

Research Article

Structural Characteristics and Physical Properties of Tectonically Deformed Coals

Yiwen Ju, Zhifeng Yan, Xiaoshi Li, Quanlin Hou, Wenjing Zhang, Lizhi Fang, Liye Yu, and Mingming Wei

Key Laboratory of Computational Geodynamics, College of Earth Science, Graduate University of Chinese Academy of Sciences, Beijing 100049, China

Correspondence should be addressed to Yiwen Ju, juyw03@163.com

Received 13 February 2012; Accepted 28 March 2012

Academic Editor: Yu-Dong Wu

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Different mechanisms of deformation could make different influence on inner structure and physical properties of tectonically deformed coal (TDC) reservoirs. This paper discusses the relationship between macromolecular structure and physical properties of the Huaibei-Huainan coal mine areas in southern North China. The macromolecular structure and pore characteristics are systematically investigated by using techniques such as X-ray diffraction (XRD), high-resolution transmission electron microscopy (HRTEM), electron paramagnetic resonance (EPR), nuclear magnetic resonance (NMR), and low-temperature nitrogen adsorption method. The results suggest that under the directional stress, basic structural units (BSU) arrangement is closer, and the orientation becomes stronger from brittle deformed coal to ductile deformed coal. Structural deformation directly influences the macromolecular structure of coal, which results in changes of pore structure. The nanoscale pores of the cataclastic coal structure caused by the brittle deformation are mainly mesopores, and the proportion of mesopores volume in ductile deformed coal diminishes rapidly. So the exploration and development potential of coalbed gas are good in reservoirs such as schistose structure coal, mortar structure coal and cataclastic structure coal. It also holds promise for a certain degree of brittle deformation and wrinkle structure coal of low ductile deformation or later superimposed by brittle deformation.

1. Introduction

Tectonically deformed coal (TDC) is a kind of coal in which, under mono- or multiphase tectonic stress fields, its primary texture and structure is significantly destroyed. Tectonic deformation could influence coal macromolecular or deform structure and enhance coalification to a certain degree [1–6]. Through structural shearing, graphitization can be facilitated [7, 8]. In fact, tectonic deformation can not only further alter coal's molecular structure at different degrees but also change the physical properties of reservoirs (such as porosity and permeability). Based on measurement of mercury intrusion, researchers compared micropore features of TDCs collected from different coalfields [9–12]. These experiments have indicated that the pore structure of TDCs showed greater medium pores and mesopore volumes than normal undeformed coals, but there was rare

difference in micropores between primary structure coals and TDCs. This indicated that structural deformation did not obviously influence micropore structure (less than 10 nm in diameter). Meanwhile, the nanoscale pore structure of TDCs is the main adsorption space for coalbed gas, and the macromolecular structure is an important factor for restricting the adsorption capacity in coal. Therefore, the problem of macromolecular structure and nanoscale pores in TDCs are worthy research topics [13–15], and the behavior of gas adsorption/desorption and permeability of micropore in coal have strong relationship with the occurrence and distribution of coalbed gas [16–24].

The deformation of coals causes changes of structure and chemical composition of coal because of differences in the property, mode, and intensity of stress and the deformation environment. Moreover, the physical properties of TDCs also vary significantly [5, 22]. Therefore, comprehensive studies

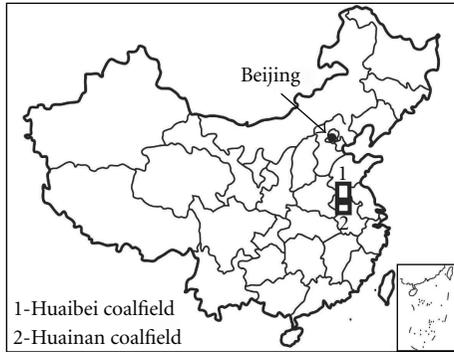


FIGURE 1: Huaibei-Huainan coalfield in the southern North China.

on stress action, strain environment, physical and chemical structures of TDCs, and physical properties of deformed coals will provide the theoretical basis for exploration and evaluation of coalbed gas resources and aid in understanding the prevention and control of coal and gas outbursts. These studies will also provide insight into gas emission, which is studied in the coalbed gas research field. In this study, the author would like to reveal the structures and the physical properties of tectonically deformed and metamorphic coals.

2. Geologic Background and Coal Structure in Huainan and Huaibei Mine Areas

The Paleozoic coal-bearing basins, especially in eastern China, underwent complexly multiphase structural evolution by compressing, shearing, and extending. This resulted in structural reworking of the primary structure of coal at different degrees, forming the different types of TDCs. The existing data from China have shown that tectonic deformation is an important factor in restricting the exploration and development of coalbed gas; it is also a direct cause of gas outbursts of coal mines.

In the Huaibei and Huainan coalfields, situated in the southern North China (Figure 1), coal measurement revealed Permo-Carboniferous systems which were formed in the Paleozoic and structurally influenced in the Mesozoic. The main coal-bearing areas were found in fault depression basins, especially in the syncline. The upwelling areas between depression basins have undergone destruction and denudation at different degrees. The different types of faults and folds in the distribution area of coal have given rise to obvious breakages and deformations in the coal seams, leading to changes of the coal structure. At the end of this process, different types of TDCs, such as cataclastic structure coal, mortar structure coal, schistose structure coal, mealy structure coal, and even wrinkle structure coal and mylonitic structure coal, were formed [5].

3. The Sample and Methods

The research areas possess six mine wells of the Suzhou mine area and the Linhuan mine area in the Huaibei coal mine region. Three of them are contained by the Xinji mine region,

Zhangji mine region and Panji mine region, which represent different tectonic units. The sample sets were collected from the Huaibei and Huainan coalfields (Figure 1). These samples belong to different kinds of TDCs, suffering from multiphase tectonic deformation and magmatic events.

In this study, two significant aspects were taken into account in the selection of the samples. The first aspect is the evolutionary degree of coal, namely, reflectance ($R_{o,max}$) of samples ranging from 0.76% to 3.59%, including low, middle, and high metamorphic coal. The second aspect is the characteristics of TDCs formed by different mechanisms. The samples of brittle deformation series, ductile deformation series, and brittle-ductile series were selected separately.

Because of different sedimentation and tectonic environments, coal often contains different maceral compositions and various kinds of mineral components, which greatly affect the results of coal structural testing. Therefore, coal samples must be prepared before testing and analysis to extract vitrinite components and take off inorganic (mineral) components.

On the basis of in situ investigation, detailed microstructural observations of these samples using optical and scanning electron microscopy (SEM) were described elsewhere. $R_{o,max}$ measurements were carried out on polished sections under halogen lamp and oil immersion conditions by using a Leitz Orthoplan/MPV-SP microscope photometer.

Using X-ray diffraction (XRD), high-resolution transmission electron microscopy (HRTEM), electron paramagnetic resonance (EPR), and nuclear magnetic resonance (NMR), systematic research has been conducted on the characteristics of macromolecular structure and chemical structure of different kinds of TDCs.

The instrument of X-ray diffraction testing is a D/Max-III B X-ray diffractometer which is made by Rigaku Inc. of Japan; the instrument of the HRTEM is a JEM-2010 HRTEM made in Japan by JEOL Inc.; the instrument of EPR testing is the E-109 electron paramagnetic resonance spectrometer made by Varian Inc. of America; the instrument of NMR testing is the Infinity 400 nuclear magnetic resonance spectrometer made by Varian Inc. of America. The experimental conditions of different instruments are discussed in detail by Ju et al. [5, 6].

The pore characteristics of TDCs have been determined by use of the low-temperature nitrogen adsorption method and scan electron microscopy (SEM). The permeability has been analyzed through experiments of in-lab permeability measurements of large diameter, in-site samples from TDCs.

The instrument used for low-temperature liquid nitrogen adsorption experiments is the ASAP-2010 made in America by Micromeritics Instrument Inc. This instrument is used for measuring the micropore specific surface area and distribution of pore diameter. In the gas-water phase permeability experiment, the whole core flow system, made by Terra Tek company, was utilized as the main instrument. This system was primarily used to simulate crustal pressure and rock permeability under natural oil reservoir pressure. The highest simulated confined pressure and fluid pressure were 70 MPa and 65 MPa, respectively. The measurement system was composed of the pressure system, the constant

temperature system, the control system, the core clamper and separator equipment.

4. Results

4.1. Forming Environments and Structure-Genetic

Classification of Tectonically Deformed Coals

4.1.1. Forming Environments of Tectonically Deformed Coals.

The Huaibei and Huainan coal mine areas underwent complexly multiphase structural and magma-thermal activities. Thus, under different metamorphic and deformed environments, various kinds of TDCs were formed in these mine areas [11, 20–22]. By combining deformational characteristics of macrocosms and microcosms of different kinds of TDCs, we recognized three kinds of metamorphic and deformed environments where TDCs were formed in the Huaibei and Huainan coal mine areas. The first type is the metamorphic and deformed environment of the low-rank coals ($R_{o,max}$ lower than 1.30%), which is based on the plutonic metamorphism; the second type is the metamorphic and deformed environments of the middle-rank coals ($R_{o,max}$ from 1.30 to 2.00%), which resulted from plutonic metamorphism and a superimposed magmatic thermal and dynamic metamorphism. The third type of metamorphic and deformed environment of the high-rank coals ($R_{o,max}$ greater than 2.00%) resulted from plutonic metamorphism and a superimposed, stronger magmatic thermal and dynamic metamorphism.

4.1.2. Structural-Genetic Classification of Tectonically Deformed Coals.

Based on the results of the in situ measurement of coal mines and microstructural observation, a structural-genetic classification system has been proposed that can be applied not only to the exploitation of coalbed gas but also to the prevention and control of coal and gas outbursts. In terms of the scale of hand specimens or samples of coal core from boreholes, TDCs formed by different deformational mechanisms have been divided into three deformation series and ten classes (Table 1). The brittle deformation series includes cataclastic structure coal, mortar structure coal, granulitic structure coal, mealy structure coal, schistose structure coal, and thin-layer structure coal. The ductile deformation series includes wrinkle structure coal, mylonitic structure coal, and ductile structure coal. The brittle-ductile series includes scaly structure coal. Compared to the previous classifications [17, 19, 25, 26] of brittle deformational coals, schistose structure coal and thin-layer structure coal have been added into this series. This is because these coals are quiet different from the cataclastic structure coals in structural and physical properties. Similarly, ductile structure coal has been divided into the ductile deformation series. For ductile deformational coals, wrinkle structure coal was considered to be formed in a compressing and shearing process, mylonitic structure coal is formed by strong ductile shearing, while ductile structure coal is formed during the creeping process of higher temperatures. Moreover, a brittle-ductile transition

type of coal has been proposed; this may include scaly structure coal.

4.2. Structural Evolution and Deformational Mechanisms of Tectonically Deformed Coals.

Temperature is the main factor that causes the change of coal chemical structure; it also facilitates metamorphism and improves coal rank [27, 28]. The temperature and stress together lead to change in coal macromolecular structure and its chemical components [4, 5]. Under thermal and stress conditions, how does the structural evolution of a deformational mechanism, over different kinds of TDCs, take place?

Thermal action and directional stress are the important factors in changing coal macromolecular structures and destroying bond forces. The vitrinite reflectance anisotropy (VRA) characteristic of deformed coals was well known and was interpreted from the ordering degree promoted by the stress [5, 29, 30]. Meanwhile, $R_{o,max}$ optical characteristics are shown in different kinds of TDCs (Table 2). Because of thermal values and stress, the value of $R_{o,max}$, the size of stacking (L_c) and extension (L_a) of basic structural units (BSU), carbon aromaticity (f_a) and radical density (N_g) increase. Accordingly, layers of the microcrystal (N) and the number of benzene rings of each layer also increase, whereas the spacing among monolayers of aromatics (d_{002}), L_a/L_c and the aliphatic spacing (d_r) continually decrease. In addition, the values of L_a/L_c (formed under the condition of stress) are obviously lower than those formed under conditions of thermal action. Therefore, the whole coalification process is the course of condensation of aromatic rings in which pores change continually [31]. Furthermore, when the structural deformation enhances, the faint orientation and bad ordering domain of schistose structure coal formed by brittle deformation extend to a local orientation, a strong orientation, and an integer orientation orderliness (Table 2 and Figure 2). It is obvious that stress has affected the chemical structure of coal. However, in the past, researchers had not convinced that the effect of tectonic stress is not the same for different kinds of ductile deformation coals. The macromolecular structure of wrinkle structure coal shows smaller changes, while the macromolecular structures of mylonitic structure coal and ductile structure coal change significantly.

4.3. Physical Properties of Tectonically Deformed Coals and Their Relation with Structure

4.3.1. Porosity of Tectonically Deformed Coals.

Coal contains a large numbers of pores [10, 13, 15], even more in TDCs. The nanoscale pore diameters of different kinds of TDCs have been grouped into four types: mesopores (15 ~ 100 nm), micropores (5 ~ 15 nm), submicropores (2.5 ~ 5 nm), and ultramicropores (<2.5 nm) [22]. This was done according to the average pore diameter of the natural distribution at three points: 15 nm, 5 nm, and 2.5 nm, and through combining the adsorbed gas and diffused features.

In this study, the parameters of pore structure were tested and the experimental results were shown in Table 3.

TABLE 1: The structural-genetic classification of TDCs.

Deformation series	Type of tectonically deformed coals	Texture and structure	Structural fracture and wrinkle	Breaking degree	Micro characteristics	Deformational mechanism and environment	
Brittle deformation series	Cataclastic structure coal	Band texture could be seen; layer structure was preserved	Multidirectional fracture cuts; no obvious displacement	More hardness; crumbing is difficult	Tensile fracture, shear fracture, and compressional fracture	Multi directional compression and extension; extension was the predominant role	
	Mortar structure coal	Little band texture was seen; lentoid structure was formed	Multidirectional fracture cuts; obvious displacement in the mortar	Broken into detritus from 1 to 5 cm; angular shape			
	Granulitic structure coal	Primary structure not present; disorder of layers	Multi directional cross fracture; particles were rotated	Broken into detritus, 1 cm			
	Mealy structure coal	Primary structure not present; powdered particles formed	No obvious directions for the particles	Be pinched into powered particles			Strong compressional fracture zone or scaly structure coal be reconstructed later
	Schistose structure coal	Band texture could be seen; layer structure was preserved	Single directional fracture; little displacement on the surface	Broken into detritus from 1 to 5 cm, shard shape			Single directional compression and extension or shearing strain environment
	Thin-layer structure coal	Little band texture was seen; layer structure was not obvious	Single directional fracture; obvious displacement on the surface	Broken into detritus, 1 cm			
Brittle-ductile series	Scaly structure coal	Primary structure not present; scaly structure was formed	Multi directional cross-fracture; coal was cut and wrinkled; the particles could be rotated	Broken into detritus, 0.5 cm; shard shape	Shear fracture, cleavages	Strong shearing or cleaving strain environment	
Ductile deformation series	Wrinkle structure coal	Primary structure not present; wrinkle structure was formed	Coal was wrinkled	Broken into detritus, 0.5 cm, shard shape	Wrinkle, foliated coal, S-C structure, eyed structure, optical anisotropy structure and wavy extinction	Strong shearing or long and low stress deformation	
	Mylonitic structure coal	Primary structure not present; mylonitic structure was formed	The particles had directional arrangement; flow structure was formed	Was pinched into powered particles			
	Ductile structure coal	Primary structure not present; lump and lentoid structures were formed	No obvious physical deformation	More hardness; crumbing was difficult			Long and low stress creep; ductile flow at high temperatures

Research was performed on the nanoscale pore structure in metamorphic-deformed environment of low-rank coal and the different deformational series of structural coal. Our results have demonstrated that the volume of mesopores reduces rapidly while the volume of micropores and pores whose diameters are lower than micropores increase in the metamorphic deformed environment of low-rank coal. Also, submicropores and ultramicropores can be found, the specific surface area of mesopores reduces greatly while and

submicropores increase rapidly. The change of pore parameters in nonhomogeneous coal structure is similar to the weak brittle deformation coal. For different types of tectonic coals formed in metamorphic-deformed environments of middle- and high-rank coal, pore parameter change is basically consistent with the metamorphic-deformed environment of low-rank coal. But, there are differences in the changes among different kinds of tectonic coals. To sum up, the conditions of temperature and confining pressure can play

TABLE 2: The characteristics of structural parameter of tectonically deformed coals.

Type of tectonically deformed coals	Coal sample's number	$R_{0,max}$ (%)	$R_{0,bl}$	NMR f_a^* (%)	EPR $N_g \times 10^{18}$ spins/g	d_{002}	L_c (nm)	L_a (nm)	X-ray diffraction d_r	N	n	L_a/L_c	HRTM lattice images
Cataclastic structure coal	QN03	1.23	0.33		75.0365	0.3667	1.3755	1.8575	0.568	3.75	51.9	1.35	
Mortar structure coal	TY02	1.51	0.32		44.8972	0.3662	1.4903	1.8546	0.535	4.07	51.8	1.24	
Mortar structure coal	XY02	1.99	0.44		45.5163	0.3699	1.2997	1.8444	0.561	3.51	51.2	1.42	
Granulitic structure coal	XY01	1.26	0.31		33.8709	0.3665	1.4379	1.9163	0.540	3.92	55.3	1.33	
Mealy structure coal	ZXZ06	1.33	0.28		65.7937	0.3561	1.5138	2.0003	0.528	4.25	60.3	1.32	Dispersedly isolated points, with weak orientation
Schistose structure coal	ZJ01	0.95	0.23	0.791	52.4347	0.3828	0.9011	1.7254	0.613	2.35	44.8	1.91	
Thin-layer structure coal	XJ03	1.01	0.23		44.1508	0.3716	0.928	1.8524	0.592	2.50	51.7	2.00	
Scaly structure coal	LL01	1.45	0.43	0.795	64.0869	0.3659	1.3645	1.8997	0.535	3.73	54.3	1.39	
Wrinkle structure coal	BJ05	1.08	0.24	0.778	36.1217	0.3716	1.0827	1.7684	0.57	2.91	47.1	1.63	
Mylonitic structure coal	QN09	0.96	0.27		46.3902	0.3665	1.3782	1.8187	0.536	3.76	49.8	1.32	Gracile worm shape, with local orientation
Mylonitic structure coal	ZXZ04	1.82	0.41	0.809	136.9129	0.3684	1.4733	1.9754	0.558	4.00	53.0	1.27	
Ductile structure coal	XY06	2.21	0.53		77.4869	0.3601	1.6195	2.1802	0.528	4.50	71.6	1.35	
Schistose structure coal	LH05	1.75	0.38	0.802	61.2011	0.3617	1.5694	2.0895	0.538	4.34	65.8	1.33	
Schistose structure coal	TT10	1.94	0.40		41.9478	0.3619	1.566	1.9791	0.534	4.33	59	1.26	
Cataclastic structure coal	HZ01	2.97	0.58	0.873	107.7825	0.3537	2.0291	2.4107	0.485	5.73	87.5	1.19	Striations patch group, with strong orientation
Mylonitic structure coal	HZ07	3.59	0.71	0.925	84.3871	0.3497	2.5172	2.4489	0.467	7.20	90.3	0.97	

* f_a : carbon aromaticity; N : layers of the microcrystal; n : numbers of benzene ring; L_c : size of stacking of basic structural units; L_a : extension of basic structural units; d_{002} : the spacing among monolayers of aromatics; d_r : the aliphatic spacing; N_g : radical density. Broad horizontal lines in the table distinguish TDCs formed in different metamorphic and deformational environments; narrow horizontal lines distinguish tectonically deformed coals of different deformational series. Parts of Table 3 are the same.

TABLE 3: The characteristics of pore structural parameter of TDCs.

Type of tectonically deformed coals	Coal sample's number	Pore volume					Pore specific surface					N ₂ adsorption volume (cm ³ /g)	
		V _t (cm ³ /g)	V ₄ /V _t	V ₅ /V _t	V ₆ /V _t	V ₇ /V _t	S _t (m ² /g)	S ₄ /S _t	S ₅ /S _t	S ₆ /S _t	S ₇ /S _t		
Cataclastic structure coal	QN03	0.00046	95.652	4.348			0.484	83.33	16.667				0.3105
Mortar structure coal	TY02	0.00079	78.481	11.392	7.595	2.532	0.224	30.357	21.429	27.678	20.536		0.9606
Mortar structure coal	XY02	0.00055	92.727	5.455	0	1.818	0.072	59.722	16.667	8.333	15.278		0.3566
Granulitic structure coal	XY01	0.00229	81.223	13.100	5.240	0.437	0.491	43.789	28.31	26.28	1.221		1.9401
Schistose structure coal	ZJ01	0.00066	86.364	7.576	6.060		0.123	46.341	15.447	38.212			3.762
Thin-layer structure coal	XJ03	0.00491	70.061	18.534	11.202	0.203	1.377	25.853	28.467	44.080	1.600		3.7620
Sealy structure coal	LL01	0.00215	82.791	10.698	6.511		0.464	41.164	22.845	35.991			1.9104
Wrinkle structure coal	BJ05	0.00407	76.167	17.700	6.143		1.008	37.401	33.929	28.670			3.1771
Mylonitic structure coal	QN09	0.00930	67.229	19.277	13.253	0.251	2.636	24.772	28.376	45.524	1.328		6.1405
Mylonitic structure coal	ZXZ04	0.01871	74.025	19.401	6.146	0.428	4.575	33.880	34.972	27.607	3.541		12.9174
Ductile structure coal	XY06	0.00078	85.900	5.128	7.692	0.282	0.158	35.443	11.392	48.101	5.064		0.5320
Schistose structure coal	LH05	0.00093	94.624	4.301	1.075		0.101	72.277	12.871	14.852			0.6216
Schistose structure coal	TT10	0.00090	96.667	3.333			0.081	85.185	13.580	1.2350			0.5834
Cataclastic structure coal	HZ01	0.00049	81.663	8.163	10.204		0.105	43.810	10.476	45.714			0.4836
Mylonitic structure coal	HZ07	0.00882	28.458	14.286	54.875	2.381	6.365	5.027	10.008	79.214	5.751		6.9903

V₄: 15 ~ 100 nm; V₅: 5 ~ 15 nm; V₆: 2.5 ~ 5 nm; V₇: <2.5 nm; V_t: Total pore volume; S₄: 15 ~ 100 nm; S₅: 5 ~ 15 nm; S₆: 2.5 ~ 5 nm; S₇: <2.5 nm; S_t: Total specific surface area.

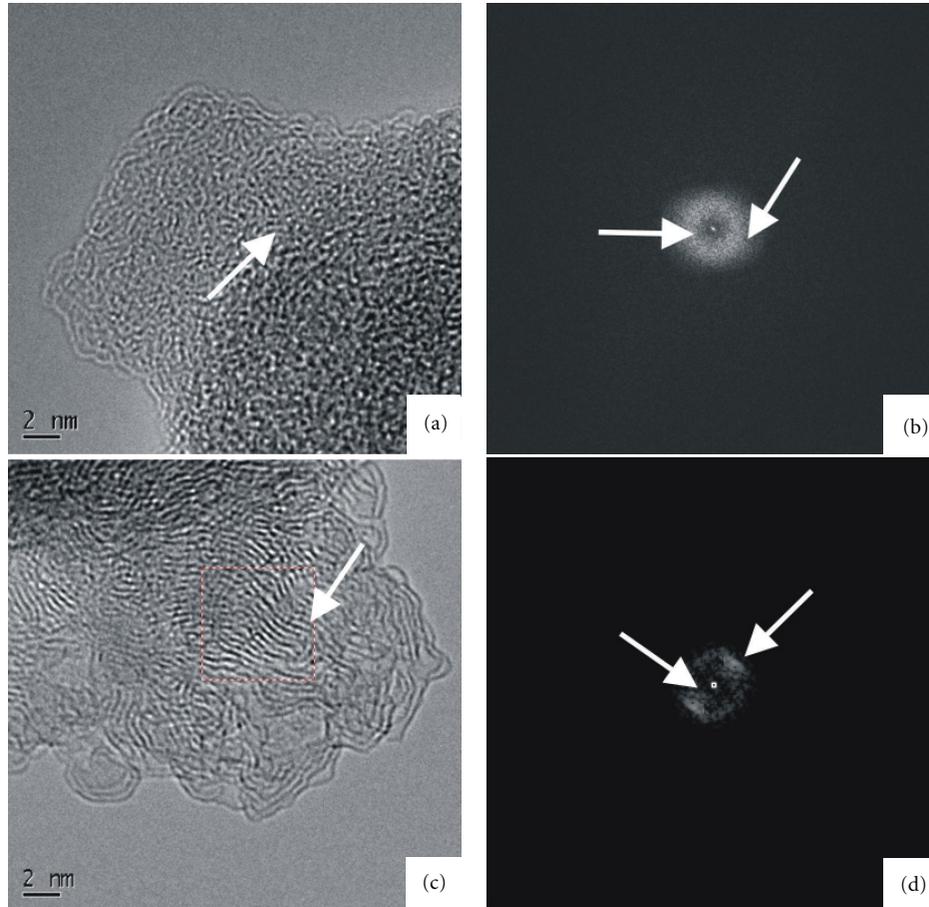


FIGURE 2: HRTEM images of TDCs. (a) Sample ZJ01 of schistose structure coal, in the lattice image; BSU is dispersive and isolated, its diameter is smaller, and BSU has weak orientation, Zhangji Coal Mine, Huainan Coalfield; (b) Sample ZJ01 of schistose structure coal, (002) the (inner ring) diffraction ring lightness is stronger, (101) outer ring looms, Zhangji Coal Mine, Huainan Coalfield; (c) Sample HZ07 of mylonitic structure coal, in the image; BSU is striation group shape, its diameter is large, and BSU has stronger orientation, HRTEM, Haizi Coal Mine, Huaibei Coalfield; (d) Sample HZ07 of mylonitic structure coal, (002) diffraction ring lightness is dispersive, (101) can be seen clearly, and symmetry light spot appears, HRTEM, Haizi Coal Mine, Huaibei Coalfield.

an important role in the evolution of nanoscale pore characteristics, but structural stress is the predominant factor influencing the formation of the characteristics of nanoscale pores.

4.3.2. Permeability of Tectonically Deformed Coals. Often, the TDCs are believed to be low-permeability coals without much value for exploitation [32–34]. In fact, this is not always true. The porosity and permeability of TDCs vary according to changes in their tectonic deformation [19, 22, 35]. In this study, the parameters of permeability in different types of TDCs were tested and the experimental results were shown in Table 4. Those samples were drilled in the coal mine samples collected from the Huaibei and Huainan coalfields. The gas-water phase permeability of TDCs is mainly determined by their type and their sample drilling style. It can be seen clearly from Table 4 that the gas-water phase permeability of brittle deformation increases sequentially. The increase begins with cataclastic structure coal and moves to mortar structure coal, then to schistose

structure coal, which is perpendicular to the fracture, and, finally, to the schistose structure coal, which is parallel to the fracture.

On the other hand, the value of gas-water phase permeability of coal in ductile deformation is less than that in brittle deformation. Moreover, the permeability of TDCs in which ductile deformation is superimposed by later brittle deformation has been significantly increased. Thus, TDCs suffering from certain degrees of deformation have a higher permeability than those of primitive structure coals or very weak TDCs [36].

5. Discussion

5.1. Structural Evolution and Deformational Mechanisms of Tectonically Deformed Coals

5.1.1. Main Geological Factors Affecting Deformation and Metamorphism of Coals. The relationship between coal

TABLE 4: The experiment results of gas-water phase permeability for TDCs.

Deformation series	Type of tectonically deformed coal	Coal sample's number*	Apparent fracture porosity (%)	K_0^{**} of CH_4 ($10^{-3} \mu\text{m}^2$)	K_0 of He ($10^{-3} \mu\text{m}^2$)	K_w ($10^{-3} \mu\text{m}^2$)
Brittle deformation	Cataclastic structure coal	HZ01	4.1	0.162	0.180	0.099
	Cataclastic structure coal	QN03	1.5	0.019	0.020	0.017
	Cataclastic structure coal	TY10	2.4	0.178	0.205	0.075
	Mortar structure coal	TY02	2.6	0.183	0.211	0.087
	Mortar structure coal	XY02	3.1	0.471	0.495	0.138
	Schistose structure coal	ZJ01 (vertical sampling)	2.9	0.534	0.535	0.155
	Schistose structure coal	ZJ01 (lateral sampling)	2.9	1.630	2.120	0.359
Ductile deformation	Wrinkle structure coal	BJ05	2.7	0.362	0.378	0.124
	Ductile structural coal	XY06	2.0	0.089	0.098	0.056

* The direction and degree of fractures affect the drilling coal samples. We made vertical drilling in samples when there is the cross of two sets of fractures (e.g., schistose structure coal, mortar structure coal). And lateral drilling when there is only one set of fracture (Cataclastic structure coal).

** K_0 : absolute permeability; K_w : water phase permeability.

structure and metamorphism has been discussed over a century [37]. It is thought that temperature, pressure, and time are the general geologic factors that control coalification. Pressure factors of coalification include static pressure (confining pressure) and dynamic pressure (tectonic stress). Previous studies focused on the influence of temperature and confining pressure on coal structure. As a result, the existed studies have indicated that temperature is the main factor causing the changes in the chemical structure of coal, facilitating metamorphism and improving coal rank [37, 38]; confining pressure benefits physical coalification but suppresses chemical coalification. Tectonic stress has seldom involved the metamorphism of coals [37, 38]. From BSU testing and the extraction of dissoluble organic molecules of tectonic coals, Cao et al. [4, 39] and Cao et al. [40] concluded that temperature and stress lead to changes in coal macromolecular structure and chemical components. The changes in coal macromolecular structure parameters are mainly caused by compression or shearing. Both brittle and ductile deformation can affect the macromolecular structure of the coal, which leads to dynamic metamorphism and elevation in the coal rank, to a certain degree. The influence of macromolecular structure in ductile deformation coals is stronger than that in brittle deformation coals. The reason is that carbon aromaticity and the degree of ring condensation have been obviously increased in the former. In this research, by using the XRD, HRTEM, EPR, and NMR, L_c , L_a , f_a , and N_g increase, while L_a/L_c , d_{002} , and d_r decrease. The results showed that the cracking and removal of straight and side-chain compounds of organic matter in coal caused reduction in molecular weights and directly changed the chemical composition of macromolecules in coal, especially in low and medium coalification stages. This shows “dynamic hydrocarbon generation”. The change of structural parameters promotes the BSU rearrangement, as well as ordering enlargement and orientation growth. The result is an increase of the aromatic condensed ring system. The stress polycondensation is usually represented by VRA increasing. Therefore, structural stress promotes physical coalification and chemical coalification.

5.1.2. *The Types of Tectonic Stress Affecting Deformation and Metamorphism of Coals.* There has been speculation as to how stress affects the optical properties of coal and whether deformed coal changes chemically when its physical structure changes. Based on the present studies, the influence model of the tectonic stress on the dynamic evolution of organic matter can be concluded as the followings: friction heat view, strain energy view, and mechanochemistry chemical view.

Stone and Cook [41] suggested that anisotropic stress may influent coalification by strain being imparted to vitrinite during coalification. Teichmüller and Teichmüller [42], Bustin [43] and Suchy et al. [44] suggested that local increases in coal rank and local graphitization resulted from frictional heating. Levine and Davis [45] assumed that the applied stress field caused increased bireflectance by the preferred nucleation and growth of favorably oriented lamellae. Based on XRD, TEM, and micro-Raman spectroscopy evaluations, Ju et al. [5, 6] concluded that the coal structure generated under shearing was changed through elongation and contraction of pores between the aromatic lamellae, thus enhancing growth of aromatic stacks. Observation of increased reflectance or local coalification can be classified either by a thermal mechanism [42–44] or mechanical strain mechanism [41].

In the brittle deformational coals, structural influence of different TDCs is not the same. As the deformation becomes stronger, coal macromolecular structures change from a cataclastic structure coal, mortar structure coal, or granulitic structure coal to mealy structure coal. It is similar when going from schistose structure coal to thin-layer structure coal. Under the effect of stress, the structural fractures quickly spread along weak belts of bond forces in coal; the coal body breaks, and the blocks and particles glide, rotate, and displace; friction heat is then produced, which promotes local coal metamorphism. From the observation of XRD, HRTEM and the highest bireflectance in shear zones, stress-chemistry and frictional heating of shearing strain areas might play a more important role in increasing brittle deformation which is because the rate of strain is high enough to destroy bond forces on the stress direction,

generate thermal heat, and locally alter coal structure under stress.

Levine and Davis [45] postulated a mechanical-chemical mechanism in which stress reoriented the lamellae and facilitated growth. Bustin et al. [8] and Ross and Bustin [46], on the other hand, thought strain energy transformation played an significant role in graphitization from coal under the shear stress regime. A previous study of ductile deformation showed that the size of aromatic stacks and reflectance increase in deformed coals [4, 6, 30]. It is obvious that for distinct kinds of ductile deformation coals, the effect of tectonic stress is not the same. There are three main types of changes in macromolecular structures for TDCs: dislocation glide between the aromatic layers of coal cores, shear glide of the aromatic cores, and destruction/recombination of the coal molecular structure bonding energy. The changes of macromolecular structure in ductile deformation become greater than that in brittle deformation. Superimposition of these types of deformations in coal macromolecular structure is, therefore, the basic process of coal structure shearing and also the essential process of strong ductile deformation of coal. These findings are consistent with the hypothesis of aromatic lamellae growing and reorientating proposed by Levine and Davis [45] and the strain energy transformation presented by Bustin et al. [8] and Ross and Bustin [46]. Since deformation was the result of strain effects in coal, the chemical coalification derived from deformation should be attributable to the strain. During deformation of the coal seam, strain might play significant roles which depend upon the strain rate and the geological setting. In strong ductile shearing, reorientation of aromatic lamellae responsible for higher bireflectance or biaxial vitrinite reflectance indicatrices may occur without brittle failure under lower strain rates. In this case, coal would deform ductilely and the aromatic lamellae would become oriented with the new stress regime [4, 5]. A progressive strain may tend to flatten porosity and shorten the distance between lamellae. If the rearrangement of lamellae was slow enough, all of the chemical structural units might conform to the new stress setting with no chemical change. If the coal was pulverized to a fine grain size by ductile shearing, grain flow would parallel the stress direction. During grain flow, strain would be translated to particle surface thus causing thin areas of strain. Changes in the physical and chemical structures of coal might occur on the particle surface, where local changes in coalification might be produced. Strain rearrangement of aromatic lamellae on the particle surface would lead to local coalification [4]. The degree to which strain rearrangement translates to the particle interior may have a profound influence on the gross change in coal structure. Greater strain on the particle surface not only causes reorientation of aromatic lamellae but also may result in some chemical change.

The fact that the highest bireflectance occurs in shear zones, similar to strain area, has been observed from the anthracite specimens deformed at high temperature and pressure [8]. Thus, it may be supposed that strain energy might play a more important role than frictional heating in increasing coal deformation and metamorphism because the

rate of strain is insufficient to generate temperatures high enough to thermally alter coal.

5.2. *Physical Properties of Tectonically Deformed Coals and Their Relation with Structure*

5.2.1. *Porosity of Tectonically Deformed Coals and Their Relation with Structure.* Coal is one of the few highly porous media found in nature. Porosity, pore diameter distribution, and pore specific surface area are the main physical properties of coals. These physical properties are important in coalbed methane exploitation and evaluation of coal and gas outburst tendency [4, 16–22, 47]. The pore volumes and specific surface areas of coals have been widely studied [9, 13, 15, 48–56]. Under a scanning electron microscope and high-resolution transmission electron microscopy, pore shape, pore diameter, the molecular structure and relationship of these features with thermal deformation mechanisms have been identified and discussed [31, 57–59]. Duber and Rouzaud [31] found that the pore structure change determined the carbon microcosmic structure and the bireflectance of coal. The size and shape of the pores depended on the processes of coalification, temperature, and tectonic stress. Coal was considered to be formed with different size and shape of pores and aromatic clusters around the pore wall. Coalification was considered to be a process of aromatic ring concentration with pores changing size and shape. Studies of pore structure are therefore scientifically significant.

Tectonic deformation can not only alter coal's macromolecular structure to distinct degrees but can also change pore structure. Based on measurements of mercury intrusion, researchers compared micropore features of TDCs collected from different coalfields [9–11]. These experiments showed that TDCs possessed with greater medium pore volume than normal coal. No difference in mesopores and micropores was found, however. This suggested that tectonic deformation did not significantly influence micropore structure (less than 10 nm in diameter). Ju et al. [5, 6] suggested that tectonic deformation can not only change the micropore structure (>10 nm) of coal but also influence coal's nanoscale pore structure (<10 nm), which is the main adsorption space for coalbed methane.

From the results listed in Tables 3 and 4, the parameters of pore structure, such as pore diameter structure, pore volume and specific surface area, are seriously influenced by changes in thermal conditions. In contrast, the structural deformation of coal results from variability in pore porosity. Brittle and ductile deformation might change the structural parameters of coal pores, but the structural parameters of different kinds of TDC pores are distinct. It can be seen that the volume and specific surface area of nanoscale pores (<10 nm) in TDCs are more than those in normal coals. This finding indicates that TDCs have a higher potential of gas accumulating and pooling. On the other hand, Zhang et al. [10] and Ju et al. [23] have showed that the volumes and specific surface areas of macro- and medium pores of TDCs

have a similar variation. The macro- and medium pores of TDCs reveal stronger laminar flow.

Thermal and stress action can facilitate coalification and change the macromolecular structure and nanoscale pore structure of TDCs [28, 39], but manner and degree of changes are different. Under thermal conditions, the meso- and micromolecules can be arranged to recombination and condensation by means of physical resultant force. The increase of ordering degree is not quite obvious in orientation, leading to the aromatic layers remaining thin. This commonly enhances the thermal stability of the aromatic structure. Because the aromatic layers are disordered, a more concentrated arrangement results in the formation of mesopores and micropores, forming one or several channels that are connected by pores. Under stressful conditions, the amount of coal macromolecules increases continuously. Additionally, the bordering aromatic rings continue to polymerize and form superpositions. For the local directional and surrounding nondirectional aromatic layers to assemble together, the dislocation of the aromatic cores and slippage of aromatic layers must occur. The larger pores are produced between the aromatic cores and aromatic layers. The pore of TDCs are mainly micropores and submicropores, while ultramicropores are also developed with worse connection. In the local direction process of aromatic structure units, special thin neck, bottle-shaped pores have developed. These are mainly built up by pores between the directional and undirectional structure units.

5.2.2. Permeability of Tectonically Deformed Coals and Their Role in CBM Exploitation. This experimental result shows that the gas-water phase permeability of TDCs is mainly determined by type. The permeability of TDCs could be very high. The permeability increases sequentially, beginning with cataclastic structure coal, to mortar structure coal, to schistose structure coal, which is vertical to the fracture, and, finally, to the schistose structure coal that is parallel to the fracture. TDC with early low ductile deformation superposed by brittle deformation, gas-water phase permeability in the range of $0.1\text{--}5.0 \times 10^{-3} \mu\text{m}^2$, which is in the middle or higher than the primary structure coals and TDCs with weak deformation (Table 4, Figure 3). If the structural fractures are filled with calcite veins or the other mineral matter (Figure 3), gas-water phase permeability would decrease. This means that traditional theory, which states that the permeability in the area where TDCs are common is low, is inaccurate. The permeability of TDCs obtained from the experiment of samples with in situ large diameter in the indoor test can be an important parameter that reflects the real permeability of the coalbed.

When we study the in situ permeability of the coalbed, we should get representative samples of each type of coal in order to obtain integrative permeability. Because of the heterogenous characteristics of the coal seams, especially in the area developing TDCs, we may predict a permeability change of the TDCs according to the types present and the coal gas mining style in different places and seams.

As the stress increases, the coals are further crushed into cataclastic-angular and/or cataclastic-granular pieces. Original cleats [60, 61] that are completely covered with deformation may wrench and tightly compress the fractures, even grinding the coals. Locally intensive deformation may wrench and tightly compress the fractures, even grinding coal into powder, which may plug sheared fractures. However, a combination of the original cleats and tectonic-induced fractures with deformation style (brittle or ductile), size, continuity, and connectivity of the sheared fractures contribute significantly to the overall permeability and are likely to play a major role in the flow of methane through structurally deformed coal in both diffusion at the micropore level and laminar flow at the cleat level [35].

In general, structurally deformed coals are formed through a brittle deformation mechanism and are called cataclastic coals and schistose coals. In such coals which have a hierarchy of open, continuous, and connecting fractures and cleats, the effective block size is not defined at present by the cleat spacing [60, 61], but somewhere between cleats and microfractures. A large number of brittle fractures in structurally deformed coals will make methane migration faster than that in normal coals. This tendency is extremely important for the development of CBM projects and may not only preserve significant gases but also increase the inherent permeability.

Nevertheless, not all structurally deformed coal possesses the properties of fast desorption and laminar flow. Ductile deformation coals, such as mylonitic coals, which are locally and ductilely generated in an intensively compressive and shearing environment, always display tightly compressed and broken fractures to be pushed into one another with worse connectivity for methane migration.

The various types of TDCs formed by the structural deformation of coal seams, especially the coals such as schistose, mortar and cataclastic structure coals subjected to a certain degree of brittle fractures, have good exploration and development potential for coalbed gas. Low ductile deformation superimposed by later brittle deformation also has this potential. On the other hand, changes in coal seam thickness from structural stress and destruction of coal structures caused by the rheology of coal seams are the major factor resulting in the gas outbursts of the coal mines [62, 63]. Therefore, according to the intensity, type and distribution of the rheology of coal seams, the zone of pooling coalbed gas and gas outbursts could be predicted. TDCs of strong brittle-ductile and ductile deformation, such as scale structure coal and mylonitic structure coal, are the main problems in preventing and controlling coal and gas outbursts and gas emissions. These problems need to be resolved urgently.

6. Conclusions

Taking Huaibei and Huainan as typical examples of coal mine areas in the southern North China, the structural characteristics and the physical properties of TDCs have

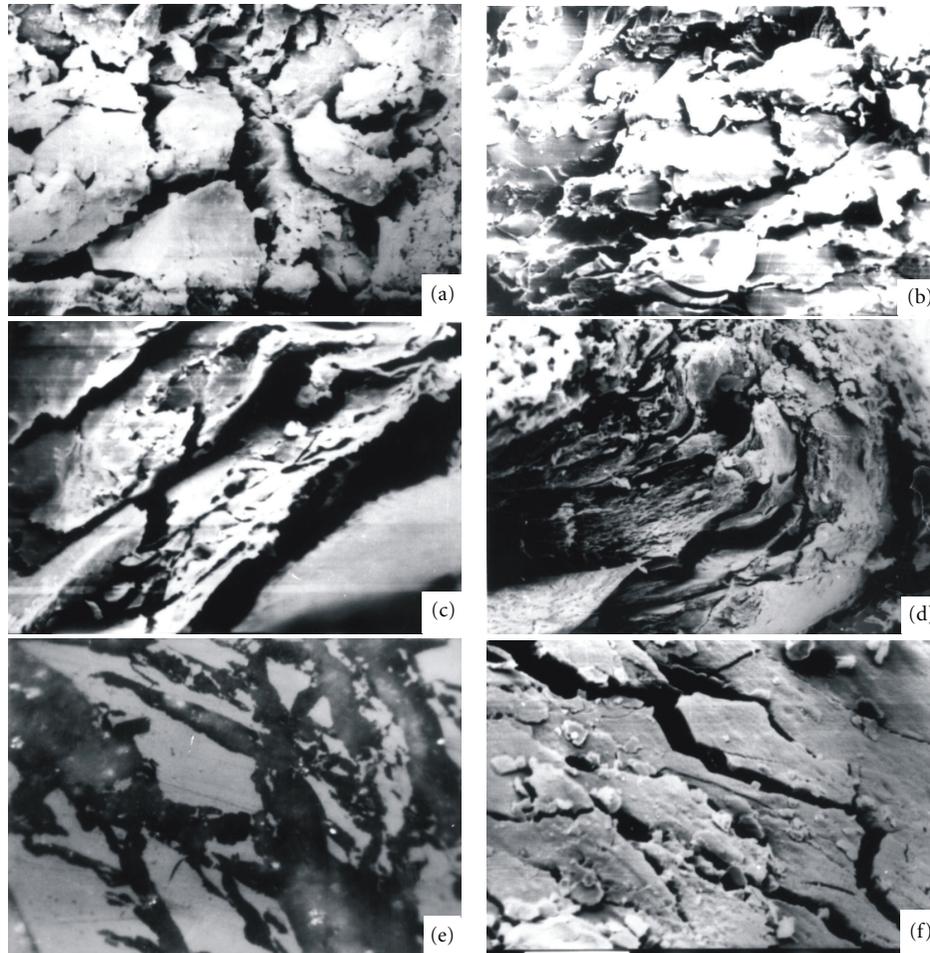


FIGURE 3: SEM and optical microscopy images of TDCs (a) cataclastic structure coal, multidirectional fracture cutting, and no obvious displacement in the blocks, SEM; (b) mortar structure coal, multidirectional fracture cutting, and obvious displacement in the blocks, SEM; (c) schistose structure coal, single directional fracture, and little displacement on its surface, SEM; (d) wrinkle structure coal; coal was wrinkled, and later superimposed by brittle fractures, SEM; (e) cataclastic structure coal with the structural fractures filled with calcite veins, optical microscopy; (f) cataclastic structure coal with the structural fractures expanded the cleats, SEM.

been studied by the authors. Some conclusions are drawn as follows.

(1) Tectonically deformed coals (TDCs) formed by different deformational mechanisms have been divided into three deformation series and ten classes. In the metamorphic-deformed environments, from brittle deformed coal to ductile deformed coal, the changes in characteristics of the macromolecular structures and chemical compositions are that as the increase in structural deformation becomes stronger, from the brittle deformation coal to ductile deformation coal, the ratio of width at the half height of the aromatic carbon and aliphatic carbon peaks (H_{fa}/H_{fal}) was increased. As carbon aromaticity is raised further, carbon aliphaticity reduced obviously and different compositions of macromolecular structure appeared as a jump and wave pattern except that in wrinkle structure coal. Under the directional stress, BSU arrangement is closer, and the orientation becomes stronger from brittle deformed coal to ductile deformed coal. Under the effect of oriented stress, the

orientation of the macromolecular structure becomes locally stronger, and the ordering degree of the arrangement of the BSU is dramatically enhanced.

(2) The characteristics of structural evolution of different kinds of tectonically deformed coals have been further investigated. Structural deformation directly influences the macromolecular structure of coal, which results in dynamic metamorphism. In the brittle deformational coals, frictional heating and stress chemistry of shearing strain areas might play a more important role in increasing brittle deformation because the rate of strain is big enough to generate high temperature, locally altering coal structure under stress. In ductile deformation coals, it may be assumed that strain areas might play a more significant role than frictional heating in increasing coal deformation and metamorphism because the rate of strain is insufficient to generate temperatures high enough to thermally alter coal.

(3) In comparison with the previous results, the characteristics of macromolecular structures and micro- and

nanoscale pore structures of different kinds of TDCs have been analyzed in detail. In particular, advanced research on the formation mechanism of TDCs has been carried out. The nanoscale pore of the cataclastic structure coals caused by the brittle deformation is mainly mesopores; the rest are micropores and submicropores. As the action of tectonic deformation increases, the volume of micropores and the pores whose diameters are even smaller, increases dramatically, and submicropores and ultramicropores can be found. In coals that have experienced ductile deformation, the proportion of mesopores volume in wrinkle structure coal diminishes rapidly, and an increase in the proportion of micropores and submicropores is observed. The proportion of mesopores volume of mylonitic structure coal decreases continually as tectonic stress increases, while the volume of micropores and submicropores increases rapidly, and ultramicropores can also be found. Ductile structure coal has a characteristic similar with the weak brittle deformation coals, but submicropores and ultramicropores are present. The main factor that produces a change in pore structure is strongly tectonic deformation. It is proposed that brittle and ductile deformation can change the pore distribution, which then change spacing and chemical structure.

(4) The permeability of different kinds of TDCs has been investigated which could be very high. The permeability increases sequentially, beginning with cataclastic structure coal, to mortar structure coal, to schistose structure coal, which is perpendicular to the fracture, and, finally, to the schistose structure coal, which is parallel to the fracture. The permeability of TDCs where weak ductile deformation was superimposed by later brittle deformation is better than primary structural coal. According to the results of the coalbed gas testing well, the permeability of TDCs obtained from the experiment of in situ major diameter samples in the indoor test can be an important parameter that reflects the real permeability of the coalbed.

Acknowledgments

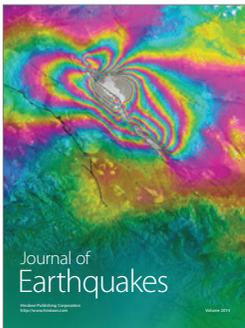
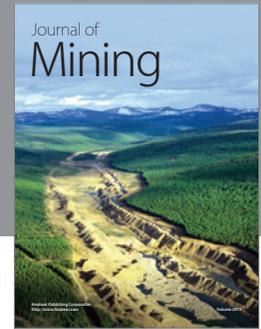
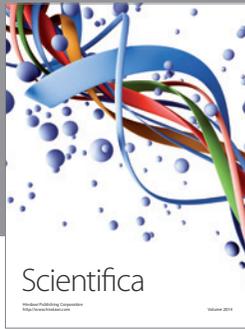
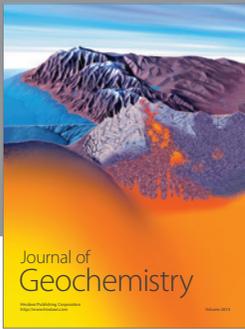
This study was financially supported by the National Basic Research Program of China (also called 973 Program) (Grant no. 2009CB219601), the National Natural Science Foundation of China (Grant no. 40772135; 41030422; 40972131; 40172058), and Strategic Priority Research Program of the Chinese Academy of Sciences (Grant no. XDA05030100).

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