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Temperature-dependent diffusion process of methane through dry crushed coal

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ABSTRACT

Temperature influence or geothermal effect is pervasive in the carbon dioxide geological sequestration, GIP (gas in place) estimation, and the Coal Mine Methane (CMM) degasification process. The temperature effects on the adsorption process are not explicitly considered in previous gas diffusion tests. This study seeks to treat the adsorption–diffusion process as a unified process and then investigate the temperature influence effect on this process. Isothermal tests under different constant temperatures show that 1) Langmuir constant-a is independent of temperature, and Langmuir constant-b can be treated as a negative linear function of temperature in engineering applications. Two groups of diffusion tests were conducted: diffusion under constant temperature and diffusion with increasing temperature. The diffusion tests show that 2) for the diffusion process with increasing temperature, the equivalent diffusion coefficient (D_{eq}) increases with increasing temperature, and this relationship can be described by the revised Arrhenius-style equation. 3) For the diffusion process under constant temperature, the diffusion coefficient (D) is dependent on temperature and independent of adsorption equilibrium pressure. The relationship between diffusion coefficient and temperature can also be described using the revised Arrhenius-style equation. 4) Diffusion coefficients range from 8.53×10^{-8} cm^2/s to 1.97×10^{-7} cm^2/s when temperature rises from 30 °C to 90 °C.

1. Introduction and background

China is the biggest coal producer and consumer in the world, and its coal production in 2013 was 3.7 billion metric tons. According to the geological survey, 61.14% of deep coal reserves are greater than 1000 m (Tang et al., 1999). There are currently more than 40 coal mines mining at depths greater than 1000 m, and the deepest mining depth is 1501 m. With increasing mining depth, the methane induced coal mine disasters are more pronounced because of the high CMM gas content and pressure, which also attributes the coal and gas outburst during mining activity (Lama and Bodziony, 1998; Hao et al., 2014). Thus, a majority of state-of-the-art coal seam degasification techniques are developed to lower the coal seam methane content and pressure (Cheng et al., 2004; Wang, 2006; Yi, 2007; Yang et al., 2008; Jiang et al., 2008; Li et al., 2011; Wang, 2012; Karacan et al., 2011; Zabetakis et al., 1973; Noack et al., 1998; Hyman et al., 1987). These techniques can be classified into two groups according to their special mechanisms. One group increases the fluidity of CMM in a coal seam by increasing coal seam permeability in combination with the mechanical approach such as hydraulic fracture, hydraulic cutting, blasting and regional in-situ stress release (Guo et al., 2011; You et al., 2010; Sun et al., 2010; Feng et al., 2001; Zhou et al., 2014). The other group enhances the CMM diffusion property on the micro-surface of coal by taking advantage of the heat effect. The hot-water injection method, flue gas injection method, water jet, alternative electromagnetic field, and ultrasonic vibrating technique utilize the heat effect by increasing the internal energy of CMM to accelerate gas desorption-diffusion process (He, 1996; Yang et al., 2008; Jiang

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et al., 2008; Li et al., 2011; He et al., 2010; Liu et al., 2004; Yang et al., 2010; Ge et al., 2011; Yang et al., 2013). Thus, the temperature effect on sorption–diffusion process should be considered.

When the GIP (gas in place) is estimated for a deep coal seam, an issue related with temperature or geotherm appears. The difference of coal seam temperature between 500 m and 1500 m is approximately 15–35 °C in U.S. (Nathenson and Guffanti, 1988), which will have a mark influence on the GIP estimation (Hildenbrand et al., 2006; Gensterblum et al., 2013, 2014; Busch and Gensterblum, 2011). In addition, the temperature difference also affects the gas desorption-diffusion property during coal sample collection stage through drilling (Sakurovs et al., 2008a, 2008b).

Geological sequestration of carbon dioxide is one approach for reducing CO_2 emissions and the greenhouse effect (Marini et al., 2006; White et al., 2003; Holloway, 2005; Gasparik, M. et al., 2014). Researches show that deep unminable coal seams are a favorable geologic medium for CO_2 sequestration, and CO_2 injection to coalbed methane reservoir has also been utilized to enhance coalbed methane (CO_2-ECBM) recovery (Gentzis et al., 2000; Van Bergen et al., 2006; Kronimus et al., 2008; White et al., 2005; Morsi and Schroeder, 2005). The combination of reservoir temperature and pressure also determines the CO_2 existing status and storage capacity of the unminable coal seam (Sakurovs et al., 2008a,b; Busch et al., 2004; Gensterblum et al., 2014).

However, the relationship between temperature and the diffusion coefficient is not clearly or quantitatively understood even though lots of scholars have done extensive experimental and theoretical studies on CH_4 diffusion process on coal (Smith and Williams, 1984; Yang and Wang, 1986; Busch et al., 2004; Gensterblum, 2012; Charrière et al., 2010; Clarkson and Bustin, 1999; Cui et al., 2004; Kumar, 2007; Mavor et al., 1990; Nandi and Walker, 1970a, 1970b; Saghafi et al., 2007; Shi et al., 2003; Siemons et al., 2007). Furthermore, even though the sorption stage determines the maximum diffusion volume of gas, the temperature effect on the sorption stage is not explicitly considered during the diffusion test, especially for estimating the maximum gas adsorption content under different temperature and pressure. Thus, this study seeks to treat the sorption-diffusion as a unified process and investigate the temperature influence on this process. Isotheral tests of methane on crushed coal under different temperatures are conducted, which shows the relation between temperature and Langmuir adsorption constants (a and b). Next, the diffusion test of methane on crushed coal is performed with particular emphasis given to the effect of temperature. Two diffusion processes, diffusion under constant temperatures and diffusion under increasing temperatures, are tested. The isothermal test is conducted by means of the volumetric adsorption approach, and diffusion kinetic data is obtained by monitoring the gathering volume of diffusion gas after pressure drop to 1 atm in sorption–diffusion experiments (Jian et al., 2012; Crosdal, 1995; Nandi et al., 1970; Gasparik et al., 2013).

2. Experimental section

2.1. Sample preparation

The blocks of coal were obtained from the Zhaogu coal mine, Jiaozuo, Henan province in China. The coal sample is high quality anthracite coal. The physical parameters of the coal were analyzed following Chinese national standards (Table 1). Next, the coal specimen was ground and sieved by 0.25 mm–0.5 mm metal sifters, and then placed into a drying oven at 200 °C to dehydrate. After dehydration was completed, the prepared sample was stored in a dehydrator for later usage.

Table 1
Physical parameters of coal sample.

Sample	A_{ad} (%)	V_{daf} (%)	TRD_{20}^{20} (g/cm)	ARD_{20}^{20} (g/cm^3)	Porosity (%)
Zhaogu coal mine	9.68	9.93	1.56	1.48	5.13

2.2. Isothermal test

The isothermal tests under different temperatures (30 °C, 40 °C, 50 °C, and 60 °C) were conducted using the isothermal ad/desorption instrument, IS-100, made by the TerraTek Corporation in U.S.

The Langmuir equation has been widely used to describe the methane sorption process in coal (Pillalamarry et al., 2011; Yu, 1992). The isothermal equation was established using the Langmuir model,

$$Q_T = \frac{a \cdot b(T) \cdot P}{1 + b(T) \cdot P} \tag{1}$$

where, Q_T is the sorption gas content (cm^3/g), P is the gas pressure of the equilibrium status (MPa), the Langmuir constant-a represents the maximum adsorbed gas at infinite pressure (cm^3/g), and the Langmuir constant-b is the reciprocal of Langmuir pressure (MPa^{-1}) and dependent on temperature.

Once the isothermal tests under different temperatures were completed, the Langmuir constants (a and b) under different temperatures can be obtained. Then the Langmuir equation involving temperature influence is obtained. Then the equation (2) is used to estimate the adsorption gas content of coal in the equilibrium status under different temperature and pressure.

$$Q_\infty = \frac{a \cdot b(T) \cdot p}{1 + b(T) \cdot p} \cdot \frac{100 - A_{ad} - M_{ad}}{100} \cdot \frac{1}{1 + 0.31 M_{ad}} \tag{2}$$

where, Q_∞ is the maximum adsorption content of coal, a is Langmuir constant, b(T) is Langmuir constant (temperature-dependent function), p is adsorption equilibrium pressure, A_{ad} is ash content and M_{ad} is moisture content. This lays a foundation for calculating diffusion coefficient late.

2.3. Diffusion test

The diffusion test was conducted with a self-assembled sorption-diffusion setup (Fig. 1). The entire system consists of two main units: an isothermal sorption unit and a diffusion gas measurement unit. The sorption unit provides the adsorption equilibrium state of methane on crushed coal under different temperatures and pressures. The sorption unit is composed of a methane charging unit, a vacuum degassing unit and an adsorption unit. The methane charging unit is used to charge methane to the canister. The vacuum degassing unit is used to degas coal samples prior to charging the sample container with methane. The thermostatic waterbath provides a constant temperature ranging ±0.1 °C of setting temperature.

The diffusion gas measurement unit gathers and measures the diffusion gas by means of the water displacement method. In order to compare and analyze methane volume under different temperatures and pressures, the following equation is needed to unify the measured gas volume,