A Dynamic Multi-component Transport Model for CO2 Enhanced Coalbed Methane Recovery

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ABSTRACT

This paper presents a dynamic multi-component transport (DMCT) model for better understanding and prediction of CO2-enhanced coalbed methane (CO2-ECBM) recovery. This model is a coupled system dealing with different processes: gas sorption equilibrium; gas diffusion and flow; and matrix shrinkage and swelling. The gas diffusion model was developed based on the bidisperse diffusion mechanism and the Maxwell-Stefan (MS) diffusion theory, which provides an improved simulation of multi-component gas diffusion dynamics. The multi-component sorption equilibria was determined using ideal adsorbed solution-real gas (IAS-RG) approach coupled with Langmuir isotherm. The variations of fracture porosity and permeability with effective stress were considered in the simulation using the Shi & Durucan (SD) model. Experimental data obtained from a series of core flush tests was employed to examine the predictive capability of the proposed DMCT model. The simulations show good agreements with the core flush experiments, and the predicted micro- and macro-pore diffusivities are comparable with measurements achieved by other researchers. These diffusivities were then used in triple-component gas diffusion for simulating flue gas injection enhanced coalbed methane (FG-ECBM) recovery. The calculated results suggest that the CH4 recovery profiles from the mixed gas injections lay between the recovery lines of pure N2 and pure CO2 injections. The mixed gas injections have the same breakthrough time as with pure N2 injection. However, injection gas composition has a significant effect on the produced gas composition.

INTRODUCTION

Carbon dioxide geosequestration in coalbeds for enhanced coalbed methane (CO2-ECBM) recovery has recently attracted growing attention due to increasing concerns about greenhouse gases and the emerging commercial significance of coalbed methane production all over the world. While considerable efforts have been made in modeling the gas sorption kinetics for CBM/ECBM production, significant limitations exist in current models. One predominant deficiency is that most of the currently available CBM/ECBM models are based on a single-component unipore diffusion formulation. These models are not suitable to simulate the simultaneous counter flow of CO2 into, and CH4 out of, the coal matrix. A further complication is the wide range in the scale of the pore spaces typically occurring in coal.

The pore structure of coal is highly heterogeneous, and it depends on the coal type and rank [1, 2]. According to International Union of Pure and Applied Chemistry (IUPAC) classification [2], pores can be classified into macropores (>50nm), transient or mesopores (2nm~50nm) and micropores (<2nm). The studies of Gamson et al [4] suggest that gas transport in coal seams, in terms of coal type and rank, can be classified into two types: a two-step transport process (viscous flow followed by diffusion) and a multi-step transport process. The multi-step process consists of gas diffusion in micropores; gas diffusion through partly blocked microfractures, which may consist of mesopores and macropores; gas flow through open, un-mineralized microfractures; and then viscous gas movement through main cleats.
So far the work found in open literature primarily focuses on the adsorption capacity and pays less attention to the
gas diffusion dynamics, especially multi-component gas diffusion dynamics. Three types of models are currently
available to simulate gas diffusion in coals: equilibrium model, non-equilibrium model and bidisperse diffusion
model. The equilibrium and non-equilibrium models basically treat the gas diffusion process as an instantaneous or
one-step diffusion [5, 6]. The non-equilibrium model is now used in most commercial CBM simulators to simulate
gas sorption kinetics, which can be subcategorized into two approaches: the pseudo-steady state approach [7]
and unsteady state approach [5, 6, 8]. The former is only accurate once the concentration gradients in the reservoir
have stabilized, after an extended period, and is less suitable for early stages of production [9, 10]. The
later accounts for the effects of the gas concentration gradient, which seems a more rigorous approach, but at the
cost of significantly more numerical burden. The bidisperse diffusion model [11-14] assumes two-step gas
diffusion in a coal matrix: surface diffusion in the microporous system and pore diffusion in the meso/macropore
system. We believe that gas diffusion in coals with multiscale pores can be described better by a bidisperse
diffusion model.

Fick’s law of diffusion is a widely used method to describe the coalbed gas diffusive process because of its
simplicity. However, its validity is severely restricted mostly in large pores and likely to be misleading in many
practical situations where the diffusion coefficients of the fluid species depend on composition. The Maxwell-
Stefan (MS) diffusion formulation applies to multi-component fluid diffusion and deals rigorously with the
interactions between multi-component gas molecules. It is therefore more appropriate than Fick’s law for
describing multi-component gas diffusion dynamics and consequently should be applied in the CBM/ECBM
models.

Cleat permeability is recognized as one of the most important parameters for coalbed methane production. Both
experimental and theoretical research indicate the permeability of coal fractures decreases exponentially with
increasing net stress [15-17]. The effect of matrix shrinkage on cleat porosity and permeability has been studied
and has led to several predictive permeability models [17-21].

There are two principal variations of ECBM recovery, namely with N₂ injection and CO₂ injection; each process
uses a distinct mechanism to enhance methane desorption and production. Several problems appear during the
pure CO₂ or N₂ injection-ECBM recovery, such as the high cost of separating CO₂ from flue gas for pure CO₂
injection scenario or low incremental reserves of CBM for pure N₂ injection scenario. Therefore, it is necessary to
better understand the ECBM recovery process performed by flue gas injection, towards improving practical
methane productivity and recovery, yet minimizing costs by foregoing CO₂ separation.

This paper presents a dynamic multi-component transport (DMCT) model to address the issues discussed above.
The model adopts a bidisperse pore structure in the coal matrix based on the approach proposed by Ruckenstein
et al [22]. The concentration-dependent micropore diffusivity is described with a polynomial and estimated by
comparing the model prediction with experimental data. The diffusion dynamics of mixed gases is simulated using
MS diffusion formulation. The permeability-stress model proposed by Shi & Durucan [23,24] was employed in
simulation to account for the stress-dependent porosity and permeability. The DMCT model is used to simulate
the mixed gas diffusion and viscous flow in the bulk coal for CO₂-ECBM and FG-ECBM recovery processes.

MATHEMATICAL MODEL

The following assumptions are used to model gas diffusion and flow in the coal matrix:

I. The coal matrix is treated as a cylindrical cell surrounded by main fractures. It contains particles with
uniform radius, between which are open microfractures (Fig. 1a);

II. The particles have bimodal pore structure, containing uniform radius microporous microparticles with the
space between microparticles making up the meso/macroporosity (Fig. 1b);

III. Gas flow through open microfractures and cleats is assumed to be viscous flow obeying Darcy’s law (water
phase is not included in the model);

IV. Gas diffusion in the coal matrix is assumed to be bidisperse pore diffusion: surface diffusion in micropores
and bulk and Knudsen diffusions in meso/macropores;

V. The micropore size distribution is ignored and the micropore size within a coal sample is assumed to be
homogeneous; and

VI. The equilibrium between the gas phase and adsorbed phase is instant.
With the above assumptions, three levels of mass balance equations can be developed for the three levels of pore sizes: open microfractures, meso/macropores and micropores.

Gas Flow in Open Microfractures

The mass balance equations for gas flow in the microfractures can be simplified as 1-dimensional Darcy flow, giving

\[ \varepsilon_c \frac{\partial(C_{ci})}{\partial t} - \frac{\partial(C_{ci} u_g)}{\partial x} + J_{m-c,i} = 0 \ (i = 1, 2, \ldots, nc) \]  

where \( C_{ci} \) is gas concentration in fractures of the gas component \( i \); \( nc \) is number of components; \( u_g \) is gas velocity depending on permeability and pressure gradient along the coal matrix cell; \( \varepsilon_c \) is fracture porosity; and \( J_{m-c,i} \) is mass exchange rate between fractures and particles per unit volume of bulk coal.

The values of fracture porosity and permeability may vary in course of gas injection/production due to coal matrix shrinkage and swelling. To evaluate the variations of these parameters under a condition of the stressed sorption, the Shi-Durucan (SD) model [22, 23] was utilized to simulate the stress-dependent permeability and porosity for multi-component system. In this model, the change in effective horizontal stress is given by

\[ \sigma - \sigma_0 = -\frac{V}{1-V} (p - p_0) + \frac{E}{3(1-V)} \sum_{j=1}^{n} \alpha_j (C_{jg} - C_{jg0}) \]  

where \( \sigma \) is effective horizontal stress; \( V \) is Poisson ratio; \( E \) is Young's modulus; \( \alpha_j \) is matrix shrinkage/swelling coefficient of component \( j \); \( p \) is pore pressure. Using the extended Langmuir isotherm, the adsorbed volume for gas component \( j \) may be approximated by

\[ C_{jg} = \frac{V_{lj} p y_j b_j}{1 + p \sum_j y_j b_j} \]  

where \( V_{lj} \) is Langmuir volume of component \( j \); \( y_j \) is the mole fraction of component \( j \) in gas phase; \( b_j \) is Langmuir constant of component \( j \). The fracture porosity \( \varepsilon_c \) varies exponentially with change in the effective horizontal stress, as follow

\[ \varepsilon_c = \varepsilon_{c0} e^{-c_f (\sigma - \sigma_0)} \]  

where \( \varepsilon_{c0} \) is initial fracture porosity; \( c_f \) is cleat-volume compressibility. The fracture permeability can be derived from the following relationship

\[ \frac{k}{k_0} = \left( \frac{\varepsilon_c}{\varepsilon_{c0}} \right)^3 \]  

where \( k \) and \( k_0 \) are fracture permeability and initial fracture permeability, respectively.

Diffusion in Meso- and macro-pores Within a Particle

Multi-component gas diffusion in meso- and macro-pores within a particle can be described by

\[ \varepsilon_m \frac{\partial(C_{mi})}{\partial t} = - \frac{1}{R^2} \frac{\partial}{\partial R} \left( R^2 N_{m,i} \right) - J_{m-\mu,i} \ (i = 1, 2, \ldots, nc) \]  

with initial and boundary conditions

\[ t = 0: \ C_{mi} = C_{mil0} \]
\[ R = 0: \quad \frac{\partial C_{mi}}{\partial R} = 0 ; \quad R = R_m: \quad C_{mi} = C_{ci} \]  

where \( \varepsilon_m \) is macroporosity; \( N_{m,i} \) is pore diffusion flux in the particle; and \( J_{m,i} \) is mass exchange rate between micropores and meso/macropores. The pore diffusion flux is defined as

\[
[N_m] = -[B_m^e]^{-1} \frac{\partial[C_m]}{\partial R} = -[L_m] \frac{\partial[C_m]}{\partial R} \]  

where \([L_m]\) is the macropore diffusivity matrix and defined by the inverted matrix of inverted MS effective diffusivity \([B_m^e]\). The \([B_m^e]\) consists of the following elements [25]

\[
B_m^e(i, j) = \begin{cases} 
\frac{1}{D_{miM}^e} + \sum_{k=1}^{nc} \frac{y_i}{D_{mik}^e} & (i = j; i = 1, 2, \ldots, nc) \\
- \frac{y_i}{D_{mij}^e} & (i \neq j; i = 1, 2, \ldots, nc)
\end{cases} \]  

where \( D_{miM}^e \) is effective Knudsen diffusivity of component \( i \); \( y_i \) is mole fraction of component \( i \); and \( D_{mij}^e \) is effective binary molecular diffusivity between species \( i \) and \( j \).

**Diffusion in Micropores Within a Microparticle**

Gas diffusion in micropores within a microparticle can be described by

\[ \varepsilon_\mu \frac{\partial C_\mu}{\partial t} = -\frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 N_{\mu,i} \right) \quad (i = 1, 2, \ldots, nc) \]  

with initial and boundary conditions

\[ t = 0: \quad C_\mu = C_{\mu 0} \quad (r < r_\mu) \]  

\[ r = 0: \quad \frac{\partial C_\mu}{\partial r} = 0 \quad (i = 1, 2, \ldots, nc) \]  

\[ r = r_\mu: \quad C_\mu = I_\mu(P) \quad (i = 1, 2, \ldots, nc) \]

where \( I_\mu(P) \) is the adsorption equilibrium which can be estimated by a sorption isotherm model; \( \varepsilon_\mu \) is the microporosity; \( N_{\mu,i} \) is the surface diffusion flux in the microparticle. According to the Maxwell-Stefan theory, micropore diffusion flux can be represented as

\[
[N_\mu] = -[B_\mu]^{-1} [\Gamma] \frac{\partial[C_\mu]}{\partial r} = -[L_\mu] \frac{\partial[C_\mu]}{\partial r} \]  

The matrix of inverted MS micropore diffusivities \([B_\mu]\) has the elements
\[
B_{\mu}(i, j) = \begin{cases} 
\frac{1}{D_{iV}} + \sum_{k=1}^{nc} \frac{\theta_{i}}{D_{ik}} & (i = j; i = 1, 2, \ldots, nc) \\
- \frac{\theta_{i}}{D_{ij}} & (i \neq j; i = 1, 2, \ldots, nc)
\end{cases}
\]

(16)

where \(\theta_{i}\) is fractional surface coverage; \([\Gamma]\) is matrix of thermodynamic factors; \(D_{iV}\) and \(D_{ij}\) are MS micropore diffusivity of component \(i\) and MS counter diffusivity between components \(i\) and \(j\), respectively.

### NUMERICAL TECHNIQUE

A loose coupling algorithm is adopted in this work for numerically solving the highly coupled model equations, as illustrated in Fig. 2. Such a numerical technique has been used in simulation of coupled processes with high accuracy [26]. Our case is a coupled system dealing with different processes, i.e., gas sorption equilibrium, gas diffusion and Darcian flow, and matrix shrinkage and swelling. Among these processes, the gas diffusion and flow are central, which is described by Eqs. (1), (6) and (11). These equations will be discretized and solved independently and then coupled with the sorption equilibrium of the gases and matrix shrinkage/swelling behavior. The multi-component sorption equilibria was determined using ideal adsorbed solution-real gas (IAS-RG) approach with Langmuir isotherm [27] in this study; and coal matrix shrinkage and swelling can be estimated by Eqs. (2) to (5). Both of these two processes are easily coupled with the core model.

### MODEL RESULTS AND DISCUSSION

#### CO\(_2\) Enhanced CBM Recovery

The experimental data obtained in our lab were used to validate the DMCT model and investigate the diffusivities of mixed gases in coals. The experiments were core flush tests where CO\(_2\) or He was injected into the coal sample to displace pre-adsorbed methane or carbon dioxide, respectively. The sample tested was originated from the open cut Blackwater Coal Mine in the central area of the Bowen Basin, Australia. The sample is an 8×8×8 cm cube, moisture in the sample is about 5%, helium density is around 1285 kg/m\(^3\) and the vitrinite random reflectance (Vro) ranges from 0.88 to 0.89, making it a high volatile bituminous coal. Total porosity of the coal sample is 11.22%. As aforementioned, three types of porosities are defined in the model to describe the multi-scale porosity system: fracture porosity (\(\varepsilon_{c}\)), macroporosity (\(\varepsilon_{m}\)) and microporosity (\(\varepsilon_{\mu}\)). According to previous research [17, 28, 29], the total porosity of coal can be apportioned 11% to fractures, 36% to meso- and macropores and 53% to micropores. The parameters adapted from the tests and used for this study are listed in Table 1.

#### Determination of kinetic parameters

The binary molecular diffusivities between different species were estimated using Fuller correlation [30], and Knudsen diffusivities of each species were calculated using the correlation presented by Jackson [31]. According to the Bosanquet approximation [31], the macropore diffusivity of a given species can be estimated from its Knudsen diffusivity and the binary molecular diffusivity of the associated binary gas. The values of the kinetic parameters calculated under the conditions of three stages are shown in Table 2.

In this work, the optimization of MS micropore diffusivity \(D_{iV}\) is simplified by fitting to the experimental data with a postulated single-component MS diffusion equation with a concentration-dependent MS micropore diffusivity

\[
D_{i} = \frac{D_{iV}(\theta_{i})}{1-\theta_{i}} = \frac{D_{iV}(0)}{1-\theta_{i}} \left( \sum_{i=1}^{N_{0}} b_{i} \theta_{i}^{i-1} \right)
\]

(17)

where \(\theta_{i}\) is total surface occupancy; \(b_{i} (i = 1, 2, \ldots, N_{0})\) and \(D_{iV}(0)\) is MS micropore diffusivity at zero surface coverage, which can be determined by fitting the calculated results to the experimental data. The optimized \(D_{iV}(0)\) of CO\(_2\) at 0.4 MPa is approximately \(8 \times 10^{-10} \text{ m}^2/\text{s}\).
Model validation: Fig. 3 shows the calculated gas composition in produced gas using the model comparing with the experimental data for stages 17 and 29 respectively. It can be seen from this figure that the simulated results fit the experimental data very well.

Predicted gas diffusivities: Fig. 4a shows the effective macropore diffusivities of each component versus gas composition in gas phase, predicted by MS diffusion model under the conditions of stage 17. The results indicated that the effective meso/macropore diffusivities of CH$_4$ and CO$_2$ gradually increase with increasing mole fraction of the corresponding component in the gas phase, respectively. The effective micropore diffusivities of CH$_4$ and CO$_2$ in this binary gas system are functions of the surface mole fraction of CH$_4$. It can be seen that both CH$_4$ diffusivity and the binary diffusivity of CH$_4$-CO$_2$ increase with surface mole fraction of CH$_4$, while CO$_2$ diffusivity and binary diffusivity of CO$_2$-CH$_4$ decrease with surface mole fraction of CH$_4$. Moreover, the simulations indicate that CH$_4$ micropore diffusivity is about 10 times less than CO$_2$ micropore diffusivity. The predicted CH$_4$ effective micropore diffusivities are consistent with experimental measurements [32].

Flue Gas Enhanced CBM Recovery

A series of three-component (CH$_4$, CO$_2$ and N$_2$) gas diffusion and flow simulations were performed to assess the behavior of a coal sample under a range of injection gas compositions. The coal properties have assumed to be the same as those considered in the last section and the thermodynamic properties of these components are summarized in Table 3. It is further assumed that the injection is operated at a constant injection pressure and at a constant injection composition. In these calculations, mobile liquid is not considered. Mixtures of CH$_4$, CO$_2$ and N$_2$ are used to represent injection gases. Several cases were designed to analyze the effects of injection gas composition on the gases diffusion and flow behavior. Five injection compositions were considered containing 0, 25, 50, 75, and 100% mole fraction of CO$_2$, with the remainder N$_2$.

Effects of injection gas composition on methane recovery: Fig. 5 shows the calculated CH$_4$ recovery for all these cases, respectively. The methane recovery is defined as the volumetric percent of recovered CH$_4$ to initial CH$_4$ in coal sample. It can be seen from the figure that all mixed gas injections are able to achieve 100% methane recovery and they have the similar profiles of methane recovery. However, a maximum difference of 15% is found between the mixture with N$_2$ of 25% and 75% that occurred at around 15 mins after gas injection. The N$_2$-enriched injection gas yields a relatively slow desorption of methane from the coal, and thus a lower methane recovery (before 60 mins), while the CO$_2$-enriched injection gas gives a fast methane desorption and a higher methane recovery (also before 60 mins), as indicated in Fig. 5. Therefore, the mixtures of N$_2$ and CO$_2$ greatly reduce the methane recovery, which is consistent with the experimental results obtained by Reznik et al [33].

The methane recovery profiles from the mixed gas injections are all similar to the profile for pure N$_2$ injection, especially the mixture with N$_2$ of 75%. This suggested that N$_2$, regardless of its concentration in the mixture, is the governing component that controls the methane recovery. The initial methane recovery by pure CO$_2$ injection is lower than the mixed gas injections and pure N$_2$ injection, while methane desorption by CO$_2$ injection increases very fast and is in the lead after about 2 mins of gas injection. The reason for this phenomenon is the higher capacity of CO$_2$ adsorption on coal surface than that of CH$_4$. The loss of injected CO$_2$ volume is greater than the volume of the CH$_4$ released from the coal surface. Therefore, it would take longer time to inject enough amount of CO$_2$ to replace methane at the initial time. After that, the methane desorption by CO$_2$ injection is larger than the amounts by mixed gas and N$_2$ injections, and thus the methane recovery profile from the mixture injections lay between the recovery lines of pure N$_2$ injection and pure CO$_2$ injection.

Effects of injection gas composition on methane mole fraction in produced gas: Fig. 6 shows the calculated mole fraction of CH$_4$ in produced gas. As the CO$_2$ is injected to the coal sample, it is rapidly adsorbed by the coal so that only a small amount of CO$_2$ can remain in the free phase. Therefore, the breakthrough time of pure CO$_2$ injection is longer than other cases. Here, the breakthrough time is defined as the time when the mole fraction of injected gas in produced gas is up to 1%. In the case of pure CO$_2$ injection, the concentration of CH$_4$ decrease sharply after CO$_2$ breakthrough in produced gas because methane desorption is faster than other cases. Moreover, the injections are assumed to be operated at a constant injection pressure, which yields higher injection rates for pure CO$_2$ injection. Therefore, the mole fraction of CO$_2$ in produced gas decrease faster than
other cases. The breakthrough times of mixed gas injections are equal to the breakthrough time of pure N₂ injection. Since the methane desorption rates of mixed gas injections decrease in the order of the mixture with N₂ of 25% > 50% > 75% (Fig. 5), the methane mole fraction in produced gas for the injection gas with 25% N₂ is highest among the three cases until the model runs about 10 mins, and then it decreases faster than other cases.

Effects of injection gas composition on N₂ adsorption: Fig. 7 shows the calculated amounts of N₂ adsorption on coal for pure N₂ injection and mixed gas injections, respectively. The DMCT model results show that none of the nitrogen is adsorbed by the coal when significant CO₂ (≥25%) is present in the injection gases.

Effects of injection gas composition on breakthrough times of gases: Figs. 8a and 8b depict the effects of injection gas composition on breakthrough times of nitrogen and carbon dioxide, respectively. It can be seen from Fig. 8a that all of mixed gas injections have the same breakthrough time of N₂, as with the pure N₂ injection. Fig. 8b shows that the mixed gas injection with CO₂ of 75% has the shortest breakthrough time of CO₂ among mixed gas injection cases, and the breakthrough time of CO₂ is more extended as its concentration in the injected gas mixture is decreasing.

CONCLUSIONS

A dynamic multi-component transport (DMCT) model has been developed to simulate diffusion and flow of mixed gases in coals for CO₂ sequestration and enhanced methane production. The Maxwell-Stefan diffusion formulation was used to model the counter-diffusion of mixed gases in coals. The model has been validated in simulations of CO₂-CH₄ counter-diffusion, showing excellent agreement with experimental measurements. Several conclusions can be made from the simulations of flue gas injection ECBM production: (1) When CO₂ is present in the injected gas, none of N₂ is adsorbed by the coal. Regardless of its concentration in the mixture, N₂ is the governing component that controls the methane recovery. (2) The CH₄ recovery profiles from the mixture injections lie between the recovery lines of pure N₂ and pure CO₂ injections, and mixtures of N₂ and CO₂ greatly reduce the recovery of CH₄. (3) All of the mixture gas injections have the same breakthrough time as with the pure N₂ injection; while CO₂ breakthrough time is more extended as its concentration in the injected gas mixture is decreasing. (4) Injection gas composition has significant effect on the produced gas composition.

NOMENCLATURE

- \( B_m^e \) matrix of inverted Maxwell-Stefan effective macropore diffusivities, s/m²
- \( B_\mu^e \) matrix of inverted Maxwell-Stefan micropore diffusivities, s/m²
- \( C_{ij} \) mole concentration of gas component i in open microfractures, mol/m³
- \( C_{mil} \) mole concentration of gas component i in macropores, mol/m³
- \( C_{mil0} \) initial gas mole concentration of component i in macropores, mol/m³
- \( C_t \) total concentration, mol/m³
- \( C_{\mu i} \) mole concentration of component i in micropores, mol/m³
- \( C_{\mu i0} \) initial mole concentration of component i in micropores, mol/m³
- \( c_m \) matrix shrinkage compressibility due to methane desorption, 1/Pa
- \( c_\mu \) differential swelling coefficients of component i
- \( D_{em0} \) initial effective macropore diffusivity, m²/s
- \( D_{eg0} \) initial effective micropore diffusivity, m²/s
- \( D_{iniM} \) Knudsen diffusivity of component i, m²/s
- \( D_{mij} \) molecular diffusivity of binary pair i-j, m²/s
- \( D_{ij}^* \) Maxwell-Stefan micropore diffusivity of component i, m²/s
- \( D_{ij}^* \) Maxwell-Stefan micropore counter diffusivity between components i and j, m²/s
Young's modulus, Pa

adsorption isotherm

dimensionless adsorption isotherm

mass exchange rate between cleats and bulk coal per unit volume of coals, mol/(m²·s)

mass exchange rate between micropores and macropores, mol/(m²·s)

fracture permeability, mD

initial fracture permeability, mD

matrix of macropore diffusivities, m²/s

matrix of micropore diffusivity, m²/s

matrix of molar fluxes in particles, mol/(m²·s)

matrix of molar fluxes in microparticles, mol/(m²·s)

number of components in the system

pressure, Pa

Langmuir pressure, Pa

particle radial coordinate, m

radius of particle, m

microparticle radial coordinate, m

radius of microparticle, m

velocity of gas phase, m/s

Langmuir volume of component j, cc/g

mole fraction of component i in gas phase

matrix shrinkage/swelling coefficient of component j

ratio of diffusion rates in the micropores to that in the macropores

matrix of thermodynamic factors

fracture porosity and initial fracture porosity, respectively

macroporosity and microporosity, respectively

fractional surface occupancy of species i

Poisson ratio

effective stress, Pa

ACKNOWLEDGMENTS

This work was supported by the Australian Research Council (ARC), Australian Postgraduate Award (APA) and CSIRO PhD scholarship.

REFERENCES

17. Massarotto P., 2002: "4-D coal permeability under true triaxial stresses and constant volume conditions", Brisbane, PhD thesis.
Table 1 Conditions and results of core flush tests.

<table>
<thead>
<tr>
<th>Conditions/results</th>
<th>Stage 17</th>
<th>Stage 29</th>
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<tr>
<td>Test</td>
<td>CO₂ displaces CH₄</td>
<td>He displaces CO₂</td>
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<td>Adsorbed pressure (MPa)</td>
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<td>Mean pore pressure (MPa)</td>
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<td>0.40</td>
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<td>Displacing time (hours)</td>
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<td>Total displaced gas (NL)</td>
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<td>Gas in connection tube (NL)</td>
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<td>Free gas in coal sample (NL)</td>
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<td>Desorbed gas (STP, NL)</td>
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<td>Initial adsorbed gas (STP, NL)</td>
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<td>Variation of effective stress (MPa)</td>
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<td>4.1 → 3.2</td>
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<td>Variation of permeability (mD)</td>
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<td>1.7 → 3.6</td>
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Table 2 Knudsen, Molecular and macropore diffusivities used for simulation.

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<th>Pore pressure (MPa)</th>
<th>Species</th>
<th>Knudsen diffusivity (×10⁻⁶ m²/s)</th>
<th>Molecular diffusivity (×10⁻⁶ m²/s)</th>
<th>Macropore diffusivity (×10⁻⁶ m²/s)</th>
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Table 3 Knudsen, Molecular and macropore diffusivities used for simulation.

<table>
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<th></th>
<th>$P_c$ (atm)</th>
<th>$T_c$ (K)</th>
<th>$V_c$</th>
<th>$Z_c$</th>
<th>$\Omega$</th>
<th>$V_L$ (cc/g)</th>
<th>$P_L$ (atm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CH$_4$</td>
<td>44.52</td>
<td>190.6</td>
<td>99.0</td>
<td>0.288</td>
<td>0.008</td>
<td>24.0</td>
<td>20.00</td>
</tr>
<tr>
<td>CO$_2$</td>
<td>71.42</td>
<td>304.1</td>
<td>93.9</td>
<td>0.274</td>
<td>0.225</td>
<td>35.0</td>
<td>6.67</td>
</tr>
<tr>
<td>N$_2$</td>
<td>32.80</td>
<td>126.2</td>
<td>89.8</td>
<td>0.290</td>
<td>0.04</td>
<td>0.44</td>
<td>18.14</td>
</tr>
</tbody>
</table>

Fig. 1. Conceptual model for multi-porosity system: a): a cylindrical coal matrix; b): a particle showing bimodal pore structure.
Fig. 2. The structure of numerical model.

Fig. 3. Calculated CH₄ molar fraction in produced gas for stage 17 and 29.
Fig. 4. Calculated results for stage 17. a): The effective meso/macropore diffusivities; b): The effective micropore diffusivities.

Fig. 5. Effects of injection gas composition on methane recovery.
Fig. 6. Effects of injection gas composition on methane mole fraction in produced gas.

Fig. 7. Effects of injection gas composition on N₂ adsorption.
Fig. 8. Effects of injection gas composition on breakthrough times: a) for N₂; b) for CO₂.