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Influence of coal petrology on methane adsorption capacity of the Middle Jurassic coal in the Yuqia Coalfield, northern Qaidam Basin, China

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ABSTRACT

The lower coalbed methane (CBM) adsorption capacity of the low rank coals is not only related to its lower maturity, but also determined by the coal maceral compositions. In this study, a total of 13 samples including 10 coals and 3 carbonaceous mudstones, were collected from the Middle Jurassic Dameigou Formation in the borehole YQ-1 of the Yuqia Coalfield, northern Qaidam Basin, NW China. Coal lithotypes, maceral compositions, coal ranks, coal facies and methane adsorption characteristics of these samples were investigated using microscopic observation, proximate analysis, porosity analysis, and isothermal adsorption experiments. The results show that the maceral composition has a great influence on the methane adsorption capacity, and the influence of vitrinite on the methane adsorption is generally stronger than that of inertinite at a similar coal rank. For most samples, there is a positive correlation between the vitrinite contents and the Langmuir volume (V_L), a negative correlation between the inertinite contents and the V_L , and no obvious correlation between the exinite contents and the V_L . Furthermore, the vitrinite/inertinite (V/I) ratio also shows a positive relationship with the V_L . However, some samples containing large amounts of unfilled fusinite and/or semifusinite have more pore spaces favoring methane adsorption and can also adsorb significant quantities of methane. Consequently, coals with higher vitrinite contents, in association with some unfilled fusinites or semifusinites, should have the greatest adsorption capacity. Coal maceral compositions vary with the types of coal facies, and thus the methane adsorption capacity of coals may be closely related to coal facies. It is found that the methane adsorption capacities of the coals in a wet forest swamp (Type I) and an intergradation forest swamp (Type II) are greater than those in a drained forest swamp (Type III) and an open water peat swamp (Type IV). It is suggested that the area developed with the wet forest swamp and in the intergradation forest swamp with high porosity should be the best target areas for the CBM exploration in the Yuqia Coalfield.

1. Introduction

The Yuqia Coalfield, which is situated in the central part of the northern Qaidam Basin, northwestern China, has abundant coal and coalbed methane (CBM) resources, including coal reserves of 38.25×10^8 t and CBM reserves of 67.87×10^8 m³, respectively (Liu et al., 2013; OGC-MLR, 2009). However, little exploration has currently been undertaken for the CBM resources in this area, possibly due to poor understanding of the CBM geological conditions and the methane accumulating mechanisms in the vast area.

As a crucial parameter in assessing the CBM potentials, the methane adsorption is an ongoing and extremely important area of research. Meanwhile, it is significant for exploiting and developing CBM resources successfully (including replacement of CH₄ by adsorb-

ing CO₂ or N₂) (Perera et al., 2011; Zhao et al., 2012; Song et al., 2012; Komatsu et al., 2013), and for preventing gas-related accidents in underground coal mines effectively. In addition, coal permeability, as another significant parameter for gas extraction and CBM exploitation, is also influenced by the changes of methane adsorption/desorption (Zhu et al., 2013; Xie et al., 2015). Moreover, the methane adsorption characteristics are influenced by the inherent physical properties of coals, and external environmental or geological factors. Inherent physical coal properties include coal rank, ash yield, moisture content, coal lithotype, maceral composition, coal facies, and pore structure (Sánchez et al., 2010; Wang et al., 2011; Gun'ko et al., 2011; Perera et al., 2012; Olajossy, 2013; Luo et al., 2014; Li et al., 2016). External influencing factors include temperature, in situ stress, granularity, and degree of deformation (Zhang et al., 2005; Wang et al., 2011; Perera

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et al., 2012; Mahdizadeh and Tayyari, 2011; Yue et al., 2015).

A number of studies showed that methane is always adsorbed in the coal organic compositions, and the methane adsorption capacity of vitrinite and inertinite is greater than that of exinite with equal amounts (Alexeev et al., 2004; Chalmers and Bustin, 2007; Jian et al., 2015). However, the relative roles of vitrinite and inertinite in methane adsorption remain controversial. Most studies have found vitrinite-rich coals have a stronger methane adsorption capacity than inertinite-rich coals (Faiz and Hutton, 1995; Clarkson and Bustin, 1996; Bustin and Clarkson, 1998; Crosdale et al., 1998; Clarkson and Bustin, 1999; Laxminarayana and Crosdale, 2002; Mastalerz et al., 2004; Hildenbrand et al., 2006). However, there is some evidence that the methane adsorption capacity of multi-cellular inertinite is higher than that of vitrinite (Блжайоко, 1980; Wang et al., 2011; Jian et al., 2014). Meanwhile, other studies suggested that there are no trends or complex relationships between macerals and the methane adsorption capacity (Zhong and Zhang, 1990; Faiz et al., 1992; Carroll et al., 2003; Olajossy, 2013).

Coal rank plays an important role in influencing the methane adsorption capacity and maceral composition (Zhang et al., 1991; Quan, 1995; Wang et al., 2011). For the middle or high rank coals, it was reported that the methane adsorption capacity is slightly relevant to the maceral composition but is strongly correlated to coal rank (Yao et al., 2009). While for low rank coals, macerals may become the main factor controlling the pore structure and the methane adsorption when the coal seam (particularly if it is an individual seam) is located in a single coalfield or is laterally small in scale (Jian et al., 2015; Li et al., 2016). In this study, all the samples were collected from the same drill cores and have similar maximum vitrinite reflectance values of around 0.7%, which, to some extent, decreases the influence of coal rank on methane adsorption.

The phrase ‘coal facies’ refers to the primary genetic types of coal (Teichmüller, 1982). Coal facies can be identified using different methods. Macerals and their combinations were widely used to characterize coal facies (Diessel, 1982; Calder et al., 1991; Staub, 2002; Bechtel et al., 2014; Sen et al., 2016). An analysis of the relationship between the coal types and the methane adsorption characteristics suggested that the methane adsorption capacity increases respectively in coals developed in drained forest swamps, intergradation forest swamps, wet forest swamps and freshwater peat swamps. (Li et al., 2014b).

Previous studies have focused on the relationships between macerals, or coal facies, and the adsorption capacities (Faiz et al., 1992; Carroll et al., 2003; Yao et al., 2008; Li et al., 2014b). However, these relationships of coal samples with a relatively narrow coal rank from a single borehole have rarely been reported. In this study, we analyzed 13 samples ($R_{o, \max}$, 0.68–0.77%) from the YQ-1 drilling cores in the northern Qaidam Basin, northwestern China. All samples underwent proximate analysis, porosity calculations, maceral identification, equilibrium moisture analysis, and isothermal adsorption experiments. Correlation analyses not only can assess the impacts of coal petrology on methane adsorption capacity, but also predict the Langmuir volumes and Langmuir pressures of coals based on acquired data of coal petrology (Verma and Sirvaiya, 2016). The final aim is to find the best target areas for the CBM exploration in the Yuqia Coalfield.

2. Geological settings

The YQ-1 well drilling performed in 2015 is located in the western portion of Yuqia Coalfield, along the northern margin of the Qaidam Basin (Fig. 1). The Middle Jurassic (coal-bearing strata) is up to 1 km thick consisting of siliciclastic rocks and coal seams within the Dameigou Formation and the overlying Shimengou Formation. The Dameigou Formation is dominated by coarse to fine-grained siliciclastic rocks, with thick coal seams intercalated within the lower and middle parts of the formation (seams M7 and M6, respectively). The

Shimengou Formation is composed of fine-grained siliciclastic rocks and the oil shales, with thick coal seams intercalated within its lower part (seam M5).

According to the previous studies, the Dameigou and Shimengou Formations were deposited in the paleoenvironments of braided river delta and lake (Liu et al., 2013; Li et al., 2014a). The minable coal seams are the M7 and M6 coals of the Dameigou Formation, and the M5 coal of the Shimengou Formation. The seams are widely distributed across the whole coalfield and the average total thickness ranges from 2 to 35 m. The coal lithotypes in the Yuqia Coalfield are mainly semi-bright to semi-dull coals, and these coals overall belong to high volatile bituminous coals.

3. Sampling and experimental methods

Thirteen samples were taken from the YQ-1 drilling cores at approximately 1.5 m intervals, comprising six samples from the M6 seam and seven samples from the M7 seam. All samples were numbered consecutively with depth, from No. 1 to No. 13 (Fig. 1).

Lithotypes classifications were performed based on the indexes of relative glossiness and light composition content, according to Chinese National standards GB/T 18023-2000. Fresh surface of the coal samples and the minimum layer thickness over 5 cm are necessary for observation. The coal porosity (Φ) measurements were calculated by the equation of $\Phi = (\rho_s - \rho_a) / \rho_s \times 100\%$, where, ρ_s is coal skeletal density and ρ_a is coal apparent density.

The Chinese National standards GB/T 30732-2014 was applied in the proximate analyses, including analysis of moisture content, ash yield and volatile content by instrumental method. Maximum vitrinite reflectance ($R_{o, \max}$) measurements and maceral analyses (500 points) were performed by oil immersion in reflected optical light using a Leitz MPV-3 photometer microscope, in accordance with Standards GB/T 6948-1998 and GB/T 8899-1998, respectively.

All samples (90–120 g each) were prepared by crushing and sieving to a particle size range of 0.18–0.25 mm. The moisture-equilibrium treatment was processed and equilibrium moisture content of each sample was analyzed. Methane adsorption experiments of moisture-equilibrated were performed to determine the Langmuir volume (V_L) and the Langmuir pressure (P_L) following Chinese Standard Method GB/T 19560–2008. The methane adsorption capacity of moisture-equilibrated samples was measured using an IS-100 high pressure isothermal adsorption apparatus at 30 °C and a maximum equilibrium pressure of 10 MPa.

4. Results and discussion

4.1. Fundamental coal analyses

Maximum vitrinite reflectance values, proximate analyses and porosity results, maceral composition, and isothermal adsorption data from the samples are presented in Table 1.

Maximum vitrinite reflectance values for all samples are shown to be similar ranging from 0.68% to 0.77% (average, 0.72%), indicating that the coals in the study area belong to the high volatile bituminous coal and relatively concentrated values of $R_{o, \max}$ exist. The moisture content on an air dried basis (M_{ad}) and the equilibrium moisture content (M_e) vary from 1.81% to 5.75% (average, 3.58%), and from 5.81% to 14.47% (average, 10.2%), respectively, which show a weak decreasing trend with increasing $R_{o, \max}$ (Fig. 2a and b). The ash yield on an air dried basis (A_{ad}) is between 4.78% and 64.76% (average, 27.47%), which also correlates negatively with $R_{o, \max}$ (Fig. 2c). Meanwhile, there are three carbonaceous mudstones (dirt bonds in the coal seams) including Nos. 2, 10, 11 samples, and all the ash yields of them are higher than 50%. In addition, the porosity of samples ranges from 5.26% to 18.06%, which also shows a decreasing trend with the increasing A_{ad} (Fig. 2d). The ash yields, derived from

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