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# Impact of fracturing fluid on multi-scale mass transport in coalbed methane reservoirs



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### article info abstract

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The effect of fracturing fluid on multi-scale mass transport, including methane desorption, diffusion and percolation, was investigated on an anthracite coal from the Qinshui Basin. Methane sorption and diffusion characteristics were measured using the volumetric method. The Langmuir equation was used to fit the sorption isotherm data and unipore model to estimate the effective diffusion coefficient  $(D_e)$ . The impairment of gas permeability, characterized by the residual resistance factor  $(F_{rr,g})$ , was determined using the pulse-decay method. In addition, changes in pore structure and surface properties were also analyzed by SEM/EDX, low-temperature  $N_2$  adsorption and XPS. The results show that the methane adsorption capacity was reduced by 10.5% after fracturing fluid treatment. The Langmuir volume  $(V_L)$  decreased while the Langmuir pressure  $(P_L)$  increased, which is beneficial for gas desorption. The effective diffusion coefficient  $(D_e)$  exhibited a negative correlation with pressure and decreased by 23.0% to 37.6%. The gas permeability was significantly diminished and  $F_{rr,g}$  for a macrofractured sample was much greater than that for a micro-fractured sample. Furthermore, due to the adsorption of fracturing fluid on pore/fracture walls, the mass transport channels were narrowed and hydrophilia was enhanced. This possibly explains the reduction of gas adsorption capacity and mass transport rate in coal.

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### 1. Introduction

Coalbed methane (CBM) is a rapidly expanding clean energy source which is efficient and low-cost. It is estimated that there is between 500 and 3000  $\times$  10 $^{12}$  scf (14.2 to 84.9  $\times$  10 $^{12}$  m<sup>3</sup>) of recoverable CBM worldwide [\(Dong et al., 2015](#page--1-0)). CBM reservoirs have unique characteristics compared with conventional gas reservoirs, related to the pore structure, gas storage and transport mechanisms [\(Clarkson and Bustin,](#page--1-0) [1999\)](#page--1-0). According to the classification provided by the International Union of Pure and Applied Chemistry (IUPAC), coal pores can be divided into micropores ( $\leq$ 2 nm), mesopores (2–50 nm) and macropores (≥50 nm), of which 90% are smaller than 10 nm [\(Charrière et al.,](#page--1-0) [2010](#page--1-0)). Coal represents a dual pore system, characterized by primary porosity related to micropores and mesopores in the coal matrix, and secondary porosity defined by macropores or cleats surrounding the matrix blocks [\(Warrant and Root, 1963\)](#page--1-0). Substantial quantities of gas  $(>95%)$  are physically adsorbed by the large internal surface area of the micropores and mesopores [\(Gray, 1987\)](#page--1-0). Generally, gas transport in coal is considered to be a multi-scale mass transport process, as shown schematically in [Fig. 1](#page-1-0). CBM desorbs from the coal particles before flowing from the matrix into the cleats via Fick's Law of diffusion,

Corresponding author. E-mail address: [hffs2008@vip.qq.com](mailto:hffs2008@vip.qq.com) (F. Huang). after which it moves through the cleats via Darcy's Law of flow, and finally into the wellbore.

The productivity of a CBM well is routinely stimulated with hydraulic fracturing. During the hydraulic fracturing operations, the injected fracturing fluid first fills and opens up the cleats, thus creating flow paths under positive pressure. The fluids introduced to the fracture network can then spontaneously imbibe into the matrix pores and move away from the fracture faces due to capillary pressure [\(Chaturvedi](#page--1-0) [et al., 2009; Ghanbari and Dehghanpour, 2015](#page--1-0)). Field observations have shown that a large percentage of the injected fracturing fluid, typically 20% to 30%, is not recovered during the clean-up phase; it remains in the pore-fracture system of the coal [\(Puri et al., 1991](#page--1-0)). As a result, the multi-scale mass transport mechanism in the CBM reservoirs, including methane desorption, diffusion and percolation, can be negatively affected by the trapped fracturing fluid.

Research on the effect of moisture on methane adsorption and diffusion in coal has been widely carried out recently [\(Joubert et al., 1974;](#page--1-0) [Crosdale et al., 2008; Day et al., 2008; Pan et al., 2010; Gensterblum](#page--1-0) [et al., 2013; Xu et al., 2015\)](#page--1-0). The results of these studies suggest that the methane adsorption capacity is negatively correlated with moisture until a critical moisture level is reached and that the methane diffusivity decreases with increasing moisture. However, the in situ coal is usually assumed to be moisture-equilibrated. This means that moisture is expected to be beyond the critical level and is present in three main forms: adsorbed, free and chemically bound water ([Charrière and](#page--1-0)





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Fig. 1. Multi-scale mass transport mechanism in CBM reservoirs.

[Behara, 2010; Arisoy and Akgun, 1994](#page--1-0)). Under such moistureequilibrated conditions, it is not clear whether the invasion of fracturing fluid would still affect the methane adsorption and diffusion in coal. [Chen et al. \(2009\)](#page--1-0) have studied the effect of fracturing fluid on the methane adsorption in coal. They found that the fracturing fluid treatment lead to a lower adsorption capacity and a higher Langmuir pressure. Nevertheless, the experimental methods used in their study had two main disadvantages. The first one was that the sample treated with fracturing fluid was prepared from dry coal. In that case, the initial water content was ignored, while in fact it plays an essential role in the water spontaneous imbibition. For this reason, the impact of fracturing fluid was inaccurately estimated. The second disadvantage of this study was that the Langmuir parameters were fitted by the adsorption isotherm, while CBM recovery is in fact a process of desorption rather than adsorption and the sorption hysteresis may affect the Langmuir measurements [\(Wang et al., 2014](#page--1-0)). [Li et al. \(2015\)](#page--1-0) studied the effect of fracturing fluid on the methane diffusion in CBM reservoirs and found that the methane diffusivity was significantly decreased by the fracturing fluid. However, their methods had the same problems mentioned above, as well as an additional issue of not considering the pressure dependence of gas diffusion in coal.

The influence of fracturing fluid on the percolation properties of CBM reservoirs has also been widely investigated ([Puri et al., 1991;](#page--1-0) [Chen et al., 2006; Gao et al., 2015; Li et al., 2015\)](#page--1-0). The results of these studies show that the fracturing fluid treatment negatively affects both the absolute and relative permeabilities. On the other hand, the permeability measurements in these studies were based on steadystate methods. In that case it may be quite difficult to determine the permeability of tight rocks, especially shale and coal, because long-time scales are involved and it is required to use instruments monitoring extremely small flow rates or pressure changes ([Walls et al., 1982; Cui](#page--1-0) [et al., 2009\)](#page--1-0). For this reason, the pulse-decay method has been widely applied instead for the low-permeability measurements [\(Wang et al.,](#page--1-0) [2015; Metwally and Sondergeld, 2011](#page--1-0)).

In this paper, we apply a series of experimental methods designed to overcome the highlighted deficiencies, which allow us to properly study the effect of fracturing fluid on the multi-scale mass transport in coal. First, methane ad/de-sorption, diffusion and permeability measurements are conducted before and after fracturing fluid treatment of a Chinese anthracite coal. This makes it possible to comprehensively analyze the multiple effects of the fracturing fluid. Second, the changes in pore structure and surface properties are analyzed using scanning electron microscopy with energy dispersive X-ray spectroscopy (SEM/ EDX), low-temperature  $N<sub>2</sub>$  adsorption and X-ray photoelectron spectroscopy (XPS) methods. This makes it possible to reveal the mechanisms governing the variations of mass transport properties induced by the fracturing fluid. Results of this research may prove valuable to understand the mechanisms of formation damage and to improve the modeling of gas flow in coal.

### 2. Experimental methods

### 2.1. Sample preparation

One anthracite coal block from the No. 15 coal seam of the Qinshui Basin in Shanxi Province (China) was prepared for this study. The results of proximate and petrographic analyses are summarized in Table 1. Details on the mineral composition and the relative content of clay minerals are given in [Tables 2 and 3.](#page--1-0) After sample acquisition, part of the coal block was ground into powder. Approximately 200 g of 60–80 mesh coal samples were sieved for the purpose of methane ad/de-sorption and diffusion measurements, and approximately 50 g of 18–20 mesh coal samples for low temperature  $N_2$  adsorption experiments and approximately 10 g of  $-200$  mesh coal samples for XPS analyses. In addition, small coal cubes with a volume of 1  $\text{cm}^3$  to 2  $\text{cm}^3$  were cut from the block for SEM/EDX studies. For the purpose of the above tests (methane ad/de-sorption, diffusion, SEM/EDX, low temperature  $N_2$  adsorption and XPS), raw moisture-equilibrated coal samples were prepared by placing the samples in a desiccator, where a super-saturated solution of  $K_2SO_4$  was used to provide ~97% relative humidity at room temperature. Furthermore, the raw samples were treated by soaking in the fracturing fluid for 48 h, in order to ensure that the samples were uniformly saturated. The soaked samples were then filtered through a strainer until the fluids stopped dripping from the bottom. Additionally, cylindrical coal samples with a diameter of ~2.54 cm were drilled from the coal block. Following this, the top and





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