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Experimental study of impact of anisotropy and heterogeneity on gas flow in coal. Part I: Diffusion and adsorption

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ABSTRACT

Gas is adsorbed in the pores of coal matrix and during gas production gas is desorbed from the pore surface and diffuses through the matrix pore structure and flows in the fracture/cleat system to the production well or boreholes. However, coal is highly heterogeneous and anisotropic. How heterogeneity and anisotropy affect the gas storage and especially the diffusion behaviour is not well studied. In this work, a series of measurements were performed on three dry cubic coal samples cut from the same coal block from the Bowen Basin, Australia, using an adsorbing gas, methane. For each sample, gas adsorption experiments with gas flowing from three principal directions were performed. The diffusion data was fitted with a bidisperse diffusion model to obtain diffusion coefficient. The three samples, although from the same coal block, showed difference in adsorption amount and significant difference in effective diffusivity. It was found that the effective macropore diffusivity increased with gas pressure and effective micropore coefficient decreased with gas pressure. The effective diffusivity showed difference among samples and directions, demonstrating coal heterogeneity and anisotropy both have a significant impact on gas diffusion behaviour. However, no generalisation can be obtained with any single pore structure parameter, such as pore size or surface area, as it may be related to all pore and fracture structures at various scales.

1. Introduction

Coalbed methane (CBM) as a kind of clean energy makes up about 7%, 35% and 3% of the annual natural gas production in the USA, Australia and China, respectively [1]. The knowledge of gas storage and migration mechanisms in coal is vital for optimising gas production and reservoir evaluation [2]. It is widely assumed that coal is dual-porosity, that is, gas transport through cleat system described by Darcy flow and in matrix pores via diffusion [3,4]. Therefore permeability of the cleat system and diffusivity of the matrix pores are two critical parameters for CBM production. The coal permeability has been studied extensively (e.g., [5–7]), however gas diffusion in coal matrix was not studied extensively (e.g., [2,3,8]).

Experimentally studying methane diffusion mechanism in coal matrix and accurately determining methane diffusivity are of great importance for CBM production. Busch and Gensterblum [2] reviewed literature on gas diffusion in coal matrix, including the experimental work [3,9–19], unipore diffusion modelling [9,10,12,18,20], and the bidisperse diffusion modelling using Ruckenstein et al. [21] approach [9,12,13,19,22,23]. Among these modelling work summarised in Busch and Gensterblum [2], most models treated coal matrix as spherical and isotropic. Often laboratory study of gas diffusion in coal matrix uses coal powder or crushed coal (e.g., [11,14,17]), therefore, these assumptions may be valid. However, crushed coal may not well possess original matrix pore structure. Hence, experimental work using coal core is required to investigate the gas diffusion behaviours [3,24].

Besides the publications summarised in Busch and Gensterblum [2], a few recent experimental attempts have been performed to investigate gas diffusion in coal. For instance, transient diffusivity using a limit approximation approach was proposed to fit laboratory desorption data using coal powder samples [25]. Experimental measurements on coal were performed and unipore diffusion model and Fick's law were used to characterise methane diffusivity, but with limited success [26]. The impact of maceral composition and coal rank on gas diffusion was

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experimentally investigated on 18 crushed Australia coals [27] and it was found that gas diffusion rate varied over six orders of magnitude for the tested coal samples, and the higher the inertinite content the higher the diffusion rate. All above mentioned work were on coal powder or crushed coal, and some other work have been using larger coal samples. For instance, diffusion experiments were performed on cylindrical coal samples and bidisperse diffusion model was used; it was found that calculated diffusivity increased with respect to pore pressure [8]. An experimental method to measure methane diffusivity on coal matrix flakes was proposed, and the impact of gas pressure, coal rank and moisture content on diffusivity was analysed [28]. A few theoretical studies on diffusion have also been conducted since the review by Busch and Gensterblum [2]. For instance, the diffusivity was found to increase with the gas pressure [29]. The diffusion kinetics was found to be different between adsorption and desorption processes, and the bidisperse model was more suitable than unipore model when modelling the diffusion process for the coal samples studied, and the macropore diffusivity was one or two orders of magnitude greater than the micropore diffusivity [30]. However, no work has been performed to study the impact of coal anisotropy and heterogeneity on gas diffusion.

Coal has anisotropic and heterogeneous pore structure, including macropores (> 50 nm), mesopores (2–50 nm), and micropores (< 2 nm) (e.g., [31]). Besides anisotropic pore distribution, the composition and fabric at all scales also determine that gas diffusion in coal is heterogeneous. Coal has strong heterogeneity due to a combination of many geological factors including sediment-source regions, depositional environments, tectonic settings, diagenesis, climate and hydrological conditions [32]. Coal samples even from the same block have various mineral components. For instance, Busch et al. [9] analysed data of grain size fractions of the coal sample from the Silesia coal mine. They found that the maceral composition, vitrinite contents, inertinite contents and the liptinite contents varied strongly with grain size fraction. Moreover, the diffusion mechanism in different components is distinguished. For instance, Laxminarayana and Crosdale [33] studied the influence of maceral composition on diffusivity and found that inertinite-rich coals usually had faster diffusion rates comparing with their rank equivalent vitrinite-rich ones. Karacan [34] performed measurements on a range of lithotypes in a bituminous coal sample and observed that CO_2 diffusion was faster in the clay + inertite region compared to the vitrinite area.

Knudsen number, the ratio of the molecular mean free path to the pore diameter, represents the relative degree of gas molecules collision with the gas molecules and pore walls and it is commonly used to classify diffusion mechanisms (Fickian diffusion, Knudsen diffusion and surface diffusion) [35]. The pores with different size have different diffusion mechanisms and different diffusivity, so gas diffusion is closely related to the pore structure in coal matrix. As the matrix pores are anisotropic and heterogeneous, diffusion is therefore anisotropic and heterogeneous in coal matrix. The availability of such knowledge may provide the foundation for more practically modelling gas diffusion process in coal matrix for CBM production. However, anisotropic diffusion process through coal matrix is still not well-understood. Thus, the impact of coal anisotropy and heterogeneity on gas diffusion requires further investigation.

In this work, a series of measurements were performed on three cubic coal samples cut from a same coal block from Bowen Basin, Australia, using adsorbing gas, methane. For each sample, gas flow in each of the three principal directions was measured. The samples were dried before measurements to avoid the influence of moisture on gas diffusion and adsorption results. The diffusion data was fitted with a bidisperse diffusion model. Then the anisotropy and heterogeneity of effective diffusivity were discussed with relation to pore structure.

2. Experimental

2.1. Sample description and preparation

A coal block was recovered from Bowen Basin, Queensland, Australia, and three cubic samples, named Sample 1, Sample 2, and Sample 3, were cut from different layers of the coal block using a diamond wire saw, then the surfaces of the cubes were grinded. The detailed process of sample preparation can be referred to our previous work [36]. The size of each sample is approximately 23 mm on each side. One principal direction of each sample is perpendicular to bedding plane and the other two principal directions are parallel to bedding plane.

To eliminate the influence of moisture on flow results and to facilitate comparison among samples, each sample was dried at 70 °C in a vacuumed oven for at least two days until the weight of the sample remained constant. It is worth noting that the inherent moisture in matrix and free phase water in cleat in in-situ coal reservoirs have significant impact on gas diffusion and adsorption [3]. Therefore, the results obtained from dry coal samples are different from moist coal samples.

The offcuts of each cubic sample were crashed and used for vitrinite reflectance, proximate analysis and ultimate analysis and the results are summarised in Table 1. Maceral compositions for each sample were also analysed based on 550 point counts of each sample and the results are summarised Table 2. From these results, it can be seen that Sample 2 has less carbon and vitrinite contents compared to the other two samples.

2.2. Adsorption and diffusion measurement methods

The adsorption and diffusion measurements were conducted using a tri-axial cell apparatus shown schematically in Fig. 1. The cubic sample firstly was placed in a 3D-printed membrane which was printed using photopolymer and had an outside diameter of 1.5 in. (3.81 cm). The combination of cubic sample and 3D-printed membrane then was put in a standard rubber sleeve, which was installed in a tri-axial cell for measurements of gas diffusion, adsorption and permeability [37]. The temperature of the sample cell was kept constant in a water bath to ensure that all measurements were carried out at 34.5 °C. An ISCO pump was used to supply confining pressure up to 9 MPa. It should be mentioned that after the sample was installed in the rig, a confining pressure of 5 MPa was applied and maintained about five hours to consolidate the sample. The gas used for experiments was pure CH_4. During experiments, CH_4 was injected to the sample from the up-stream side. Diffusion and adsorption were measured under four gas pressure steps. Pressure change against time was recorded at each pressure step until the adsorption equilibrium was reached when pressure remained unchanged. Then the permeability measurements were performed,

Table 1
Vitrinite reflectance and the compositions of the three samples.

	$R_{o,max}$ (%)	Proximate analysis (mass%)			Ultimate analysis (mass%)					
		volatile	carbon	ash	C	O	Al	Si	Fe	N
Sample 1	0.90	1.51	88.6	9.9	84.4	9.3	1.7	1.7	1.7	1.1
Sample 2	1.02	1.54	72.9	25.5	77.8	14.1	3.1	3.3	0.6	1.1
Sample 3	1.05	1.58	83.0	15.4	81.6	13.1	1.5	1.6	0.7	1.5