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## **Laboratory measurement and modelling of coal permeability with different gases adsorption**

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**Abstract:** Adsorption of gas on coal will induce coal matrix swelling. Under reservoir conditions these strain changes affect the coal permeability. A series of laboratory measurements of gas permeability of a coal sample from Qinshui basin has been conducted, using a new apparatus developed for measuring coal permeability with various gases adsorption, by use of the transient pulse decay technique. Results show that permeability measured using CH<sub>4</sub>, N<sub>2</sub> and CO<sub>2</sub> all decrease with increasing gas pressure under the constant effective stress condition. The permeability decrease shows some variations with gas species. Permeability measured by CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub> decrease almost 91%, 70% and 32% respectively, from gas pressure of 0.5 MPa to 4 MPa. The experimental results are then modelled using the Robertson and Christiansen permeability model. Results show the agreement between the modelled results and the experimental results in the pressure range of the experiments, especially for CH<sub>4</sub> and N<sub>2</sub>. [Received: June 30, 2012; Accepted: November 5, 2012]

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## 1 Introduction

Gas injected into coal for enhanced coalbed methane (ECBM) recovery is an innovative technique which appeared in the early 1990s (White et al., 2005). The injected gas can be two types, inert gas such as N<sub>2</sub>, and affinity gas such as CO<sub>2</sub> (Puri and Yee, 1990). Injecting CO<sub>2</sub> into unminable coal seams has been paid special attention in the past decade. The reason is that unminable coal seams are potentially large storage reservoirs for permanent sequestration of greenhouse gas (GHG), i.e., CO<sub>2</sub>, and offer the benefit of enhanced methane production, which can offset some of the costs associated with CO<sub>2</sub> sequestration (Robertson, 2009).

However, the field test experiences in the past decade reveal that there is a main technical barrier faced in CO<sub>2</sub>-ECBM. That is CO<sub>2</sub> induced coal matrix swelling reduce gas injectivity of the well. Take the Allison CO<sub>2</sub>-ECBM field test as an example, a permeability reduction of over two orders of magnitude was observed in the injection well (Reeves et al., 2003). Significant decline in CO<sub>2</sub> injectivity has also been reported in other trials (Shi et al., 2008; Van Bergen et al., 2006). However, dramatic reduction in injectivity has not been observed in trials where pure N<sub>2</sub> or flue gas was used, as shown in the ARC flue gas micro-pilot (Gunter et al., 2004). On the contrary, injection of N<sub>2</sub> or flue gas has shown to not only reverse the permeability reduction caused by CO<sub>2</sub> injection but also enhance the well injectivity (Gunter et al., 2004). The China CO<sub>2</sub>-ECBM pilot test also faced the same problem, namely, injectivity decreased for coal matrix swelling with CO<sub>2</sub> injection (Alberta Research Council, 2007).

As we all know, coal permeability is a key parameter to control gas flow in coalbed (Ashwani, 2005). The coal permeability reduction as just mentioned above not only be disadvantageous to CO<sub>2</sub> injection, but also be harmful to CH<sub>4</sub> production. N<sub>2</sub> has the effect of enhancing coal permeability, as shown in the ARC micro-pilot. All these coal permeability change phenomena are due to gas adsorption on coal, or more precisely, adsorption-induced coal swelling (Pan and Connell, 2007). Coal is a dual porosity media composed of matrixes and fractures (cleats) (Warren and Root, 1952). Swelling of coal matrix reduces the cleat width and thus coal permeability. Coal adsorption capacities of different gases are not the same, so coal permeability changes differ with different gases adsorption (Cui et al., 2007; Pini et al., 2009, 2010; Robertson and Christiansen, 2005; Zhang et al., 2008; Mazumder and Wolf, 2008; Wang et al., 2007). Therefore, it is

significant to investigate the change of coal permeability with different gases (i.e., CO<sub>2</sub>, N<sub>2</sub>, CH<sub>4</sub>, etc.) adsorption.

In order to predict the effects gas adsorption-induced coal swelling on coal permeability, a number of permeability models have been developed (Gray, 1987; Sawyer et al., 1987, 1990; Seidle and Huitt, 1995; Harpalani and Chen, 1995; Levine, 1996; Palmer and Mansoori, 1996, 1998; Gilman and Beckie, 2000; Shi and Durucan, 2004, 2005; Cui and Bustin, 2005; Cui et al., 2007; Robertson and Christiansen, 2006; Zhang et al., 2008; Pini et al., 2009; Liu and Rutqvist, 2010; Liu et al., 2010; Connell et al., 2010; Izadi et al., 2011; Wang et al., 2011, 2012). Among these models, the Palmer and Mansoori (P&M) and the Shi and Durucan (S&D) coal permeability models are the most widely used in reservoir simulation. The P&M model presented below assumes uniaxial strain and constant vertical stress conditions. Based on these assumptions, the following equation was derived to estimate the changes in cleats porosity due to both pore pressure and coal swelling/shrinkage:

$$\frac{\phi}{\phi_0} = 1 + \frac{C_m}{\phi_0} (p - p_0) + \frac{\varepsilon_l}{\phi_0} \left( \frac{K}{M} - 1 \right) \left( \frac{p}{p_c + p} - \frac{p_0}{p_c + p_0} \right) \quad (1)$$

where  $C_m$  is given as:

$$C_m = \frac{1}{M} - \left[ \frac{K}{M} + f - 1 \right] \gamma \quad (2)$$

where  $\phi$  is the porosity at pressure  $p$ ,  $\phi_0$  is the porosity at reference pressure  $p_0$ ,  $\varepsilon_l$  and  $p_c$  are fitting parameters for Langmuir-like model to describe volumetric strain with gas adsorption,  $\gamma$  is the solid compressibility,  $f$  is a fraction between 0 and 1.  $K$  (bulk modulus) and  $M$  (constrained axial modulus) are related to Young's modulus,  $E$ , and Poisson's ratio  $\nu$ , through isotropic elasticity theory,

$$\frac{M}{E} = \frac{1 - \nu}{(1 + \nu)(1 - 2\nu)} \quad \text{and} \quad \frac{K}{M} = \frac{1}{3} \left( \frac{1 + \nu}{1 - \nu} \right) \quad (3)$$

The change in permeability was then determined using the following cubic equation:

$$\frac{k}{k_0} = \left( \frac{\phi}{\phi_0} \right)^3 \quad (4)$$

Palmer et al. (2007) modified the original P&M model to account for the exponential increase of absolute permeability, with a newly defined  $C_m$  function:

$$C_m = \frac{g}{M} - \left[ \frac{K}{M} + f - 1 \right] \gamma \quad (5)$$

where  $g$  is a geometric term related to the orientation of the natural cleat system.

S&D proposed a permeability model, also based on matchstick representation of coalbed. They also assumed uniaxial strain and constant vertical stress conditions and the model are presented below:

$$\sigma - \sigma_0 = -\frac{\nu}{1-\nu}(p - p_0) + \frac{E}{3(1-\nu)}\varepsilon_l \left( \frac{p}{p + p_\varepsilon} - \frac{p_0}{p_0 + p_\varepsilon} \right) \quad (6)$$

The variation in permeability was then given by an equation similar to that proposed by Seidle et al. (1992):

$$\frac{k}{k_0} = \exp[-3C_f(\sigma - \sigma_0)] \quad (7)$$

where  $C_f$  is referred to the cleat volume compressibility with respect to changes in the effective horizontal stress normal to the cleats.

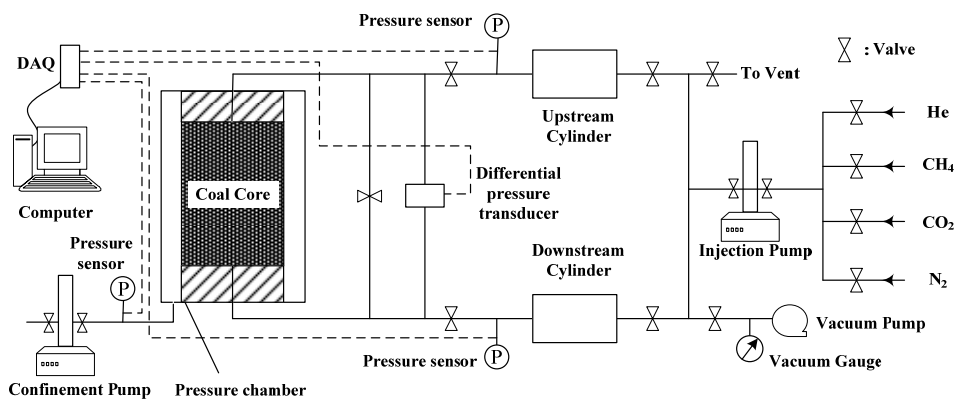
Robertson and Christiansen (2006) developed a permeability model that describes the permeability behaviour of a fractured, sorptive-elastic media, such as coal, under typical laboratory conditions where common radial and axial pressures are applied to core sample during permeability measurements.

In this work, a new apparatus has been developed for measuring coal permeability with various gases adsorption, by use of the transient pulse decay (TPD) technique. The permeability measurement experiments have been conducted, using  $\text{CO}_2$ ,  $\text{N}_2$ , and  $\text{CH}_4$ , respectively. The experimental results are then analysed detailed and modelled using the Robertson and Christiansen permeability model which is commonly used during measurement of permeability data in the laboratory.

## 2 Experimental work

### 2.1 Experimental apparatus and procedure

Figure 1 shows the schematic diagram of the TPD testing device used for this study (Brace et al., 1968; Hsieh et al., 1981; Siriwardane et al., 2009; Pan et al., 2010; Wang et al., 2011). A pressure chamber was used for the experimental measurement of gas permeability under hydrostatic pressure conditions. A confinement pump was used for applying confining pressure. The core sample, about 25 mm in diameter and 50 mm in length, is wrapped with a thin lead foil then a heat shrinkable tube before it is installed in the pressure chamber. The thin lead foil is to prevent gas diffusion from the core to the confining fluid at high pressures (Mazumder et al., 2006).  $\text{CO}_2$  would diffuse through the heat shrinkable tube to the confining fluid if the lead foil was not in place. The apparatus is engineered to sustain a maximum pore pressure of 16.7 MPa and a maximum confining pressure of 25 MPa. The sample chamber and other parts of the apparatus are kept in the same constant temperature during the experiment. The temperature control device is not presented in Figure 1.

**Figure 1** Schematic diagram of the TPD testing device

The testing procedure started with testing leakage rate of the entire system, and installing the core sample into the pressure chamber, followed by applying confining pressure. The core sample was connected with a vacuum pump, for at least 24 hours to 1 week, in order to eliminate gas and water or moisture. Then gas was injected continuously into the core from both ends of the core by the injection pump through upstream cylinder and downstream cylinder simultaneously at a constant pressure  $P$ . After a period of time, usually several days, residual gas volume of the injection pump remains unchanged, it reveals that the adsorption of the sample reaches equilibrium. After that, the upstream and downstream tubing was separated by closing valve between injection pump and downstream cylinder. The pulse pressure was imposed to the core using the injection pump. Then, according to the time curve of pressure difference between upstream pump and downstream pump, the permeability can be calculated by use of the method described later.

If we change the injection pressure of upstream pump and downstream pump, we can measure permeability at different gas pressures.

## 2.2 Experimental conditions description

The raw coal samples were collected from Qinshui basin. The adsorption and fluid flow characteristics of coal from this basin have been widely investigated (Han et al., 2010a, 2010b, 2012; Shen et al., 2011). The raw sample was cored to about 25 mm in diameter and 50 mm in length (as shown in Figure 2). Meanwhile, the cutoffs from coal column ends were collected, crushed and ground to 60 to 80 mesh for coal petrology and proximate analyses. Vitrinite reflectance ( $R_{o,max}$ ) of the coal sample was 2.87%. The vitrinite, inertinite, exinite and mineral contents were 71.6%, 23.2%, 0% and 5.2%, respectively. The proximate analysis showed that moisture content of the sample is about 4.16%, ash content 13.11% and volatile content 7.32%. Three pure gases were used including  $CO_2$ ,  $N_2$  and  $CH_4$ . The purities of the three gases were all 99.995%. All measurements were conducted at a constant temperature of 40°C. In order to eliminate the impact of effective stress on permeability, all tests were carried out at a constant effective stress of 2 MPa, which were controlled by tracking the gas injection pressure.

**Figure 2** Photos of the coal core sample (see online version for colours)

### 2.3 The TPD permeability measurement method

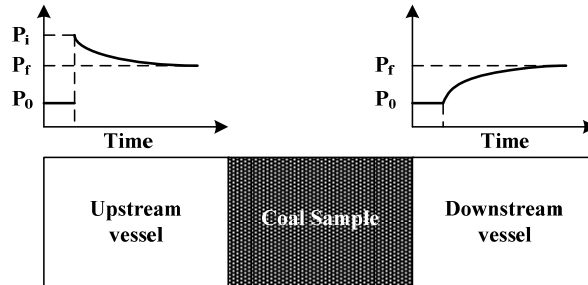
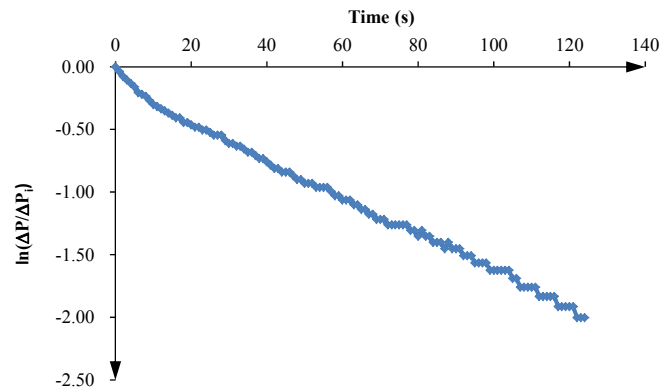
The TPD method was first proposed by Brace et al. (1968). The main advantage of this method is the shorter test duration required than steady-state methods. The TPD method involves observing the decay of a differential pressure between upstream and downstream cylinders across the sample. This pressure decay is combined with the cylinder volumes in the analysis to relate the flow through the sample and thus determine the permeability (Brace et al., 1968). A schematic diagram describing the principle of the TPD method is shown in Figure 3. The permeability ( $k$ ) of the sample can be calculated by equation (8) and (9):

$$\frac{\Delta P(t)}{\Delta P_i} = \exp(-\alpha t) \quad (8)$$

$$\alpha = \frac{kA}{\mu\beta L} \left( \frac{1}{V_u + V_d} \right) \quad (9)$$

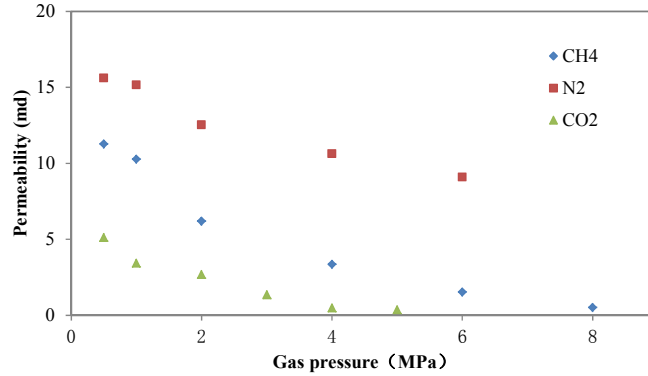
where  $\Delta P(t)$  is the pressure difference between the upstream cylinder and downstream cylinder, measured by a differential pressure transducer;  $\Delta P_i$  is the pressure difference between the upstream cylinder and downstream cylinder at the initial stage,  $t$  is time;  $k$  is permeability;  $A$  is the cross-sectional area of the core  $\mu$  is gas viscosity at the test condition, which is calculated from the NIST webbook at <http://webbook.nist.gov/chemistry/fluid/>;  $\beta$  is the compressibility of the fluid in the cylinder;  $L$  is the length of the core;  $V_u$  and  $V_d$  are the volumes of the upstream cylinder and downstream cylinder, respectively.

The calculation process including determining  $\alpha$  according to the log differential pressure vs. time curve (a typical time curve of differential pressure is shown in Figure 4). Then the permeability can be calculated by equation (8) and (9).

**Figure 3** Schematic diagram describing the principle of the TPD method**Figure 4** A typical time curve of differential pressure (see online version for colours)

### 3 Experimental results and discussion

Permeability measured results using  $\text{CH}_4$ ,  $\text{N}_2$  and  $\text{CO}_2$  with respect to gas pressure at a constant effective stress of 2 MPa are shown in Figure 5. It can be seen that, for the gas pressure range of 0.5 to 8 MPa, the permeability measured by  $\text{N}_2$  is higher than that measured by  $\text{CH}_4$  at the same gas pressure; the permeability measured by  $\text{CO}_2$  is the lowest. We can also see that permeabilities measured using  $\text{CH}_4$ ,  $\text{N}_2$  and  $\text{CO}_2$  all decrease with increasing gas pressure. This permeability decrease mainly attributes to coal cleat width decrease due to gas adsorption-induced coal matrix swelling since there is no impact of the effective stress, because of the constant effective stress condition. However, the permeability decrease shows some variations with gas species. Permeability measured by  $\text{CO}_2$ ,  $\text{CH}_4$  and  $\text{N}_2$  decrease almost 91%, 70% and 32% respectively, from gas pressure of 0.5 MPa to 4 MPa at the constant effective stress of 2 MPa. This is attributed to the different adsorption-induced swelling behaviour. Therefore, above results also reveal the highest adsorption-induced swelling effect with  $\text{CO}_2$  adsorption in coal.

**Figure 5** Permeability measured results using CH<sub>4</sub>, N<sub>2</sub> and CO<sub>2</sub> (see online version for colours)

## 4 Modelling of experimental results

### 4.1 Model selection

Robertson and Christiansen (2006) developed a permeability model that describes the permeability behaviour of a fractured, sorptive-elastic media, such as coal, under typical laboratory conditions where common radial and axial pressures are applied to core sample during permeability measurements. In Robertson's model, the expression for permeability based on a cubic geometry and hydrostatic confining pressure is as following:

$$k = \frac{b^3}{12a} \quad (10)$$

where  $k$  is permeability,  $a$  is fracture spacing,  $b$  is fracture width. Permeability is a much stronger function of the cleat width  $b$  than the matrix block width  $a$  because of the cubic exponent attached to  $b$ . A simplifying assumption is to consider only the changes in  $b$  due to changes in pressure. Taking the derivative of above equation with respect to pressure and letting  $b$  be a function of pressure results in the following equation:

$$\frac{dk}{dp} = \frac{3b^2}{12a} \frac{db}{dp} \quad (11)$$

The total change in fracture width  $\Delta b_t$  caused by changing pressure conditions is the sum of the change caused by fracture compressibility, mechanical elasticity, and sorption of gases, according to Robertson's derivation (Robertson and Christiansen, 2006),  $\Delta b_t$  can be expressed by following equation:

$$\Delta b_t = b_0 c_f (\Delta p_p - \Delta p_{ob}) + \frac{a_0(1-2\nu)}{E} \Delta p_p - \frac{a_0 S_{\max} p_L}{(p_L + p_{p0})(p_L + p_p)} \Delta p_p \quad (12)$$



where  $b_0$  is initial fracture width,  $p_p$  is pore gas pressure,  $p_{ob}$  is overburden pressure,  $a_0$  is initial fracture spacing,  $\nu$  is Poisson's ratio,  $E$  is Young's modulus,  $S_{\max}$  is a constant representing the strain at infinite pore pressure and the Langmuir pressure  $p_L$  is another constant representing the pore pressure at which the measured strain is equal to one-half  $S_{\max}$ .

Substituting equation (12) into equation (11) for  $\Delta b$  results in the following:

$$\frac{\Delta k}{\Delta p} = \frac{3b^2}{12a} \frac{1}{\Delta p} \left\{ \begin{array}{l} b_0 c_f (\Delta p_p - \Delta p_{ob}) + \frac{a_0(1-2\nu)}{E} \Delta p_p \\ - \frac{a_0 S_{\max} p_L}{(p_L + p_{p0})(p_L + p_p)} \Delta p_p \end{array} \right\} \quad (13)$$

In the case of our experimental work described in the previous section, the overburden pressure and pore pressure varied simultaneously, while keeping the different pressure constant. In this case, equation (13) reduces to the following form:

$$\frac{\Delta k}{\Delta p_p} = \frac{3b_0^2}{12} \left[ \frac{1-2\nu}{E} - \frac{S_{\max} p_L}{(p_L + p_{p0})(p_L + p_p)} \right] \quad (14)$$

Multiplying the right term in the above equation by unity ( $a/b_0 \cdot b_0/a$ ) results in:

$$\frac{\Delta k}{\Delta p_p} = \frac{3b_0^3}{12a} \frac{a}{b_0} \left[ \frac{1-2\nu}{E} - \frac{S_{\max} p_L}{(p_L + p_{p0})(p_L + p_p)} \right] \quad (15)$$

Recall that  $a/b = 3/\phi$  and that  $b^3/12a = k$ . Substituting these relationships into the above equation results in:

$$\frac{\Delta k}{\Delta p_p} = 3k \left[ \frac{3}{\phi} \left( \frac{1-2\nu}{E} - \frac{S_{\max} p_L}{(p_L + p_{p0})(p_L + p_p)} \right) \right] \quad (16)$$

Rearranging this equation to fit a form capable of integration and letting  $\Delta p$  approach zero results in:

$$\frac{dk}{3k} = \frac{3}{\phi} \frac{1-2\nu}{E} dp_p - \frac{3}{\phi} \frac{S_{\max} p_L}{(p_L + p_{p0})(p_L + p_p)} dp_p \quad (17)$$

The permeability equation can now be integrated and results in the following equation:

$$\frac{k}{k_0} = e^{3 \left\{ \frac{3}{\phi} \left[ \frac{1-2\nu}{E} (p_p - p_{p0}) - \frac{S_{\max} p_L}{(p_L + p_{p0})} \ln \left( \frac{p_L + p_p}{p_L + p_{p0}} \right) \right] \right\}} \quad (18)$$

## 4.2 Input parameters

Table 1 is a list of required model input parameters, which were determined by previous study (Fang and Li, 2012). The values of initial porosity, Young's modulus and Poisson's ratio were measured for samples cored from the same batch of raw coal used in this study. Langmuir-type parameters for sorption-induced strain ( $S_{\max}$  and  $p_L$ ) were estimated

from the adsorption, sorption-induced strain, and permeability simultaneously measurement experiment, using a coal sample also from the same batch of raw coal.

**Table 1** Input parameters required for Robertson and Christiansen permeability model

Input parameters	Value		
	CH <sub>4</sub>	CO <sub>2</sub>	N <sub>2</sub>
Initial porosity ( $\phi_0$ ), dimensionless	0.03	0.03	0.03
Young's modulus ( $E$ ), MPa	4000	4000	4000
Poisson's ratio ( $\nu$ ), dimensionless	0.40	0.40	0.40
Langmuir curve-match for shrinkage ( $p_L$ ), MPa	4.72	3.05	6.25
Langmuir curve-match for shrinkage ( $S_{\max}$ ), dimensionless	0.004632	0.006527	0.003344

### 4.3 Modelling results and discussion

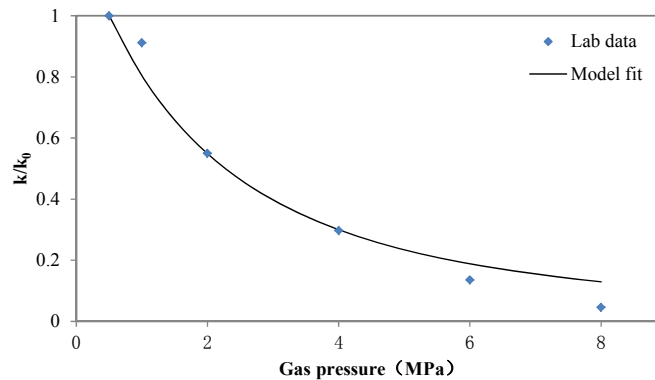
Using the values of input parameters given in Table 1, the variation in permeability for increasing gas pressure was estimated using the Robertson and Christiansen model. The modelled results, along with the experimental results for CH<sub>4</sub>, N<sub>2</sub>, and CO<sub>2</sub>, are shown in Figures 6, 7, and 8, respectively.

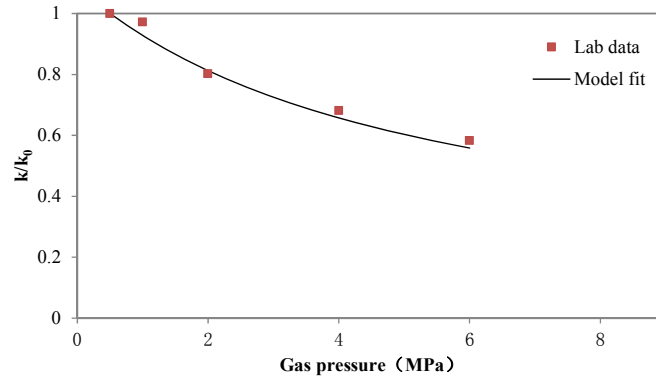
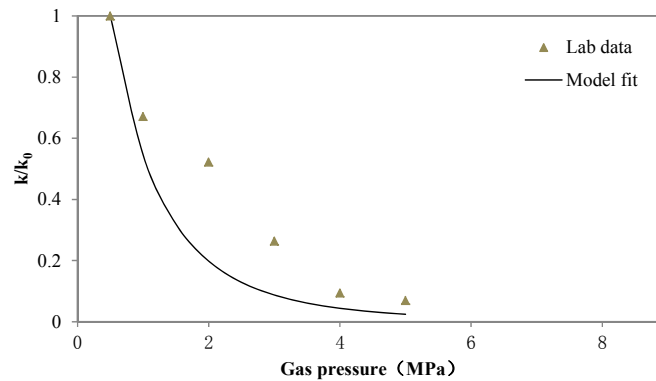
As can be seen from Figure 6, the agreement between the modelled results and the experimental results is good for CH<sub>4</sub>, especially in the pressure range of 0.5 MPa to 4 MPa. As gas pressure is greater than 4 MPa, the modelled results begin to deviate from the experimental results.

Figure 7 shows that the agreement between the modelled results and the experimental results is good for N<sub>2</sub> in the pressure range of the experiments, which is similar with CH<sub>4</sub>.

However, the model overestimates the permeability reduction of CO<sub>2</sub>, especially in the pressure range of 1 MPa to 4 MPa, as shown in Figure 8. The reason may be that in the modelling process, we supposed that the strength of coal kept constant with gas adsorption, however, some studies results, for example, Viete and Ranjith (2006), showed that the adsorption of CO<sub>2</sub> on coal causes a decrease in the coal strength. Thus, coal weakening by the introduction of CO<sub>2</sub> to a coal seam may induce fracturing, causing a permeability increase under in situ conditions. Therefore, the Robertson's model may overestimate the permeability reduction of coal caused by CO<sub>2</sub> adsorption.

**Figure 6** Measured and modelled permeability variation – CH<sub>4</sub> (see online version for colours)



**Figure 7** Measured and modelled permeability variation – N<sub>2</sub> (see online version for colours)**Figure 8** Measured and modelled permeability variation – CO<sub>2</sub> (see online version for colours)

## 5 Conclusions

This paper has presented a new apparatus developed for measuring coal permeability with various gases adsorption, by use of the TPD technique. The effects of gas adsorption-induced coal swelling on coal permeability has been studied via a series of measurements of gas permeability of a coal core sample from Qinshui basin, using three gases including CO<sub>2</sub>, N<sub>2</sub> and CH<sub>4</sub>.

Based on the results obtained, we can see that permeability measured using CH<sub>4</sub>, N<sub>2</sub> and CO<sub>2</sub> all decrease with increasing gas pressure. This mainly attributes to coal cleat width decrease due to gas adsorption-induced coal matrix swelling since there is no impact of the effective stress, because of the constant effective stress condition. The permeability decrease shows some variations with gas species. Permeability measured by CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub> decrease almost 91%, 70% and 32% respectively, from gas pressure of 0.5 MPa to 4 MPa at the constant effective stress of 2 MPa. This is attributed to the different adsorption-induced swelling behaviour.

The experimental results are then modelled using the Robertson and Christiansen permeability model which is commonly used during measurement of permeability data in the laboratory. Model results show the agreement between the modelled results and the experimental results in the pressure range of the experiments, especially for CH<sub>4</sub> and N<sub>2</sub>.

### Acknowledgements

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