strong brittle deformed coal; under low pressure conditions, the adsorption capacity of strong brittle deformed coal was significantly higher than that of weakly brittle deformed coal; however, with the increase of pressure, the adsorption capacities of the two coals tended to be the same. Cheng and Pan, 2020 found that the Langmuir volume and Langmuir pressure of tectonic and untectonic coals showed a "U" shape relationship with the degree of metamorphism. It was believed that the affinity between methane and tectonic coal was stronger than that of untectonic coal, so tectonic coal had stronger methane adsorption capacity.

The most fundamental factor affecting the macroscopic experimental results of the gas adsorption capacity and behavior of tectonic coal is the change of its microstructure. Therefore, it is difficult to explore the adsorption characteristics of gas in tectonic coal from the mechanism only by means of experimental methods, and molecular simulation can be used to analyze the adsorption mechanism of gas molecules in coal reservoirs from the molecular level at the microscopic scale. Thus, in recent years, molecular dynamics simulation has gradually become an important way to solve the complex adsorption behavior of coal. At present, the molecular simulation methods of coal are mainly based on molecular dynamics and molecular mechanics theory, and use molecular dynamics (MD), molecular mechanics (MM), finite element simulation and other molecular simulation and statistical

methods to model the molecular structure and pore structure of coal. This allows people to more comprehensively and intuitively understand the structural characteristics of coal, and helps to study the physical and chemical properties of coal through computer means. Wang et al. (2016) used the Monte Carlo simulation method to study the relationship between the surface structure of coal and the amount of methane adsorption based on the advanced evolution structure characteristics of tectonic coal, believing that the methane adsorption capacity of coal per unit mass decreased with the increase of the number of stacking layers of aromatic units, and the existence of defects and oxygen-containing functional groups was not conducive to the adsorption of methane. Han et al. (2017) used quantum chemical calculations and discussed the role of structural defects in anthracite-grade tectonic coals and their effects on macromolecular structure. The results showed that stress could promote the evolution of chemical structure of coal. Song. (2019) constructed a macromolecular structure model of mylonite coal, and a grand canonical monte carlo (GCMC) simulation of methane adsorption was performed. By comparing with the interaction between untectonic coal and methane, the gas adsorption characteristics and microscopic interaction mechanism of mylonitic coal were explained.

A large number of studies have shown that the research on the complex and exclusive characteristics of tectonic coal not only establishes the basic theory of geological body structure and tectonic changes under the action of geotectonic movement, but also plays an important guiding role in engineering practice such as coal seam outburst risk prediction and underground gas drainage in CBM low-permeability enrichment areas (Pan et al., 2012; Li et al., 2019; Zhang et al., 2022). Based on the long-term research and knowledge in structural composition of organic matter in tectonic coal and coal-methane adsorption through the author's research group (Huan et al., 2015; Zhang et al., 2018; Wang et al., 2021a; Yang et al., 2021), this paper summarizes and prospects the progress made in the study of methane adsorption capacity and behavior of tectonic coal in recent years, hoping to share and communicate, and promote further research on the adsorption of tectonic coal, so as to provide a reference for the prediction and prevention of gas disasters in outburst coal seam and to improve the recovery ratio of coal seam methane.

Methane adsorption capacity of tectonic coal

Determination of the methane adsorption capacity of tectonic coal

On the research of methane adsorption capacity of tectonic coal, there are many reports at home and abroad, but the understanding is different. Early research results show that there is no significant difference in the methane adsorption capacity of tectonic coal and untectonic coal (Wang et al., 1993), and the volume of methane adsorption does not increase with the increase of the degree of coal damage; In recent years, relevant research results also show that there is indifference in gas adsorption volume compare with tectonic coal and untectonic coal, while the gas adsorption rate of tectonic coal is higher than that of untectonic coal (Dong, 2018). However, Zhang et al. (2006) pointed out that the methane adsorption capacity of tectonic coal is significantly higher than that of untectonic coal when discussing the controlling factors of coal adsorption capacity; Wang et al. (2012) and Hu et al. (2016) conducted a comparative study on tectonic coal and untectonic coal in Jincheng, Jiaozuo, WuYi, Pingdingshan and other mining areas, concluding that the adsorption limit of tectonic coal is greater than that of untectonic coal on the whole.

In the experimental study of high pressure isothermal adsorption of methane on coal (Figure 1), it has been found that: the supercritical adsorption phenomenon occuring in the high pressure section, in which the adsorption pressure increases and the adsorption capacity decreases (Figure 1A), is quite different from the I~V adsorption isotherms that are generally believed to conform to the Langmuir equation (Figure 1), and the adsorption mechanism is different from that of gases below the critical temperature (Gao. 2011). The experimental results of Ju and Li, 2009 show that the methane adsorption capacity of weakly deformed coal increases with the increase of adsorption pressure, which conforms to the Langmuir equation, while the adsorption isotherms of strong and ductile deformed coal show different types (Figure 2). According to its analysis, desorption occurs when the adsorption pressure reaches a certain level, and the tectonic coal is compressed and deformed during the process, causing the deformation of its macromolecules and pore structure, and forming thin bottleneck pores; with the increase of adsorption pressure, the pores gradually closes, resulting in the decrease of adsorption heat and the desorption behavior, which leads to the decrease of methane adsorption capacity.

In view of the different understandings of the adsorption capacity of different types of tectonic coals and untectonic coal, and the factors affecting the capacity of methane adsorbed by coal are vast and complex, it is obviously meaningless to find out the strong and weak relationship between the methane adsorption capacity of different types of tectonic coals and co-existing untectonic coal only by discussing the difference in adsorption amount. Under normal formation conditions, CH_4 and N_2 in coal with extremely low critical temperature are supercritical gas, and CO_2 in coal will also be in supercritical state with the increase of burial depth. Apparently, the author suggests it is necessary to start from the perspective of studying the difference in methane adsorption energy between different types of tectonic coals and symbiotic untectonic coal, combine with the research idea of gas supercritical adsorption, and determine which coal of different



FIGURE 1

Type of adsorption isotherm: (A) Adsorption isotherm of methane and coal under critical temperature, (I~V) Adsorption isotherms of the Langmuir equation under critical temperature (modified from Gao., 2011).



coal in brittle deformation coals; (**D**) schistose-wrinkle structure coal in brittle-ductile superimposed coals; (**E**) scaled structure coal in brittle-ductile deformation coals; (**F**) mylonitic structure coal in ductile deformation coals. (Ju and Li, 2009, reprinted with permission of the copyright holders).

structure types has a stronger or weaker adsorption capacity for methane through experimental analysis and theoretical estimation, the coal seam gas content is also calibrated accordingly. Jian et al. (2014), Wang et al. (2016) and Song et al. (2017), have carried out a lot of research in this area, and the progress is slow at present, but it does not affect the theoretical value of the problem exploration and the engineering application value of the development of high-performance coal-based methane storage materials with tectonic coal as precursor (Zhang et al., 2017).

Thermodynamic characterization of methane adsorption of tectonic coal

As mentioned above, people have different opinions on whether the methane adsorption capacity of different types of tectonic coals must be greater than that of symbiotic untectonic coal. It is a new idea to use adsorption energy to examine the adsorption capacity of coal of different damage types. Since there is no method to directly test the adsorption energy of coalmethane, the adsorption heat can be used instead of the



adsorption energy. The basic process of adsorption heat index characterization is: based on the adsorption isotherm experiment, the real adsorption capacity, that is, the absolute adsorption capacity, is calculated with the help of the adsorption model, and the adsorption heat index and the thermodynamic effect of coal-methane adsorption are combined to calibrate the adsorption capacity of tectonic coal for methane. Two common adsorption heats, limit adsorption heat and isosteric adsorption heat, can be calculated from adsorption isotherms. Among them, limit adsorption heat is the effect of adsorption heat when the adsorption amount tends to zero, and is often used as a representative value of heat of adsorption; isosteric adsorption heat is a fractional amount, that is, the thermal effect generated by the increase in the amount of adsorption during the adsorption process.

This research group has carried out the isotherm adsorption experiment of methane on fat coal rank tectonic coal and coexisting untectonic coal from low temperature to room temperature, and has obtained a series of supercritical adsorption isotherms of methane on tectonic coal at 263-303 K, showing supercritical adsorption characteristics, namely: the methane adsorption capacity of both tectonic coal and untectonic coal increases with the increase of equilibrium pressure (Figure 3), and the adsorption curve decreases when the equilibrium pressure reaches a certain value, indicating that the adsorption capacity has a maximum value; the adsorption curve is type I before the maximum appears (Figure 1), and the pressure point at which the maximum appears lags with increasing temperature (Wei., 2012). The adsorption capacity measured by the volumetric method above is the excess adsorption capacity, not the real adsorption capacity of coal to methane, that is, the absolute adsorption capacity, and cannot objectively describe the adsorption of coal reservoirs. Therefore, before calculating the heat of adsorption, in order to explore the methane adsorption characteristics of tectonic coal, it is necessary to establish an absolute adsorption capacity calculation model and obtain the absolute adsorption capacity curve of methane adsorption of tectonic coal according to the experimental data (Zhou and Zhou, 1998; Wei., 2012). As shown in (Figure 4), the absolute adsorption curve no longer has a maximum point, before reaching the adsorption equilibrium, the adsorption amount increases with the increase of pressure. It belongs to the I-type isotherm adsorption amount is the real value of coal seam adsorption of methane, which has important guiding significance for accurately predicting coal and gas outburst.

The coal surface is the main place for the adsorption/ desorption of coalbed methane. When the adsorption of methane and coal occurs, the coal surface tension will decrease (surface thermodynamics), so the change of coal surface tension in the process of coal-methane adsorption can be calculated by the Gibbs formula. Different from the aforementioned adsorption heat studies, in order to further investigate "whether the methane adsorption capacity of tectonic coal is stronger than that of symbiotic untectonic coal" from the perspective of the surface energy drop of coal adsorbing methane, we used the IS-100 isothermal adsorption/ desorption instrument from Terra Tek, United States. The isothermal adsorption experiment of methane with tectonic/ untectonic coal with 293, 303, and 313 K coal ranks being coking coal was carried out (Figure 5), and the maximum adsorption equilibrium pressure was 8 MPa (Jian et al., 2014). The results show that: the process of coal adsorption of methane



Diagram of excess methane adsorption and absolute adsorption isotherms of fat coal rank tectonic/untectonic coal. (A) tectonic coal; (B) untectonic coal (modified from Wei, 2012).



is a process of coal surface energy reduction (Table 1), the overall performance of the experimental coal samples is that the surface energy reduction of tectonic coal adsorbing methane is greater than that of co-existing untectonic coal, both the increase in temperature and the increase in equilibrium pressure make the surface energy drop value of the tectonic coal larger than that of the co-produced untectonic coal (Figure 6). Berkheim has pointed out that "the expansion deformation of solid is proportional to its surface energy drop," and combined with the surface energy drop of coal adsorbing methane and theoretical calculations, it is believed that adsorption deformation can promote layer slip and dislocation of coal. Obviously, the surface energy reduction of tectonic coal adsorbing methane is greater than that of co-existing untectonic coal, but its coal strength is lower, and it is prone to adsorption expansion, deformation and damage, which may be one of the important reasons why tectonic coal is closely related to coal and gas outburst.

Methane adsorption behavior of tectonic coal

Quantum chemical calculation of methane adsorption of tectonic coal

The high complexity and heterogeneity of coal structure make its adsorption behavior more complicated than that of conventional solid adsorbents (Crawford and Mainwaring, 2001; Wang et al., 2021a; Wang et al., 2021b). Theoretical and experimental studies show that the structure and composition of coal are closely related to the coal organic polymer network structure and the low molecular compounds dispersed in it, which are the intrinsic factors affecting the adsorption/desorption of coalbed methane (Ji et al., 2015; Yang et al., 2021). The quantum chemical calculation of the coal surface

Untectonic coal	293 K	Р	1.6	2.57	3.66	4.77	5.91	6.92	7.89
		$ riangle \gamma$	6729.69	10001.32	131183.73	16017.10	18595.63	20655.44	22469.66
	303 K	Р	1.6	2.57	3.63	4.69	5.77	6.86	7.89
		$ riangle \gamma$	7274.68	10779.02	14084.47	16975.93	19591.49	21961.69	23998.70
	313 K	Р	1.64	2.71	3.72	4.77	5.72	6.62	7.77
		$ riangle \gamma$	7870.48	11887.58	15145.09	18117.88	20521.76	22595.56	25007.63
Tectonic coal	293 K	Р	1.99	2.99	4.03	5.1	6.17	7.14	8.28
		$ riangle \gamma$	11284.40	15345.04	18907.35	22061.95	24825.99	27066.88	29442.06
	303 K	Р	1.95	2.94	3.98	4.99	6.02	7.07	8.08
		$ riangle \gamma$	11539.22	15744.6142	19467.4509	22586.6485	25385.8766	27928.4180	30135.6364
	313 K	Р	2.01	3.1	4.1	5.13	6.06	6.94	8.05
		$ riangle \gamma$	12341.84	17068.34	20710.154	23958.04	26553.65	28774.43	31309.89

TABLE 1 Statistics of surface energy drop of tectonic/untectonic coal adsorbed methane at different temperatures (modified from Jian et al., 2014).

Note: P- adsorption equilibrium pressure, MPa; △γ-surface energy drop, J/m².



can reflect the coal surface properties, and the adsorption and interaction studies of methane molecules on solid surfaces are multi-faceted, so the theoretical calculation of methane adsorption on coal surfaces by quantum chemical methods is more rigorous.

Chen et al. (2000) has modeled atomic clusters on the coal surface and has carried out quantum chemical *ab initio* calculations with base set (STO-4-31G). The results show that the interaction between methane and the coal surface is anisotropic. When methane overlaps with a regular triangular pyramid on the coal surface, the energy is the lowest, and the maximum adsorption potential is 2.65 kJmo1⁻¹, which belongs to the category of physical adsorption and is manifested as surface

condensation. Wang et al. (2008) has proceeded DFT calculation on the mechanism of coal and gas outburst, showing that the adsorption of coal and methane is physical adsorption, and the adsorption energy is 1.16 kJ mo1⁻¹, when the coal-methane adsorption system absorbs electromagnetic waves in the 23-61 cm⁻¹ frequency range, the methane molecules can be desorbed, that is, coal and gas outburst occurs. In this paper, according to the XRD analysis data of co-existing tectonic coal and untectonic coal in the same layer (Table 2), and corresponding to each index in the anthractie coal aromatic ring structure, the untectonic coal and tectonic coal are constructed combined with the unit cell parameters of graphite in the inorganic crystal structure database (ICSD), the models are denoted as M1 and M2 (Jing et al., 2010), respectively (Figure 7). Using the adsorption isotherm calculation task in the sorption module of Materials Studio software, at 298 K and 303 K, the isotherm adsorption curves of untectonic coal (M1), tectonic coal (M2) and methane are calculated respectively (Jing et al., 2010) (Figure 8), shows that the methane adsorption curves of the two models are consistent with Langmuir adsorption characteristics. At 298 K, the saturated adsorption capacity of untectonic coal (M1) is about 28 methane molecules, and that of tectonic coal (M₂) is about 50 methane molecules, and the methane adsorption capacity of the tectonic coal is about twice that of the untectonic coal (Figure 8A). At 303 K, the methane adsorption behavior of the tectonic/untectonic coal has the same characteristics as that at 298 K (Figure 8B). Theoretical calculation studies combining quantum chemistry and model intermolecular interactions suggest that: during the formation of tectonic coal, the coal deformation increases, the structure of the side chain on the aromatic core of the coal changes, and the steric hindrance weakens, reducing the rate of methane passing through the coal body. In addition, the side groups and even the polymer main chain are partially broken to form a high free

Coal sample number	Structure type of coal body	Ro,max/%	d002/Å	Lc/Å	La/Å
M1	untectonic coal	3.99	3.463	22.908	16.201
M2	tectonic coal	4.78	3.446	25.798	19.937

TABLE 2 R_{o,max} and XRD analysis data of different types of anthractie coal (modified from Jing et al., 2010).



energy center, which is conducive to the diffusion of methane, when the energy of free methane is equal to or higher than the binding energy of the side groups on the aromatic core of coal, methane can naturally penetrate into the side groups of the coal structure, and occupy vacancies (adsorption sites) there, which adsorption behavior reduces the free energy of coal. The adsorption energy of methane on the surface of tectonic coal is greater than that of untectonic coal (Table 3), indicating that tectonic coal can store a larger amount of methane than coexisting untectonic coal (Wang et al., 2009). The study of methane activation and adsorption by coal surface structure is helpful to deeply understand the methane adsorption behavior on coal surface, and reveal the mechanism of coal and gas outburst from the molecular level.

Determination of gas content in tectonic coal based on compensation for instantaneous emission loss quantity

A large number of on-site tests and survey statistics show that the two tentative indicators for predicting the risk of coal seam outbursts tentatively set in the "Regulations on Prevention and Control of Coal and Gas Outbursts," the initial velocity of gas emission (ΔP , the critical value is 10 ml s⁻¹) and coal seam gas content (W, the critical value is 8 m³ t⁻¹), almost become insensitive indicators (or the critical value is inappropriate) in the coal seam developed in tectonic coal, which lacks scientificity and reliability, and should attract the attention of researchers. However, in the current engineering practice, the risk of coal seam outburst is still judged accordingly. Most of predecessors



TABLE 3 Gas and different types of coal adsorption distance, adsorption energy and bond order (modified from Wang et al., 2009).

Adsorbed gas	Structure type of coal body	Adsorption distance/Å	Adsorption energy/(kJ/mol)	Bond order
CH ₄	untectonic coal	2.11	-12.34	0.01
	tectonic coal	1.94	-15.75	0.02



have used isothermal adsorption/desorption experiments to explore the gas properties of coal, but the research on the instantaneous emission mechanism of coal and gas is weak, which is the basis for the determination of gas content in tectonic coal and the prediction of coal seam outburst risk (Zhang and Y, 2014). It can be seen that revealing the initial gas desorption law of tectonic coal, finding out the loss quantity in the determination of gas content in tectonic coaldeveloped coal seams, and establishing a compensation method for the loss quantity have important practical significance for mine gas disaster prediction and coalbed methane resource evaluation.

Based on coal seam gas content, the "Regulations on Prevention and Control of Coal and Gas Outbursts" sets the critical value for predicting regional outburst risk as 8 m3 t-1. However, when the developed coal seam of tectonic coal is drilled, the instantaneous gas emission is large (Figure 9), which makes it difficult to accurately measure the gas content (Fu et al., 2008). It is generally believed that the main reason is: coal seam gas content includes loss, on-site desorption and residual gas, among which the measurement errors of on-site desorption and residual gas are negligible, and the error mainly comes from the calculation of the loss; when drilling a developed coal seam of tectonic coal to measure the gas content, the rapid expansion of the coal micro-cracks makes the relatively closed original cracks and pores communicate and connect, resulting in that the instantaneous gas desorption rate in the coal is great, and the instantaneous amount of escape is large, so accurately calculating the gas loss of tectonic coal is a difficult point in the determination of coal seam gas content.

Domestic and foreign scholars have done a lot of research on the problem of loss compensation in the process of gas content determination (Qi et al., 2007; Liu et al., 2013; Song et al., 2021; Yi et al., 2022). The established empirical/semi-



FIGURE 10

SEM images comparing morphological features of fat coal rank tectonic/untectonic coal (modified from Gao., 2011). Note: T_1 , T_2 and T_3 are tectonic coal samples; U_1 , U_2 and U_3 are untectonic coal samples.

empirical loss calculation formula plays an important role in revealing the gas desorption law and calculating the gas loss, but it also has certain limitations. For example, the error of Barrel's formula increases with the extension of desorption time; the Venter formula conforms to the law of initial gas desorption, but the error increases with the extension of desorption time; the Bott formula is inconsistent with the initial desorption amount of gas; the exponential formula is the basis for determining the gas content of the coal seam by the underground desorption method of coal mines in China, but when the measured coal seam is of tectonic coal, the calculated gas loss has a large deviation from the actual value. In recent years, China has reported many studies on gas desorption law and gas loss. For example, He (2010) has obtained the influence coefficient of temperature on gas desorption of tectonic coal through regression analysis of gas desorption of tectonic coal at different temperatures $\eta = \exp [0.012 \cdot (T-10)]$, and then obtains the calculation formula $Q_T = \eta \cdot Q_{I0}$ of gas loss at different temperatures; Li (2010) has investigated the effects of desorption empirical formulas, desorption time and



desorption temperature on the gas desorption amount, and consideres that $Q_{T(t)} = \exp [0.011 \cdot (T-20)] K \cdot \sqrt{t}$ is applicable to the calculation of gas loss when the desorption temperature changes; Li et al. (2013) have believed that the Vent method has good stability and reliability in calculating the gas loss of tectonic coal.

Based on the relationship between the residual gas volume $(W_C, \text{ ml/g})$ and gas pressure (P, Mpa), remaining in the Changcun mine's tectonic coal and co-existing untectonic coal after being exposed for a certain period of time, the research group has established the following content-pressure calculation formula (Xie., 2014):

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\begin{cases} \text{untectonic: } W_c = 11.483 P^{0.4491}, (R^2 = 0.9787, 0 \le t < 30 \min) \\ \text{untectonic: } W_c = 4.6927 \ln P + 10.949, (R^2 = 0.9835, 30 \min \le t < 1h) \\ \text{tectonic: } W_c = 10.51 P^{0.4603}, (R^2 = 0.97710 \le t < 30 \min) \\ \text{tectonic: } W_c = 4.3722 \ln P + 9.7734, (R^2 = 0.9849, 30 \min \le t < 1h) \end{cases}
(1)
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After on-site inspection, it is considered that the proposed calculation method of gas loss, that is, the method of calculating gas content from residual gas, is accurate and reliable.

Understand the methane adsorption capacity and behavior characteristics of tectonic coal from the structureproperty relationship

The complex structure and heterogeneity of tectonic coal make it difficult to accurately characterize the influence of various factors on its adsorption capacity and behavior, so people are more inclined to use the academic thought "structure-property" to explore its structural relationship. The adsorption properties of coal are not only related to the physical properties such as specific surface area and pore size distribution (Wang et al., 2018), its surface chemical structure/composition also has an important influence on the adsorption behavior. The essence of the difference in the methane adsorption capacity and behavior characteristics of coal with different coal structure is differences in physicochemical structure of coal, which is initiated by different degrees of brittle cracking, crushing or ductile deformation or superimposed failure of primary structure of tectonic coal under the action of one or more periods of tectonic stress.

Xu et al. (2010) observed the surface structure of briquette with different particle sizes, noticing that the smaller the particle size of briquette samples was, the higher the degree of pore development was, and the shear strength decreased with the increase of particle size; adsorption experiments showed that the briquette made of large particle size coal powder had larger porosity, better connectivity and permeability. Li et al. (2003) investigated the pore structure of tectonic coal in China's Pingdingshan mining area by Mercury intrusion and found that compared with co-existing non-tectonic coal, the porosity of tectonic coal was 3-8 times higher and the specific surface area was 2-10 times higher. Among them, brittle deformed coal had higher porosity, large specific surface area and wider cracks, while ductile deformed coal had supramaximal specific surface area and narrow cracks. The introduction of High Resolution Transmission Electron Microscopy (HRTEM) technology has facilitated the identification of nano-scale pores in coal, which is considered to be the main storage space for coalbed methane (Song et al., 2019; Song et al., 2020). HRTEM results of different types of tectonic coals show: from brittle deformed coal to ductile deformed coal, the cumulative pore volume, total specific surface area and nitrogen adsorption increase rapidly with the increase of deformation degree (Ju et al., 2005). Li et al. (2012) discussed the influence of deformation of different mechanisms on the coal macromolecules-nano-scale pore structure, and established a coupling model of tectonic coal macromolecules-nano-scale pore structure, believing that the change of the macromolecular structure of coal during the formation of tectonic coal is the main reason for the change of the nano-scale pore structure of coal. Our research group has observed the surface morphology of tectonic coal/symbiotic untectonic coal with Scanning Electronic Microscopy (SEM), finding that as shown in Figure 10, micropores on the inner surface of tectonic coal are prevalent, and compared with untectonic coal, it has a more complex pore structure (Gao. 2011). It is considered that the change of coal macromolecular structure caused by tectonic extrusion and shearing promotes the development of coal microporous structure, which is the main space for methane adsorption and diffusion. This is consistent with the research on the correlation between the ultrastructure of tectonic coal and chemical/physical coalification carried out by Ju and Li, 2009.



between L_a/L_c and $R_{o,max}$ (modified from Zhang et al., 2016).

Adsorption will occur on the new surface generated by the deformation and rupture of the coal, combining the genesis and physical properties of tectonic coal, our research group believes that adsorption is involved in the formation process of tectonic coal (Zhang and Y, 2014). It is expressed as: during the formation of tectonic coal, the structure of coal is broken and pulverized, increasing the pore space and inner surface area of coal, and providing more space for methane accumulation; the gas adsorption causes the expansion and deformation of the coal rock, which generates the change of the pore structure of the coal rock. Methane in coal is mainly in the adsorbed state, and compared with the symbiotic untectonic coal, the microstructure characteristics of tectonic coal show: on the one hand, the development of micro-cracks and pore structures in tectonic coal provides space for gas accumulation, which may increase the gas storage capacity of coal (Qu, 2011); on the other hand, the tectonic coal structure evolves in advance (Figure 11), and the aromatic layers of coal nuclei are stacked vertically and stretched horizontally (Figure 12), which may enhance the adsorption capacity of coal (Zhang et al., 2016), and the difference in adsorption capacity of methane should be one of the reasons for the prominent danger of tectonic coal. In addition, in terms of nature that making tectonic coal prone to trigger coal and gas outbursts, adsorption behavior aggravates the fragmentation of the coal structure, and while reducing the coal strength, new pore spaces and inner surfaces may be formed to accumulate more gas; the coal shrinkage after gas discharge can cause pressure relief. The uneven distribution of gas in the coal seam will cause uneven distribution of stress, and the release of adsorption behavior will constitute a potential driving force for starting and promoting coal and gas outbursts.

Conclusion

The research on the methane adsorption capacity and behavior characteristics of tectonic coal is of great significance to the production practice activities, such as coalbed methane resource exploration (energy development) and mine gas outburst (safety mining). Combing with the author's long-term exploration and practice in the study of structural composition of tectonic coal and structure relationship of coal-methane adsorption, in this paper, the author summarized recent-years' researches on adsorption capacity and behavior of tectonic coal-methane at home and abroad from the dispute of adsorption ability determination, the thermodynamic characterization of methane adsorption capacity, and the methane adsorption behavior of quantum chemical calculation of the adsorption and the gas content calculation based on loss compensation, respectively. It is believed that the coal structure controls the methane adsorption capacity and behavior characteristics of different tectonic coals, and from the perspective of thermodynamics, the adsorption capacity of different types of coals can be better distinguished. In the future, a more scientific and complete quantum chemical calculation of methane adsorption by tectonic coal and a compensation method based on instantaneous emission loss should be established, so as to better reveal the methane adsorption behavior of tectonic coal and the mechanism of coal and gas outburst. It is hoped that we can share and communicate, and promote further research on the adsorption of tectonic coal, so as to provide scientific reference for the prevention

and control of outburst coal seam gas disasters and the improvement of coalbed methane recovery.

Author contributions

HZ: data curation, formal analysis, writing—original draft. XZ: conceptualization, data curation, supervision. YZ: conceptualization, funding acquisition, supervision. ZW: supervision.

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