Control of coal facies to adsorption-desorption divergence of coals: A case from the Xiqu Drainage Area, Gujiao CBM Block, North China

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ABSTRACT

Adsorption and desorption of coal to methane are substantially related to coal facies, referring to the production potential of a coalbed methane (CBM) well. Based on analyses of the stratified channel samples from coal seams No. 2, No. 4 and No. 9 in the Xiqu Drainage Area, Gujiao CBM Block, North China, the relationship between coal facies and adsorption-desorption behaviors was first proposed and discussed. Using the coal facies diagrams suggested by previous investigators, four types of coal facies were distinguished in the coal seams, including the upper delta plain wet forest swamp (I), the lower delta plain fen (II), the lower delta plain marsh (III) and the back barrier low moor (IV). The upper delta plain wet forest swamp occurred dominantly, developing at the bottom and top of all the coal seams. Both the ash yield and groundwater index (GWI) tend to increase from coal facies I to IV, whereas the volatile compounds, fixed carbon, organic sulfur, total sulfur, micropore volume and specific surface area, tissue preservation index (TPI) and vegetation index (VI) decreased, which indicates changes in the vegetation types, water supplements and disturbance, acid-base properties and redox conditions in the paleomires. Paleomire evolution accompanied the rise and drop of the sea level, with a transgression or regression process that could be represented by the coal facies sequence. Coal seams No. 2, No. 4 and No. 9 were discriminated into two, three and two evolving stages, respectively, and these stages were characterized by complete cycles, except stage 4-I from the lower delta plain marsh. Mudstone partings in the coal seams deposited during the highest sea level. Collectively, paleomire environments, especially the variation of vegetation types and mire water condition, result in adsorption and desorption divergences among different coal facies. Arborescent plants decrease from coal facies I to IV as the hydrodynamic intensity increases, leading to corresponding changes in botanical tissue preservation, medium oxidizability, mineral content and, ultimately, adsorption and desorption behaviors. In view of the gas content and desorption capacity of the coal layers in the same coal seam, the layers from the upper delta plain wet forest swamp may have the best potential for CBM drainage, followed by those from the lower delta plain fen and marsh, while those from the back barrier low moor may have the worst potential.

Keywords: coal facies; paleomire; adsorption; desorption; Gujiao Block

1. Introduction

As an emerging clean energy with a huge reserve, coalbed methane (CBM) is the unconventional gas resource that is most likely to implement industrial exploitation (Flores, 1998). CBM productivity and recovery efficiency are directly affected by coal's adsorption-desorption behaviors, which are mainly influenced by the substance composition and structure of coal determined by the depositional setting and paleomire type. However, the particular

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44 relevance between the facies and adsorption-desorption behaviors of coal has seldom been researched before now.

45 Coal facies, as the original genetic types of coal formed in given paleomires, have drawn extensive attention since
46 the 1950s (Han et al., 1996). Research on the types of peat accumulation, plant community, depositional conditions,
47 redox potential and geochemistry may provide information about coal facies and its evolution (Dai et al., 2007). Coal
48 petrography, palynology, paleobotany and organic geochemistry are the main methods used to define the coal facies
49 or the depositional environment of peat (Teichmüller, 1989; Nowak and Górecka-Nowak, 1999; Michaelsen and
50 Henderson, 2000; Diessel, 2007; Zdravkov et al., 2011). Many paleodepositional models of coal formation have been
51 established in previous studies (Diessel, 1992; DiMichele and Phillips, 1994). The macerals, microlithotypes and
52 lithotypes of coal can provide evidence about the nature and type of plant community, intensity and duration of
53 humification and the type of depositional milieu (Singh and Singh P.K., 1996). Hence, quantitative relationships
54 between the macerals and microlithotypes have been proposed and applied as useful methods of coal facies study. A
55 ditrigonal coal facies diagram was proposed by Hacquebard and Donaldson (1969) and modified by Marchioni
56 (1980). It has been suggested that the environment of coal deposition is controlled to a large extent by the water depth
57 of the paleomires, which, in turn, controls the extent and nature of the floral development, the mode of plant debris
58 preservation and the formation of different macerals and microlithotypes. Various types of coal facies under the
59 terrestrial, telmatic, limno-telmatic and limnic settings are then identified with quantitative microlithotype parameters.
60 The T-F-D concept, originally developed by Diessel (1982) and later prevailing in coal facies analysis (Marchioni and
61 Kalkreuth, 1991; Obaje and Ligouis, 1996), defines the coal facies in terms of a ternary diagram, in which the end
62 members are also parameters of microlithotypes. As methods of coal facies identification from maceral composition,
63 the GI-TPI diagram (Diessel, 1986) and the GWI-VI diagram (Calder et al., 1991) are the most popular and have
64 been applied to the coal formed under delta setting.

65 Research on the adsorption and desorption mechanism of CBM has been conducted for more than 50 years.
66 Experimental studies and theoretical calculations of the adsorption and desorption behaviors of coal have been
67 conducted with different gases, temperatures, pressures, humidity conditions, coal ranks, petrographic compositions
68 and mathematical models (Daines, 1968; Joubert et al., 1973; Yang and Saunders, 1985; Day et al., 2008). In
69 adsorption studies, single adsorbate, monolayer and polymolecular theories are the most common. The Langmuir
70 equation (Langmuir, 1918) has been applied to the investigations of methane adsorption on coal as a typical
71 monolayer adsorption theory (Zhou and Zhou L., 2009). Even though gas adsorption on coal is not monolayer
72 adsorption, the Langmuir model is still applicable because the adsorption of methane on coal has a type I adsorption
73 isotherm (Wang et al., 2012). The BET equation and D-A equation have been attached in the polymolecular layer and
74 adsorption potential theory, respectively, of methane adsorption on coal. The applicability of various models to
75 methane adsorption on coal and the mechanism interpretation in the light of the model parameters with physical
76 meanings have also been extensively investigated.

77 The Gujiao Block in the Xishan Coalfield, Shanxi Province, North China, is an important commercial CBM
78 production area in China, with estimated CBM resources of approximately 82 billion m³. The Xishan Lanyan CBM
79 Corporation has drilled more than 700 CBM wells in the block since 2010, mainly targeting coal reservoirs No. 2
80 (Shanxi Formation), No. 8 and No. 9 (Taiyuan Formation) (Wang et al., 2015). The adsorption-desorption behaviors
81 of coal have been shown to be critical to the productivity and economic benefit of CBM wells. The coal facies, as a
82 comprehensive index of the matter composition and structure of coal, has significant influence on its
83 adsorption-desorption characteristics and ultimately relates to the CBM drainage. In the present study, we conducted
84 a series of analyses on the stratified channel samples collected from coal reservoirs No. 2, No. 4 and No. 9 in the
85 Xiqu Drainage Area, Gujiao CBM Block. We expected (1) to distinguish the coal facies of the coal samples collected,
86 (2) to describe the features and evolution of the paleomires, (3) to learn about the adsorption-desorption behaviors of
87 coal in the light of some mathematical models and (4) to understand the relation between the coal facies and
88 adsorption-desorption behaviors of the coal in the block.

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2. Geological setting

The Xiqu Drainage Area is located in the northern Gujiao CBM Block, central Shanxi Province, North China (Fig. 1). A series of NE-SW-striking normal faults of various scales occur in the area. The synclines and anticlines are of smaller scale and are parallel or perpendicular to the faults nearby. Coal-bearing strata in the block include the upper Carboniferous Benxi (C_2b) and Taiyuan formations (C_2t) and the lower Permian Shanxi (P_1s) and Xiashihezi formations (P_1x) (Fig. 2A).

Fig. 1. Structure outline and sampling location map of the Xiqu Drainage Area.

The Taiyuan Formation, with thicknesses of 81.5~136.1 m, conformably overlies the Benxi Formation and is mainly composed of sandstone, siltstone, mudstone, limestone and coal seams, in which coal seams No. 8 and No. 9 are two major CBM reservoirs. Coal seam No. 9, located in the lower part of the Taiyuan Formation, has a thickness of 0.52~2.86 m (average 1.65 m) in the Xiqu Drainage Area. The Shanxi Formation, with thicknesses of 23.8~44.9 m, conformably overlies the Taiyuan Formation and is composed of sandstone, siltstone, mudstone and coal seams, in which coal seams No. 2 and No. 4 are another two major CBM reservoirs with thicknesses of 0.5~5.98 m (average 3.52 m) and 0~4.09 m (average 2.12 m), respectively. These major CBM reservoirs were formed in a marine-terrestrial transitional area and the peat mires developed mainly in the delta plain.

3. Samples and methods

3.1. Sampling and preprocessing

A total of 40 stratified channel samples were collected through vertical succession of the coal seams under the mine considering macrolithotypes such as bright coal, semibright coal, semidull coal and dull coal, following the

113 procedures described in Sakorafa and Michailidis (1997), Taylor et al. (1998) and Kalkreuth et al. (2000). After
114 freshening the non-weathered faces of the seams, coal seams No. 2, No. 4 and No. 9 were stratified into 15, 12 and 13
115 continuous parts, respectively, from the floors to the roofs, and every part, including the roofs and floors, was
116 separately sampled (Fig. 2B). Unfortunately, a more detailed division of the coal seams, which would make for a
117 more detailed coal facies study, could not be implemented owing to the limited sampling condition.

118 Coal samples were reduced incrementally and ground to a particle size of approximately 1 mm for polished
119 sections, 0.4 mm for mercury porosimetry, 0.25 mm for isothermal adsorption-desorption experiment and 0.18 mm
120 for proximate analysis and sulfur determination. Rock samples from the roof, floor and partings were prepared for
121 sulfur determination.

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Fig. 2. Columnar sections in the Xiqu Drainage Area of the (A) Permo-Carboniferous coal-bearing strata and (B) samples.

3.2. Micropetrographic analyses

A fraction of the coal samples with 0.6~1.0 mm of granularity was used to prepare the polished sections, following the procedures established in and Taylor et al. (1998).

Vitrinite reflectance was measured with a CCD UVNIR microphotometer equipped with an Axio Imager and an M1m microscope, following the recommendations of Taylor et al. (1998). The examination was conducted using a

131 50× magnification objective lens and a 10× magnification ocular lens under oil immersion ($N_e=1.518$ at 23°C). On
132 each sample at a wavelength of 546 nm, maximum vitrinite (telocollinite) reflectance had fifty readings taken, by
133 which mean maximum vitrinite reflectance was then calculated.

134 During maceral analysis, data of at least 500 points with a fixed step of 0.5 mm between each point were
135 collected for each polished section, according to the method established by Taylor et al. (1998) and ISO 7404-3: 2009
136 (2009). Macerals were identified in white incident light as well as with blue irradiation, with a 50× magnification
137 objective lens and a 10× magnification ocular lens under oil immersion using a Leitz Ortholux II POL-BK
138 microscope.

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140 3.3. Isothermal adsorption-desorption experiments

141 Following the procedures in Yao et al. (2009) and Xu et al. (2012), the Terra Tek ISO-300 was used for the
142 isothermal adsorption-desorption experiments of 24 coal samples. The adsorption experiment was conducted from 0
143 MPa to approximately 10 MPa at a constant temperature of 30°C (close to the actual reservoir temperature) on an
144 air-dry basis. When equilibrium adsorption at the highest set pressure was reached, desorption experiments started at
145 the same constant temperature. The residual gas content at every pressure point was recorded during depressurization
146 until the amount of desorbed gas was less than $1.25 \times 10^{-2} \text{ cm}^3/\text{g}$ per day. Methane with an ultra-high purity (99.999%)
147 was used as the testing gas. Deviation of temperature during the course of the experiments was $\pm 0.5^\circ\text{C}$. The accuracy
148 of the pressure transducer was $\pm 0.1\%$.

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150 3.4. Auxiliary analyses

151 A series of auxiliary analyses were carried out. Moisture content, ash yield, volatile compounds and fixed carbon
152 were determined with proximate analyzer SDTGA 5000. Mercury Porosimeter IV9500 with the pore diameter
153 resolving power of 3 nm was used to measure the pore volume and specific surface area of coal samples. An
154 Intelligence Sulfur Analyzer ZCS-8 was used to determine total sulfur content and occurring forms of sulfur.

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156 4. Results

157 4.1. Macroscopic appearance of the coals

158 The studied coal seams were stratified on the basis of the macrolithotypes including the bright, semibright,
159 semidull and dull coals (Fig. 2B). The bright and semibright coals are banded and have horizontal bedding, while dull
160 coal appears relatively homogeneous and in massive structures, often associated with thin lutaceous interlayers. Coal
161 seam No. 2 is predominantly composed of banded semibright coal, which constitutes 42.15% of the accumulated coal
162 seam thickness, while bright coal is the least occurring type and accounts for merely 9.64%. Bright and semibright
163 coal dominate the upper and lower parts of coal seam No. 2, while the middle part contains mainly duller coal. Coal
164 seam No. 4 is dominated with dull coal, which constitutes 32.62% of the total thickness of the coal seam. Semibright
165 coal comprises only 10.75% of coal seam No. 4 and is the least-found macrolithotype in the seam. The upper and
166 lower parts of coal seam No. 4 are predominantly dull and semidull coals; meanwhile, the middle part is brighter.
167 Coal seam No. 9 is mainly dominated by bright and semibright coals from the floor to the roof, accounting for 33.33%
168 and 30.53%, respectively, of the accumulated coal seam thickness. Dull coal is seen in the middle and upper parts of
169 seam No. 9.

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171 4.2. Microscopic characteristic of the coals

172 Vitrinite maximum reflectance ($R_{o, \max}$) ranges between 1.45% and 1.62% for coal seam No. 2, between 1.56%

173 and 1.70% for seam No. 4 and between 1.79% and 1.95% for seam No. 9 (Table 1), indicating that the former two
174 seams rank coke coal and the last seam ranks meagre coal after the scheme established by Han et al. (1996), or all
175 three seams rank bituminous A after the scheme established by ISO 11760: 2005 (2005). In general, there is a
176 tendency of increasing vitrinite reflectance with the increasing depth of the studied coals.

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178 Table 1. Data of vitrinite reflectance, coal quality and pore analyses of studied samples ($R_{o, \max}$, Proximate analysis and Sulfur analysis
179 in %; V_m in cm^3/g ; and S_m in m^2/g).

Sample	$R_{o, \max}$	Proximate analysis				Sulfur analysis				V_m	S_m
		M_{ad}	A_{ad}	V_{ad}	FC_{ad}	$S_{p, d}$	$S_{s, d}$	$S_{o, d}$	$S_{t, d}$		
2-R	--	--	--	--	--	13.03	0.52	0.82	14.37	--	--
2-1	1.45	0.78	15.70	19.37	64.93	0.30	0.02	0.62	0.94	0.0175	14.462
2-2	1.57	0.83	18.64	21.25	60.12	0.10	0	0.50	0.60	0.0176	14.447
2-3	1.60	0.69	42.77	15.20	42.04	0.03	0	0.23	0.26	0.0073	6.093
2-4	1.55	0.86	24.85	18.62	56.54	0.06	0	0.40	0.46	0.0144	11.834
2-5	1.60	0.77	15.50	17.94	66.58	0.02	0	0.39	0.41	0.0177	14.614
2-6	1.56	0.78	13.61	20.76	65.63	0.03	0	0.48	0.51	0.0204	16.726
2-7	1.58	0.82	11.88	18.97	69.17	0.05	0.01	0.41	0.47	0.0199	16.401
2-8	1.62	0.80	17.26	17.25	65.51	0.26	0.03	0.46	0.75	0.0166	13.751
2-9	1.55	0.80	12.43	19.49	68.09	0.05	0.01	0.44	0.50	0.0206	17.000
2-10	1.55	0.81	8.17	19.72	72.12	0.03	0	0.43	0.46	0.0210	17.262
2-11	1.60	0.81	8.66	19.96	71.39	0.24	0.02	0.54	0.80	0.0220	18.084
2-12	1.55	0.85	8.26	19.80	71.95	0.10	0.01	0.49	0.60	0.0222	18.412
2-13	1.61	0.89	6.37	21.87	71.78	0.06	0.01	0.53	0.60	0.0218	17.870
2-F	--	--	--	--	--	2.37	0.14	0.29	2.80	--	--
4-R	--	--	--	--	--	0.44	0.08	0.06	0.58	--	--
4-1	1.56	0.68	16.09	21.54	62.38	0.08	0.02	0.40	0.50	0.0176	14.483
4-2	--	--	--	--	--	0.03	0	0.03	0.06	--	--
4-3	1.65	0.74	49.36	14.59	36.05	0.08	0.01	0.11	0.20	0.0062	5.188
4-4	1.61	0.72	30.56	15.99	53.46	0.11	0.02	0.31	0.44	0.0096	7.978
4-5	1.61	0.75	23.15	19.82	57.04	0.07	0.01	0.38	0.46	0.0140	11.488
4-6	1.62	0.91	16.02	18.59	65.39	0.10	0.02	0.39	0.51	0.0170	14.016
4-7	1.59	0.84	12.02	23.38	64.60	0.08	0.02	0.52	0.62	0.0178	14.614
4-8	1.63	0.83	11.67	20.32	68.01	0.08	0.02	0.44	0.54	0.0189	15.531
4-9	1.62	1.18	20.07	16.54	63.40	0.05	0.02	0.35	0.42	0.0142	11.754
4-10	1.61	0.79	17.82	18.82	63.37	0.03	0	0.39	0.42	0.0148	12.252
4-11	1.70	0.95	27.65	16.38	55.98	0.05	0.01	0.29	0.35	0.0109	9.057
9-R	--	--	--	--	--	0.82	0.07	0	0.89	--	--
9-FR	--	--	--	--	--	10.96	0.94	0.73	12.63	--	--
9-1	1.95	1.75	8.14	14.72	77.15	0.95	0.14	0.59	1.68	0.0216	17.964
9-2	1.94	1.22	13.75	14.85	71.41	0.06	0.04	0.44	0.54	0.0202	16.684
9-3	1.90	0.87	35.26	13.69	51.06	0.06	0.02	0.20	0.28	0.0110	9.121
9-4	1.91	1.14	20.09	15.72	64.20	0.04	0.01	0.39	0.44	0.0147	12.084
9-5	1.79	1.14	14.33	16.87	68.81	0.20	0.02	0.51	0.73	0.0174	14.386
9-6	--	--	--	--	--	0.03	0	0.07	0.10	--	--
9-7	1.89	0.98	27.13	13.56	59.32	0.03	0.01	0.26	0.30	0.0130	10.864
9-8	1.85	0.97	14.24	15.17	70.60	0.03	0.01	0.38	0.42	0.0177	14.613
9-9	1.91	1.02	11.79	16.29	71.93	0.03	0.02	0.41	0.46	0.0198	16.386
9-10	1.83	1.08	10.79	17.07	72.14	0.22	0.04	0.57	0.83	0.0206	16.887
9-F	--	--	--	--	--	0.02	0.02	0.04	0.08	--	--

180 $R_{o, \max}$, maximum vitrinite reflectance; M_{ad} , moisture content, air-dry basis; A_{ad} , ash yield, air-dry basis; V_{ad} , volatile compounds, air-dry
181 basis; FC_{ad} , fixed carbon, air-dry basis; $S_{p, d}$, pyrite sulfur, dry basis; $S_{s, d}$, sulfate sulfur, dry basis; $S_{o, d}$, organic sulfur, dry basis; $S_{t, d}$, total

182 sulfur, dry basis; V_m , volume of micropore; S_m , specific surface area of micropore; R, coal seam roof; FR, false roof; F, coal seam floor;
 183 --, no data.

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Table 2. Petrographic composition of studied coal samples (vol%).

Sample	Vitrinite group					Inertinite group						Liptinite group			Mineral		
	T	TC	DC	CD	ΣV	F	Sf	Ma	Mi	ID	ΣI	MaS	Cu	ΣL	Cla	Py	ΣM
2-1	8.9	28.2	25.1	2.1	64.3	1.3	16.4	7.9	0.4	8.5	34.6	0.6	0	0.6	0	0.6	0.6
2-2	1.4	15.6	64.8	3.7	85.4	1.6	3.7	2.6	0.2	4.5	12.6	0.6	0.2	0.8	0.8	0.4	1.2
2-3	1.0	7.9	46.2	7.3	62.4	2.3	17.7	8.5	0	6.9	35.5	0	0.4	0.4	0.8	1.0	1.7
2-4	1.7	12.8	68.5	0.4	83.4	1.0	9.3	3.5	0	1.5	15.3	0.2	0	0.2	1.0	0.2	1.2
2-5	5.4	23.1	27.7	1.7	57.9	2.5	23.8	7.0	1.0	6.8	41.1	0.2	0	0.2	0.6	0.2	0.8
2-6	2.1	31.4	44.7	5.5	83.7	1.0	9.3	2.9	1.0	0.6	14.6	0.4	0.2	0.6	0.8	0.4	1.1
2-7	1.4	33.4	38.9	3.9	77.5	1.8	10.4	5.3	0.6	3.7	21.7	0.2	0	0.2	0.2	0.4	0.6
2-8	0.7	15.9	53.6	3.8	74.1	2.2	16.4	3.8	0.2	1.6	24.3	0	0.4	0.4	0.9	0.4	1.3
2-9	2.4	31.5	41.1	3.0	78.0	3.0	9.9	6.2	0.8	1.7	21.6	0.4	0	0.4	0	0	0
2-10	9.5	41.8	19.9	3.0	74.2	2.6	18.6	2.2	0.2	1.3	24.9	0.4	0	0.4	0.2	0.4	0.6
2-11	7.8	32.1	23.7	4.3	67.9	1.8	15.3	7.0	0.6	6.7	31.3	0.4	0	0.4	0	0.4	0.4
2-12	5.8	43.5	24.3	1.3	74.9	1.9	16.9	2.4	0.6	2.0	23.8	0	0.6	0.6	0.4	0.4	0.7
2-13	6.3	36.4	34.3	2.1	79.0	3.0	10.7	2.9	0	3.6	20.2	0.2	0	0.2	0.4	0.2	0.6
4-1	6.5	36.4	39.9	1.3	84.1	0.9	11.1	2.4	0	1.1	15.5	0.4	0	0.4	0	0	0
4-3	0.6	6.4	19.9	7.4	34.3	2.5	23.8	23.2	0	13.8	63.4	0.8	0	0.8	1.2	0.4	1.6
4-4	0.8	21.9	41.6	3.8	68.1	1.0	19.8	6.5	0	3.2	30.4	0.4	0	0.4	0.6	0.6	1.1
4-5	3.7	33.3	43.6	2.7	83.3	1.4	9.9	2.7	0	1.4	15.4	0	0.4	0.4	0.6	0.4	1.0
4-6	2.7	40.6	10.7	4.1	58.1	3.7	27.8	3.7	0	5.4	40.6	0	0.6	0.6	0.4	0.4	0.8
4-7	5.2	36.5	35.5	2.3	79.5	1.4	12.2	4.8	0	1.2	19.5	0.4	0	0.4	0.2	0.4	0.6
4-8	6.4	39.3	20.4	1.5	67.6	1.7	19.7	8.8	0	1.9	32.0	0	0.4	0.4	0	0	0
4-9	1.9	42.7	28.5	5.9	78.9	1.1	7.8	1.7	0	9.3	19.9	0	0.4	0.4	0.6	0.2	0.8
4-10	5.7	31.7	23.2	6.7	67.3	2.6	17.7	5.7	0	5.9	31.9	0.4	0	0.4	0	0.4	0.4
4-11	0.6	21.1	43.0	7.0	71.7	1.1	18.4	4.6	0	3.0	27.2	0.4	0	0.4	0.4	0.4	0.8
9-1	4.6	24.0	24.2	2.6	55.4	2.0	26.8	7.6	0.4	6.5	43.3	0	0	0	0.4	0.9	1.3
9-2	1.4	35.5	32.6	8.0	77.5	1.0	12.3	3.3	0.8	3.7	21.1	0.6	0	0.6	0.6	0.2	0.8
9-3	2.1	6.2	20.6	6.0	35.0	3.5	40.6	16.9	0	1.9	62.9	0.8	0	0.8	0.8	0.6	1.4
9-4	4.8	38.5	26.2	7.1	76.6	0.8	8.2	9.6	1.1	2.9	22.6	0.4	0	0.4	0.4	0	0.4
9-5	4.3	41.3	30.2	7.6	83.3	0.6	7.4	6.2	0	1.7	15.9	0.4	0	0.4	0.4	0	0.4
9-7	1.3	18.3	50.4	4.9	74.9	1.3	14.9	4.5	0.2	2.6	23.6	0	0.4	0.4	0.8	0.4	1.1
9-8	4.0	28.4	20.5	1.7	54.6	1.7	26.4	12.1	0	4.1	44.3	0	0.4	0.4	0.6	0.2	0.8
9-9	4.5	40.6	31.5	5.8	82.4	1.1	8.7	5.8	1.1	0.4	17.1	0	0	0	0.6	0	0.6
9-10	7.4	54.4	26.5	4.6	93.0	1.7	3.5	0.7	0	0.4	6.3	0	0	0	0.4	0.4	0.7

186 T, telinite; TC, telocollinite; DC, desmocollinite; CD, vitrodetrinite; V, vitrinite group; F, fusinite; Sf, semifusinite; Ma, macrinite; Mi,
 187 micrinite; ID, inertodetrinite; I, inertinite group; MaS, macrosporinite; Cu, cutinite; L, liptinite group; Cla, clay mineral; Py, pyrite; M,
 188 mineral matter.

189

190 A total of 11 macerals were counted in all coal samples and the vitrinite group is at an advantage in the macerals
 191 (Table 2). Vitrinite group in coal seam No. 2 accounts for 57.9~85.4 vol% of the overall composition. Comparatively,
 192 vitrinite group macerals vary by a broader margin in other seams, with 34.3~84.1 vol% found in coal seam No. 4 and
 193 35.0~93.0 vol% found in coal seam No. 9. In this group, the predominant macerals are telocollinite and
 194 desmocollinite while telinite and vitrodetrinite are found in small amounts. The inertinite group is less abundant,
 195 accounting for 12.6~41.1 vol%, 15.4~63.4 vol% and 6.3~62.9 vol% in coal seams No. 2, No. 4 and No. 9,
 196 respectively, and is mainly composed of semifusinite. Macrinite occurrence is characterized by the broadest
 197 distribution, ranging between 0.7~23.2 vol%. A small amount of elliptical micrinite is found in the inertinite group
 198 and is often associated with telocollinite. The liptinite group is rare and constitutes less than 0.8 vol% of the overall

composition. Only macrosporinite and cutinite are observed. The highest content of visible mineral matter is 1.7 vol%, and only clay mineral and pyrite are found. Pyrite occurring as small, euhedral crystals or in framboidal form is often associated with desmocollinite. This type of pyrite is usually interpreted as syngenetic pyrite during the peat formation (Taylor et al., 1998). Epigenetic pyrite is present as filling of open cell lumens in some macerals such as telocollinite and semifusinite.

4.3. Isothermal adsorption-desorption characteristics

Adsorption and desorption experiments on 24 coal samples were conducted successively at a constant temperature of 30°C. The results are listed in Table 3, Table 4 and Fig. 3. The maximum adsorbed gas content ($V_{\max}$) was measured when equilibrium adsorption was reached at the highest set pressure of approximately 10 MPa. $V_{\max}$ of coal seams No. 2, No. 4 and No. 9 range between 7.34~19.35 cm³/g (mean 15.16 cm³/g), 6.86~17.00 cm³/g (mean 11.47 cm³/g) and 10.41~20.86 cm³/g (mean 16.75 cm³/g), respectively.

Table 3. Isothermal adsorption data of studied coal samples (p in MPa and V_{ad} in cm³/g).

2-1		2-2		2-3		2-5		2-7		2-10		2-12		2-13	
p	V_{ad}	p	V_{ad}	p	V_{ad}	p	V_{ad}	p	V_{ad}	p	V_{ad}	p	V_{ad}	p	V_{ad}
0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
0.56	4.74	0.68	3.32	0.71	1.45	0.66	3.53	0.67	3.14	0.53	4.79	0.56	3.90	0.58	4.11
2.40	12.03	2.67	8.24	2.89	4.07	2.64	8.80	2.64	8.79	2.37	12.50	2.47	10.29	2.27	10.55
4.48	15.04	4.78	10.24	5.03	5.31	4.75	11.17	4.61	11.40	4.52	15.67	4.88	13.31	4.89	14.39
6.57	17.23	6.65	11.11	6.86	6.25	6.58	12.56	6.52	13.15	6.52	17.43	6.60	14.63	6.65	16.02
8.38	18.34	8.30	11.55	8.12	6.66	8.25	13.33	8.35	14.34	8.28	18.38	8.06	15.39	8.19	17.00
10.33	19.35	10.24	11.92	10.67	7.34	10.33	13.87	10.35	15.33	10.32	19.25	10.39	16.37	10.40	17.85
4-3		4-4		4-5		4-6		4-7		4-9		4-11		9-1	
p	V_{ad}	p	V_{ad}	p	V_{ad}	p	V_{ad}	p	V_{ad}	p	V_{ad}	p	V_{ad}	p	V_{ad}
0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
0.84	1.93	0.81	1.55	0.69	2.60	0.58	3.80	0.59	3.42	0.75	2.43	0.72	2.21	0.56	4.24
2.88	4.22	2.91	4.27	2.75	6.75	2.67	9.99	2.64	9.37	2.82	6.36	2.94	6.07	2.33	11.35
4.94	5.20	4.97	5.64	4.88	8.65	4.66	13.17	4.63	11.73	4.81	8.08	4.80	7.81	4.86	14.84
6.83	6.16	6.57	6.78	6.66	9.85	6.51	14.92	6.52	13.20	6.45	9.42	6.72	9.61	6.57	16.31
8.54	6.58	7.98	7.33	8.54	10.57	8.11	16.05	8.07	13.94	8.36	10.22	8.25	10.64	8.30	17.30
10.49	6.86	10.56	8.11	10.59	11.31	11.18	17.00	10.33	14.51	10.36	10.80	11.26	11.72	10.31	18.32
9-2		9-3		9-4		9-5		9-7		9-8		9-9		9-10	
p	V_{ad}	p	V_{ad}	p	V_{ad}	p	V_{ad}	p	V_{ad}	p	V_{ad}	p	V_{ad}	p	V_{ad}
0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
0.56	3.05	0.68	2.88	0.65	3.20	0.62	3.56	0.72	3.08	0.43	4.08	0.50	4.67	0.61	4.40
2.54	9.46	2.70	6.89	2.63	8.95	2.54	10.12	2.70	7.68	2.29	13.00	2.43	13.32	2.51	12.35
4.67	12.29	4.84	8.48	4.62	11.42	4.81	13.47	4.97	10.39	4.27	16.20	4.60	16.56	4.65	15.73
6.69	13.96	6.66	9.39	6.61	12.91	6.64	15.21	6.38	11.82	6.40	18.52	6.64	18.86	6.66	17.78
8.02	14.60	8.30	9.86	8.36	13.87	8.29	16.42	7.94	12.82	8.25	19.68	8.17	19.73	8.47	18.82
10.70	15.72	10.6	10.41	10.33	14.83	10.40	17.20	10.54	13.70	10.37	20.86	11.18	20.28	10.64	19.39

p , pressure; V_{ad} , adsorbed gas content.

For a better understanding of the isothermal adsorption behavior, Langmuir equation (1) (Langmuir, 1918), BET equation with two constants (2) and D-A equation (3), listed below, were used as representative adsorption models of monolayer, polymolecular layer and adsorption potential theory, respectively.

$$V = V_L \frac{p}{p_L + p} \quad (1)$$

218 where V is the gas content within the sample, cm^3/g ; V_L is Langmuir volume, cm^3/g ; p is pressure, MPa; and p_L is
 219 Langmuir pressure, MPa.

$$V = \frac{V_m C p}{(p^0 - p)[1 + (C - 1) \frac{p}{p^0}]} \quad (2)$$

220 where V_m is the maximum monolayer gas content within the sample, cm^3/g ; C is constant relevant to adsorption heat;
 221 and p^0 is saturated vapor pressure, MPa.

$$V = V_0 \exp \left[-D \ln^n \left(\frac{p^0}{p} \right) \right] \quad (3)$$

222 where V_0 is the volume of micropore, cm^3/g ; D is constant relevant to adsorption heat; and n is constant relevant to
 223 pore distribution.

224

225

Table 4. Isothermal desorption data of studied coal samples (p in MPa and $V_{r, ad}$ in cm^3/g).

2-1		2-2		2-3		2-5		2-7		2-10		2-12		2-13	
p	$V_{r, ad}$	p	$V_{r, ad}$	p	$V_{r, ad}$	p	$V_{r, ad}$	p	$V_{r, ad}$	p	$V_{r, ad}$	p	$V_{r, ad}$	p	$V_{r, ad}$
8.44	19.24	8.04	11.81	8.31	7.21	7.87	13.76	8.02	14.96	8.56	19.28	8.21	15.99	7.59	17.53
6.99	18.38	6.76	11.59	6.98	6.76	7.03	13.25	6.60	13.86	7.10	18.53	7.08	15.30	6.93	17.03
5.55	17.40	5.57	11.16	5.87	6.38	6.11	12.77	5.51	13.03	5.70	17.65	5.95	14.53	6.12	16.46
5.08	17.02	4.67	10.63	4.94	5.95	4.88	11.91	4.42	12.03	5.07	17.15	5.25	13.97	5.01	15.47
3.77	15.84	3.63	9.85	3.96	5.47	3.80	11.01	3.45	10.97	3.82	16.01	3.71	12.49	3.67	13.99
2.76	14.53	2.66	8.84	2.83	4.80	2.84	10.01	2.51	9.73	2.84	14.75	2.40	10.69	2.83	12.78
1.93	13.11	1.72	7.37	1.88	4.06	1.95	8.74	1.63	8.07	2.01	13.33	1.65	9.10	2.00	11.24
1.22	11.24	1.09	5.92	1.25	3.51	1.18	7.24	1.06	6.66	1.31	11.51	1.30	8.22	1.27	9.43
0.82	9.74	0.71	4.62	0.69	2.66	0.77	5.97	0.64	5.02	0.90	10.02	0.88	6.78	0.86	7.91
4-3		4-4		4-5		4-6		4-7		4-9		4-11		9-1	
p	$V_{r, ad}$	p	$V_{r, ad}$	p	$V_{r, ad}$	p	$V_{r, ad}$	p	$V_{r, ad}$	p	$V_{r, ad}$	p	$V_{r, ad}$	p	$V_{r, ad}$
8.61	6.82	8.11	8.10	8.29	10.90	8.43	16.67	7.81	14.37	8.06	10.73	8.43	11.53	8.20	17.98
7.05	6.52	7.04	7.75	7.14	10.35	6.64	15.62	7.02	13.94	7.17	10.32	6.75	10.88	7.13	17.30
5.75	6.21	5.92	7.38	6.03	9.80	5.45	14.74	6.16	13.44	6.23	9.95	5.48	10.28	6.07	16.57
5.07	6.03	4.86	6.88	5.29	9.33	4.51	13.99	4.88	12.50	5.05	9.27	4.57	9.76	5.37	16.00
3.75	5.67	3.85	6.36	3.84	8.34	3.52	13.13	3.78	11.51	3.74	8.45	3.56	9.23	3.75	14.38
2.72	5.21	2.83	5.72	2.59	7.25	2.42	11.71	2.57	10.12	2.83	7.75	2.59	8.41	2.57	12.70
1.86	4.76	1.88	4.93	1.57	5.86	1.74	10.61	1.86	8.89	1.96	6.77	1.78	7.67	1.61	10.62
1.11	4.03	1.19	4.27	1.19	5.22	1.06	8.85	1.17	7.47	1.17	5.75	1.05	6.26	1.15	9.27
0.69	3.52	0.62	3.29	0.92	4.52	0.67	7.48	0.79	6.21	0.76	4.75	0.63	5.38	0.82	7.88
9-2		9-3		9-4		9-5		9-7		9-8		9-9		9-10	
p	$V_{r, ad}$	p	$V_{r, ad}$	p	$V_{r, ad}$	p	$V_{r, ad}$	p	$V_{r, ad}$	p	$V_{r, ad}$	p	$V_{r, ad}$	p	$V_{r, ad}$
8.12	15.69	8.20	10.20	7.87	14.39	7.79	17.08	8.03	13.58	8.20	20.82	8.49	20.14	8.61	19.06
6.80	14.90	7.03	9.71	6.56	13.50	6.45	15.90	6.81	13.20	6.95	20.05	6.45	18.83	7.39	18.39
5.55	14.16	5.84	9.26	5.54	12.83	5.25	14.87	5.67	12.64	5.56	19.09	5.03	17.60	6.08	17.53
4.56	13.35	5.18	8.93	4.44	11.91	4.38	13.92	4.60	12.14	5.06	18.68	4.31	16.86	5.18	16.91
3.55	12.30	3.93	8.24	3.38	10.82	3.33	12.60	3.43	11.20	3.77	17.47	3.43	15.88	4.20	15.95
2.55	10.91	2.68	7.31	2.49	9.64	2.41	11.15	2.50	10.13	2.86	16.25	2.42	14.23	2.94	14.89
1.64	9.20	1.67	6.13	1.63	8.00	1.56	9.21	1.58	8.80	2.03	14.84	1.77	12.88	1.98	13.19
1.08	7.65	1.08	5.17	1.07	6.57	0.91	7.23	1.00	7.73	1.32	12.98	1.10	10.73	1.38	11.52
0.66	5.91	0.70	4.19	0.66	4.94	0.58	5.57	0.59	6.48	0.90	11.46	0.73	9.03	0.92	9.70

226 p , pressure; $V_{r, ad}$, residual gas content.

227

228

Fig. 3. Adsorption and desorption isotherms of studied coal samples. Note: ad., adsorption; de., desorption.

Matlab R2010b was used and parameters of equations (1), (2) and (3) were obtained using nonlinear fitting solvers at least squares principle. Convergence cannot be obtained for equation (2). Valid parameters for equations (1) and (3) are listed in Table 5.

The Langmuir equation can also be used to evaluate desorption experiments and V_L and p_L for the desorption experiments are marked as $V_{L,D}$ and $p_{L,D}$ in present study (Table 5). Depletion gas content (V_d) was calculated using the assumed depletion pressure of 0.7 MPa, an empirical value of the lowest reservoir pressure obtained during CBM drainage in America (Fu et al., 2007). Eventually, desorption rate (R_D) was worked out while the desorbed gas content (V_D) was divided by the maximum adsorbed gas content (V_{max}) (Table 5).

Both the V_L and V_0 correlate positively with V_{max} (Fig. 4A), indicating the ultimate adsorption capacity of coal seam No. 9 is the highest and that of coal seam No. 4 is the lowest. Similarly, the V_d positively correlates with V_{max} (Fig. 4A), meaning the higher the adsorbed gas content is in the adsorption, the higher the residual gas content will be after desorption. $V_{L,D}$ is directly proportional to and consistently lower than V_L (Fig. 4B). Positive correlations also occur between p_L and D (Fig. 4C) and between R_D and $p_{L,D}$ (Fig. 4D), while p_L and n are negatively correlated (Fig. 4C).

Table 5. Parameters of isothermal adsorption and desorption models of studied coal samples (V_L , V_0 , $V_{L,D}$, V_d and V_D in cm^3/g ; p_L and $p_{L,D}$ in MPa; and R_D in %).

Sample	Adsorption					Desorption				
	V_L	p_L	V_0	D	n	$V_{L,D}$	$p_{L,D}$	V_d	V_D	R_D
2-1	23.47	2.32	19.33	0.25	1.53	21.50	1.20	7.92	11.43	59.06
2-2	14.53	2.13	11.88	0.18	1.87	14.11	1.53	4.43	7.49	62.84
2-3	10.34	4.49	7.43	0.40	1.34	8.63	1.99	2.25	5.09	69.42
2-5	17.43	2.60	13.96	0.25	1.60	16.12	1.57	4.97	8.90	64.17
2-7	20.77	3.73	15.39	0.33	1.48	18.00	1.97	4.72	10.61	69.21
2-10	22.93	2.03	19.14	0.21	1.67	21.69	1.23	7.87	11.38	59.14

2-12	19.94	2.35	16.30	0.24	1.58	19.12	1.80	5.35	11.02	67.30
2-13	22.24	2.57	17.87	0.25	1.58	20.95	1.65	6.24	11.61	65.05
4-3	8.98	3.23	6.99	0.33	1.40	7.44	1.02	3.03	3.83	55.88
4-4	12.54	5.75	8.27	0.44	1.34	9.38	1.56	2.91	5.20	64.17
4-5	14.70	3.27	11.31	0.31	1.46	13.12	1.95	3.47	7.84	69.35
4-6	21.39	2.84	17.10	0.30	1.47	18.66	1.30	6.53	10.47	61.59
4-7	18.12	2.49	14.58	0.24	1.65	17.02	1.61	5.16	9.35	64.45
4-9	14.85	3.84	10.95	0.34	1.47	12.43	1.52	3.92	6.88	63.72
4-11	17.03	5.18	11.85	0.45	1.27	12.74	1.17	4.77	6.95	59.29
9-1	22.38	2.38	18.15	0.23	1.63	20.97	1.55	6.52	11.80	64.39
9-2	20.19	3.02	15.60	0.27	1.61	18.44	1.62	5.56	10.16	64.61
9-3	12.64	2.31	10.35	0.23	1.62	11.74	1.48	3.77	6.64	63.78
9-4	19.31	3.20	14.75	0.28	1.56	17.39	1.88	4.72	10.11	68.19
9-5	22.70	3.26	17.29	0.29	1.56	20.42	1.84	5.63	11.57	67.28
9-7	18.65	3.76	13.94	0.35	1.43	15.11	1.05	6.04	7.66	55.87
9-8	25.17	2.45	20.66	0.22	1.63	23.19	1.11	8.97	11.89	57.00
9-9	24.25	2.03	20.32	0.21	1.71	22.80	1.35	7.79	12.49	61.61
9-10	24.39	2.58	19.38	0.22	1.74	21.41	1.25	7.69	11.70	60.36

V_L , Langmuir volume; p_L , Langmuir pressure; V_0 , volume of micropore; D , constant relevant to adsorption heat; n , constant relevant to pore distribution; $V_{L,D}$, Langmuir volume in desorption; $p_{L,D}$, Langmuir pressure in desorption; V_d , depletion gas content; V_D , desorbed gas content; R_D , desorption rate.

Fig. 4. Relationship between the isothermal adsorption and desorption parameters of studied coal samples.

4.4. Proximate, micropore and sulfur analyses

A series of auxiliary analyses were conducted to measure the moisture content, ash yield, volatile compounds, fixed carbon, pore volume and specific surface area and sulfur of the samples (Table 1). As the micropore plays a

259 decisive role in gas adsorption in coal (Fu et al., 2007), pore with diameter less than 10 μm , which was classified as
 260 the micropore by XOQOT (1966), was particularly taken into consideration in this study (Table 1). Ash yield obtained
 261 from proximate analysis correlates inversely with fixed carbon, micropore volume (V_m) and specific surface area (S_m)
 262 at determination coefficients (R^2) all higher than 0.92 (Fig. 5A and Fig. 5B). Likewise, the coefficient between ash
 263 yield and organic sulfur is 0.73 (Fig. 5A). Micropore volume (V_m) correlates strongly with specific surface area (S_m)
 264 at a determination coefficient of nearly 1.00 (Fig. 5C). Simultaneously, these two pore parameters positively correlate
 265 with fixed carbon and organic sulfur at determination coefficients higher than 0.69 (Fig. 5C and Fig. 5D).
 266

Fig. 5. Relationship between the coal quality and pore parameters of studied coal samples.

270 5. Discussion

271 5.1. Types of coal facies

272 5.1.1. Terminology and methodology of paleomire

273 Peat-forming environments are commonly described using several terms, such as swamp, marsh, mire, moor and
 274 bog. Moore (1987) proposed the term 'mire' as the most general and defined major categories of peat-forming
 275 systems based largely on hydrology, especially the source of water and ion composition and concentration. In general,
 276 rheotrophic mires are recharged both from groundwater and rainfall. Ombrotrophic mires, in contrast, are solely
 277 rain-fed. Moore (1987) adopted the term bog to describe the ombrotrophic type of mire while dividing rheotrophic
 278 mires into fens and swamps on the basis of flooding range with respect to the mire surface. Marshes are another type
 279 of rheotrophic mire in which herbaceous plants prevail and peat accumulation is minimal. In this paper, the terms
 280 'mire' and 'paleomire' are used as collective descriptions of peat-forming environments in the Xiqu Drainage Area.
 281 Specific coal facies are denominated in the light of petrographic indices used as indicators of plant material and
 282 depositional conditions to define particular paleopeat-forming environments.

283 Numerous schemes have been proposed in the past to enumerate the depositional environments of coal based on
 284 the quantitative relationships among the tissue and non-tissue derived constituents. [Diessel \(1986\)](#) established a
 285 correlation between the coal facies indicators and corresponding environments by both the Gelification Index (GI)
 286 and Tissue Preservation Index (TPI) for Permian coals in the Sydney Basin. This scheme was then adopted widely
 287 and prevailed in facies studies of coals from different areas and ages ([Kalkreuth and Leckie, 1989](#); [Strehlau, 1990](#);
 288 [Kalkreuth et al., 1991](#); [Lamberson et al., 1991](#); [Obaje et al., 1994](#); [Kalkreuth et al., 2000](#); [Dai et al., 2007](#)).
 289 Modifications have also been made to apply these two indices in low rank coal ([Singh and Singh P.K., 1996](#); [Sakorafa](#)
 290 [and Michailidis, 1997](#); [Gmur and Kwiecińska, 2002](#); [Bechtel et al., 2003](#); [Amijaya and Littke, 2005](#)).

291 The TPI is a measure of the relationship between the plant tissue and groundmass components in coal and the
 292 degree of humification, emphasizing the degree of plant cell tissue preservation versus destruction. Meanwhile, the
 293 TPI represents the effects of the input of woody material and its preservation prior to final deposition. Increased TPI
 294 values are thought to indicate a dominance of arborescent plants in the primary paleomire. The GI is the ratio of
 295 gelified and fusinitized macerals and is considered to reflect the height of the water table or the continuity in moisture
 296 availability during peat accumulation. Wet conditions of peat formation are distinguished by high GI values. The GI
 297 is also regarded as an oxidation index in the mire; a decrease in GI values implies an increase in oxidation. In this
 298 study, the TPI and GI were calculated by the formulae (modified from [Diessel, 1986](#)) given below:

$$299 \quad TPI = \frac{\text{Telinite} + \text{Telocollinite} + \text{Fusinite} + \text{Semifusinite}}{\text{Desmocollinite} + \text{Vitrodetrinite} + \text{Gelocollinite} + \text{Corpocollinite} + \text{Inertodetrinite} + \text{Macrinite}}$$

$$301 \quad GI = \frac{\text{Vitrinite} + \text{Macrinite}}{\text{Fusinite} + \text{Semifusinite} + \text{Inertodetrinite}}$$

302 Another prevalent method for evaluating coal's depositional environment was proposed by [Calder et al. \(1991\)](#)
 303 for Westphalian coal of Nova Scotia. A paleomire environment diagram based on a Groundwater Index (GWI) and a
 304 Vegetation Index (VI) were suggested. The GWI evaluates the intensity of rheotrophic conditions as a ratio of
 305 strongly gelified vitrinite macerals and mineral matter contents over the weakly gelified vitrinite macerals. High GWI
 306 values indicate a relatively high water table in the precursor mires and vice versa. The VI is the ratio of the macerals
 307 with remnant botanical cell structure to the matrix, detrital and granular macerals such as desmocollinite,
 308 vitrodetrinite, inertodetrinite, sporinite, cutinite and liptodetrinite. In other words, the VI represents the vegetation
 309 type by contrasting the macerals of forest affinity with those of herbaceous and marginal aquatic affinity. High VI
 310 values are indicative of forested mires, whereas low VI values suggest a deposition in a limnic environment and
 311 mainly herbaceous source material. These two measures in this study were calculated following the formulae
 312 (modified from [Calder et al., 1991](#)) below:

$$313 \quad GWI = \frac{\text{Vitrodetrinite} + \text{Gelocollinite} + \text{Corpocollinite} + \text{Clay Mineral} + \text{Quartz}}{\text{Telinite} + \text{Telocollinite} + \text{Desmocollinite}}$$

$$314 \quad)$$

$$315 \quad VI = \frac{\text{Telinite} + \text{Telocollinite} + \text{Corpocollinite} + \text{Fusinite} + \text{Semifusinite} + \text{Resinite} + \text{Suberinite}}{\text{Desmocollinite} + \text{Vitrodetrinite} + \text{Inertodetrinite} + \text{Liptodetrinite} + \text{Sporinite} + \text{Cutinite}}$$

317 The coal-bearing strata in the Gujiao were typically deposited in a delta plain-littoral transitional area and coal
 318 seams No. 2, No. 4 and No. 9 were principally formed under the setting of a delta. In the facies analysis of the coal
 319 seams studied, the methods based on maceral composition of coals established by [Diessel \(1986\)](#) and [Calder et al. \(1991\)](#)
 320 were applied. Facies diagrams of both the GI-TPI and GWI-VI show applicability to a certain extent in these
 321 coals.
 322

Fig. 6. GI-TPI diagram for coal facies types of studied coal samples (base map modified from Diessel, 1986).

5.1.2. Characteristics of coal facies

Based on the GI-TPI diagram, the coal samples examined in this study were classified into four types of coal facies (Fig. 6) and similar divisions were subsequently found in the GWI-VI diagram, even though all these types were in the ombrotrophic section (Fig. 7). These four types of coal facies are denominated as the upper delta plain wet forest swamp (I), the lower delta plain fen (II), the lower delta plain marsh (III) and the back barrier low moor (IV).

The upper delta plain wet forest swamp is the mire occupying an area of phytogenic sedimentation on the delta furthest to the sea with dominant arborescent, in fact, huge perennial plants, where both telinite and telocollinite are richer than the other three types. The coals were formed under the wet condition of peat formation, and botanical structures were weakly destructed. The relatively wide ranges of the TPI and VI imply a certain variation in the plant species composition, although arborescent plants were always the prime species.

The lower delta plain fen occurs as an intermediate stage between herbaceous and forest swamp (Diessel, 1992), and a mixture of herbaceous arborescent assembly of plants supplied the peat formation under wet condition. The lower delta plain marsh is dominated by herbaceous plants growing in the vicinity of seawater. Arborescent plants were rare in this area and botanical tissues were strongly decomposed. The macerals are predominantly composed of desmocollinite of 41.6~68.5 vol%.

The back barrier low moor is frequently influenced by seawater. The paleomire here was salinized to some extent and botanical tissues were decomposed intensely. The detrital macerals, clay mineral and pyrite in this coal facies were the richest among the four types.

Divergence and regularity among these four types of coal facies are obvious. The pyritic sulfur, sulfate sulfur, moisture content and GI are variable among different coal facies with no consistent trend in the studied coal seams.

347 By contrast, the ash yield, **volatile compounds**, fixed carbon, organic sulfur, total sulfur, micropore volume and
 348 specific surface area, TPI, VI and GWI vary regularly among the coal facies. In general, both the ash yield and GWI
 349 tend to increase from coal facies I to IV in each coal seam, whereas the **volatile compounds**, fixed carbon, organic
 350 sulfur, total sulfur, micropore volume and specific surface area, TPI and VI decrease (Fig. 8).
 351

352
 353 Fig. 7. GWI-VI diagram for coal facies types of studied coal samples (base map modified from [Calder et al., 1991](#)).
 354

355 The paleomire environments are responsible for these variations among the coal facies. In the upper delta plain
 356 wet forest swamp, huge perennial plants prevailed in humid condition, providing abundant precursor material for coal.
 357 Meanwhile, the rapid burial of paleopeat ensured the preservation of the botanical cell structure of arborescent plants.
 358 The peat accumulated in peaceful mire water with distributary channels and crevasses seldom affected. This moist
 359 and reductive condition without frequent disturbance contributed to the preservation of organic sulfur and less
 360 extraneous mineral matter in the peat. Therefore, the ash yield and GWI in the upper delta plain wet forest swamp are
 361 low, while the **volatile compounds**, fixed carbon, organic sulfur, total sulfur, micropore volume and specific surface
 362 area, TPI and VI are high (Fig. 8).
 363

Fig. 8. Regularization parameters of coal facies of studied coal seams.

The lower delta plain fen occurs in a lower transitional area that is closer to the sea. Both herbaceous and arborescent plants supplied the peat formation under wet condition (Diessel, 1992). The botanical tissues of the herbaceous plants are more easily decomposed and made less contribution to the micropore. Occasional crevasses and overflows increased the extraneous mineral matter in the paleomire. Reductive condition for organic sulfur preservation was also affected by the intrusion of extraneous flows.

Herbaceous plants prevailed in the lower delta plain marsh and botanical tissues were strongly decomposed. More frequent extraneous flows from the distributary channels and sea brought more mineral matter to the paleomire, leading to higher ash yield and lower organic sulfur remaining (Fig. 8). In the back barrier low moor, high contents of detrital macerals, clay mineral and pyrite are attributed to marginal aquatic/herbaceous plants and salinized and active mire water.

5.2. Paleomire evolutions

Peat accumulation is a long process. Paleopeat-forming environments were changing with prime matter, water

380 and ions supplements, acid-base properties and redox conditions (Flores, 1993; Hacquebard, 1993; Kalkreuth et al.,
381 1993), leading to paleomire evolutions. In general, when the hydrodynamic condition is weaker, fewer terrigenous
382 substances are brought into the mire, and the ash yield of the coal is lower. Meanwhile, peaceful mire water is always
383 related to a relatively reductive condition that is advantageous to the gelification and decomposition of plant debris.
384 Conversely, reactive oxygen and allochthonous deposition increase with increasing water activity, resulting in the
385 increase of inertinite maceral group, granulated macerals (such as corpocollinite) and resinite, schistic and detrital
386 macerals, which can reduce the organic sulfur in peat or coal. Analyses results in this study show that the organic
387 sulfur negatively correlates with the ash yield (Fig. 8 and Fig. 9). Positive correlation is also established between the
388 GWI and ash yield, while the TPI and ash yield are poorly correlated (Fig. 9). Therefore, the ash yield, organic sulfur
389 and GWI are effective indicators of paleomire water conditions and the TPI is an auxiliary one.

390 The facies of the three coal seams studied are predominantly composed of the upper delta plain wet forest swamp
391 (Fig. 6, Fig. 7 and Fig. 9). In addition, almost all the coal seams began and ended with this type of facies except for
392 coal seam No. 4, which began with the lower delta plain marsh. The paleomires increased in proximity to the sea in
393 the sequence of coal facies I to IV. Paleomire evolution in this sequence represents the process of transgression, while
394 the inverse sequence represents regression. The maceral composition, ash yield, organic sulfur, TPI, VI and GWI all
395 show some regularities in regard to this facies sequence (Fig. 8). According to this understanding, two, three and two
396 evolutionary stages for coal seams No. 2, No. 4 and No. 9, respectively, can be discriminated.

397 For coal seam No. 2, stage 2-I began with the upper delta plain wet forest swamp of sample 2-13 (Fig. 9)
398 overlying the coal seam floor of dark carbonaceous mudstone (Fig. 2). The paleomire changed little until the sea level
399 rose slowly and the lower delta plain fen of sample 2-9 took advantage. Likewise, the lower delta plain marsh of
400 sample 2-8 soon replaced it. Then, the sea level dropped, and the lower delta plain fen of sample 2-7 prevailed again.
401 This stage finally ended with the upper delta plain wet forest swamp of sample 2-5. Stage 2-II started right after the
402 end of stage 2-I. The sea level rose so quickly that the paleomire was replaced by the lower delta marsh directly and
403 soon by the back barrier low moor. During the second half of this stage, which was just the symmetrical version of
404 the first half of this stage, the lower delta plain marsh and the upper delta plain wet forest swamp quickly prevailed in
405 the paleomires successively. The peat-forming environments then finished and dark mudstone deposited as the coal
406 seam roof (Fig. 2).

407 Paleomires in coal seam No. 4 developed in three stages (Fig. 9). Stage 4-I was not a complete coal facies cycle
408 and was composed merely of the lower delta plain marsh of sample 4-11. Both stage 4-II and stage 4-III were the
409 complete cycles with the processes of transgression and regression. Stage 4-II proceeded rapidly, and in it quick
410 transformations occurred between the upper delta plain wet forest swamp and the lower delta plain fen. During the
411 comparatively long and complicated stage 4-III, the steady sea level promoted the occurrence of the upper delta plain
412 wet forest swamp of samples 4-7 and 4-6. Along with slow rising of the sea level, the lower delta plain fen, the lower
413 delta plain marsh and the back barrier low moor successively thrived. Gray mudstone parting of sample 4-2 deposited
414 during the highest sea level. In the regression process, the sea level dropped so quickly that the upper delta plain wet
415 forest swamp prevailed subsequently. The paleomires terminated by silty mudstone deposition as the coal seam roof
416 (Fig. 2).

417 Paleopeat of coal seam No. 9 formed in two complete cycles with the sea level rise and drop (Fig. 9). Stage 9-I
418 started with the steady upper delta plain wet forest swamp overlying the mudstone deposition (Fig. 2). Fast sea level
419 rise later led to the thriving of the lower delta plain marsh of sample 9-7 and then the mudstone parting of sample 9-6.
420 Afterwards, a quick drop of the sea level led to the development in the lower delta plain fen of sample 9-5.
421 Paleomires changed quickly in stage 9-II. The lower delta plain fen of sample 9-4 was directly replaced by the back
422 barrier low moor of sample 9-3. Likewise, during the regression, the lower delta plain fen and the upper delta plain
423 wet forest swamp became dominant successively. Finally, carbonaceous mudstone and mudstone deposited as the
424 coal seam roof (Fig. 2).

425

Fig. 9. Columnar sequences of coal facies and corresponding parameters of studied coal seams.

5.3. Adsorption and desorption divergences among coal facies

As adsorption and desorption behaviors of coal were influenced by the coalification (Yee et al., 1993), divergences of adsorption and desorption among coal facies in the three coal seams will be discussed separately due to different $R_{o, \max}$ among them (Table 1). The parameters of the coal samples from the Langmuir equation and D-A equation are listed in Table 6 and Fig. 10.

Effective parameters of the Langmuir equation and D-A equation were taken as the representations for the adsorption models of monolayer and adsorption potential theory, respectively. Both V_L and V_0 represent the ultimate adsorption capacity of the coals in these equations. As shown in Table 6 and Fig. 10, V_L and V_0 decrease from coal facies I to IV. As a comprehensive indicator of the adsorption rate and desorption rate, p_L , which is actually a function of the temperature and adsorption heat (Fu et al., 2007), changes in almost the same trend as the adsorption heat index D , while changing inversely with the pore distribution index n (Fig. 4C; Table 6). Fluctuations of these three parameters exist among the coal facies with no consistent trends in the studied coal seams (Table 6 and Fig. 10).

Characteristics of desorption have also been described by the Langmuir equation. The desorbed gas content (V_D) is in agreement with the V_L and V_0 in the adsorption process (Table 6 and Fig. 10), indicating larger adsorption amounts from the respective coals correlate with stronger desorption capacities. The $p_{L, D}$ and R_D are well correlated (Fig. 4D) but their variation trends among the coal facies are not uniform in these three coal seams (Fig. 10).

Fig. 10. Regularization isothermal adsorption-desorption parameters of coal facies of studied coal seams.

Adsorption and desorption divergences among different coal facies result from different paleomire environments, especially the variation of vegetation types and mire water condition. The arborescent plants decrease from coal facies I to IV along with the increase of hydrodynamic intensity, leading to corresponding changes in botanical tissue preservation, medium oxidizability, mineral content and ultimately, adsorption and desorption behaviors (Fig. 11). Therefore, contributing to the highest adsorption and desorption capacities are the rapid accumulation and burial of paleopeat with little mineral matter due to abundant precursor material and peaceful and reductive mire water in the upper delta plain wet forest swamp. In the lower delta plain fen and marsh, the prevailing herbaceous plants were more easily decomposed during coalification and the paleopeat also accumulated and buried slowly (Moore, 1987). The more intense hydrodynamic condition of these facies also result in a more oxidized medium and richer mineral matter. Thus, adsorption and desorption capacities of coal from the lower delta plain fen and marsh are relatively lower. In the back barrier low moor, the dominant marginal aquatic/herbaceous plants were intensely decomposed and the paleomire water was active. As ash yield from coal is counterproductive to the gas adsorption in coal (Ozawa et al., 1976; Laxminarayana and Crosdale, 1999), the lowest micropore volume and specific surface area and highest ash yield of the coal from the back barrier low moor (Fig. 8) are not conducive to gas adsorption in coal.

The adsorption heat-related parameters p_L and D and the pore distribution index n lack consistent variation trends among the coal facies in the studied coal seams (Table 6 and Fig. 10) owing to their intricate relations to the plant species, ash yield, redox conditions, etc.

Fig. 11. Simple relationship between coal facies and adsorption-desorption behaviors. Note: I, the upper delta plain wet forest swamp; II, the lower delta plain fen; III, the lower delta plain marsh; IV, the back barrier low moor; APA, arborescent plants abundance; TPD, tissue preservation degree; HI, hydrodynamic intensity; MO, medium oxidizability; MC, mineral content; AC, adsorption capacity; DC, desorption capacity; and DP, drainage potential.

5.4. Application of coal facies to CBM industry

On the above basis, the adsorption and desorption behaviors of the delta plain-littoral transitional coal are obviously influenced by coal facies. In the same coal seam, gas contents of coals reduce gradually with the increasing sea water influence to the paleomires. Coal layers from the upper delta plain wet forest swamp, with the highest gas content and strongest desorption capacity, have the best CBM drainage potential (Fig. 11). Coal layers from both the lower delta plain fen and marsh, with the higher gas content and stronger desorption capacity, also have better CBM drainage potential. However, coal layers from the back barrier low moor might lose the drainage value due to the low gas content.

Unfortunately, the experiments in this study were only measured on an air-dry basis. Further work can be conducted on moisture-equilibrium condition to obtain more applicable results.

Table 6. Statistics of the isothermal adsorption-desorption parameters of the coal facies of studied coal seams (V_L , V_0 and V_D in cm^3/g ; p_L and $p_{L,D}$ in MPa; and R_D in %).

	Coal facies	V_L				V_0				V_D				p_L			
		Min	Max	Mean	StDev	Min	Max	Mean	StDev	Min	Max	Mean	StDev	Min	Max	Mean	StDev
No. 2	I	17.43	23.47	21.20	2.24	13.96	19.33	17.32	2.00	8.90	11.61	10.87	1.00	2.03	2.60	2.37	0.21
	II	20.77	20.77	20.77	0	15.39	15.39	15.39	0	10.61	10.61	10.61	0	3.73	3.73	3.73	0
	III	14.53	14.53	14.53	0	11.88	11.88	11.88	0	7.49	7.49	7.49	0	2.13	2.13	2.13	0
	IV	10.34	10.34	10.34	0	7.43	7.43	7.43	0	5.09	5.09	5.09	0	4.49	4.49	4.49	0
	Coal facies	D				n				$p_{L,D}$				R_D			
		Min	Max	Mean	StDev	Min	Max	Mean	StDev	Min	Max	Mean	StDev	Min	Max	Mean	StDev
	I	0.21	0.25	0.24	0.02	1.53	1.67	1.59	0.05	1.20	1.80	1.49	0.24	59.06	67.30	62.94	3.30
	II	0.33	0.33	0.33	0	1.48	1.48	1.48	0	1.97	1.97	1.97	0	69.21	69.21	69.21	0
	III	0.18	0.18	0.18	0	1.87	1.87	1.87	0	1.53	1.53	1.53	0	62.84	62.84	62.84	0
	IV	0.40	0.40	0.40	0	1.34	1.34	1.34	0	1.99	1.99	1.99	0	69.42	69.42	69.42	0
No. 4	Coal facies	V_L				V_0				V_D				p_L			
		Min	Max	Mean	StDev	Min	Max	Mean	StDev	Min	Max	Mean	StDev	Min	Max	Mean	StDev
	I	18.12	21.39	19.76	1.64	14.58	17.10	15.84	1.26	9.35	10.47	9.91	0.56	2.49	2.84	2.67	0.18
	II	14.70	14.85	14.78	0.08	10.95	11.31	11.13	0.18	6.88	7.84	7.36	0.48	3.27	3.84	3.56	0.29
	III	12.54	17.03	14.79	2.25	8.27	11.85	10.06	1.79	5.20	6.95	6.08	0.88	5.18	5.75	5.47	0.29
	IV	8.98	8.98	8.98	0	6.99	6.99	6.99	0	3.83	3.83	3.83	0	3.23	3.23	3.23	0
	Coal facies	D				n				$p_{L,D}$				R_D			
		Min	Max	Mean	StDev	Min	Max	Mean	StDev	Min	Max	Mean	StDev	Min	Max	Mean	StDev
	I	0.24	0.30	0.27	0.03	1.47	1.65	1.56	0.09	1.30	1.61	1.46	0.16	61.59	64.45	63.02	1.43
	II	0.31	0.34	0.33	0.02	1.46	1.47	1.47	0.01	1.52	1.95	1.74	0.22	63.72	69.35	66.54	2.82
	III	0.44	0.45	0.45	0.01	1.27	1.34	1.31	0.04	1.17	1.56	1.37	0.20	59.29	64.17	61.73	2.44
	IV	0.33	0.33	0.33	0	1.4	1.4	1.4	0	1.02	1.02	1.02	0	55.88	55.88	55.88	0
No. 9	Coal facies	V_L				V_0				V_D				p_L			
		Min	Max	Mean	StDev	Min	Max	Mean	StDev	Min	Max	Mean	StDev	Min	Max	Mean	StDev
	I	22.38	25.17	24.05	1.02	18.15	20.66	19.63	0.97	11.70	12.49	11.97	0.31	2.03	2.58	2.36	0.20
	II	19.31	22.70	20.73	1.44	14.75	17.29	15.88	1.06	10.11	11.57	10.61	0.68	3.02	3.26	3.16	0.10
	III	18.65	18.65	18.65	0	13.94	13.94	13.94	0	7.66	7.66	7.66	0	3.76	3.76	3.76	0
	IV	12.64	12.64	12.64	0	10.35	10.35	10.35	0	6.64	6.64	6.64	0	2.31	2.31	2.31	0
	Coal facies	D				n				$p_{L,D}$				R_D			
		Min	Max	Mean	StDev	Min	Max	Mean	StDev	Min	Max	Mean	StDev	Min	Max	Mean	StDev

I	0.21	0.23	0.22	0.01	1.63	1.74	1.68	0.05	1.11	1.55	1.32	0.16	57.00	64.39	60.84	2.65
II	0.27	0.29	0.28	0.01	1.56	1.61	1.58	0.02	1.62	1.88	1.78	0.11	64.61	68.19	66.69	1.52
III	0.35	0.35	0.35	0	1.43	1.43	1.43	0	1.05	1.05	1.05	0	55.87	55.87	55.87	0
IV	0.23	0.23	0.23	0	1.62	1.62	1.62	0	1.48	1.48	1.48	0	63.78	63.78	63.78	0

V_L , Langmuir volume; V_0 , volume of micropore; V_D , desorbed gas content; p_L , Langmuir pressure; D , constant relevant to adsorption heat; n , constant relevant to pore distribution; $p_{L,D}$, Langmuir pressure in desorption; R_D , desorption rate; Min, minimum; Max, maximum; StDev, standard deviation; I, the upper delta plain wet forest swamp; II, the lower delta plain fen; III, the lower delta plain marsh; and IV, the back barrier low moor.

490 6. Conclusions

491 Four types of coal facies were distinguished based on the GI-TPI and GWI-VI diagrams, including the upper delta
492 plain wet forest swamp (I), the lower delta plain fen (II), the lower delta plain marsh (III) and the back barrier low
493 moor (IV), in coal seams No. 2, No. 4 and No. 9 in the Xiqu Drainage Area, Gujiao CBM Block, North China. The
494 upper delta plain wet forest swamp occurred dominantly, developing at the bottom and top of all the coal seams. Both
495 the ash yield and GWI tend to increase from coal facies I to IV, whereas the **volatile compounds**, fixed carbon,
496 organic sulfur, total sulfur, micropore volume and specific surface area, TPI and VI decreased. The vegetation types,
497 water supplements and disturbance, acid-base properties and redox conditions of the paleomires were responsible for
498 these divergences.

499 Paleomire evolution accompanied the rise and drop of the sea level, with a transgression or regression process
500 that could be represented by the coal facies sequence. Coal seams No. 2, No. 4 and No. 9 were discriminated into two,
501 three and two evolving stages, respectively, and these stages were characterized by complete cycles except stage 4-I
502 from the lower delta plain marsh. Mudstone partings in the coal seams deposited during the highest sea level.

503 In general, with the paleomire milieu change, especially the decrease of arborescent plants and increase of mire
504 water activity from the coal facies I to IV in each coal seam, the ultimate adsorption capacity and desorbed gas
505 content of coals decreased. Intricate influence of the plant species, terrigenous input and redox conditions of the
506 paleomire led to the lack of consistent evolution trends of the adsorption heat-related parameters p_L and D and the
507 pore distribution index n . In view of the gas content and desorption capacity of the coal layers in the same coal seam,
508 the layers from the upper delta plain wet forest swamp may have the best potential for CBM drainage, followed by
509 those from the lower delta plain fen and marsh, while those from the back barrier low moor may have the worst
510 potential.

511

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