Analytical modeling of CO\textsubscript{2} storage and enhanced coal bed methane recovery

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Abstract

Analytical solutions are reported for one-dimensional, two-phase, three- and four-component flow in a coal bed with adsorption and volume change as components adsorb and desorb. Mixtures of N\textsubscript{2}, CH\textsubscript{4}, CO\textsubscript{2} and H\textsubscript{2}O are used to represent an enhanced coal bed methane recovery process (ECBM). N\textsubscript{2} and CO\textsubscript{2} propagate differently through a coal bed. CO\textsubscript{2} preferentially adsorbs onto the coal surface, resulting in a self-sharpening composition variation, while N\textsubscript{2} adsorbs more weakly than CH\textsubscript{4}, which causes a spreading composition wave to form. When mixtures of CO\textsubscript{2} and N\textsubscript{2} are injected, the displacement exhibits both shock and spreading wave features.

Volume reduction as CO\textsubscript{2} adsorbs delays breakthrough of injected gas. When N\textsubscript{2} is injected, flowing vapor volume increases as CH\textsubscript{4} desorbs, which results in earlier recovery of CH\textsubscript{4}. In CO\textsubscript{2} injection, complete separation of CO\textsubscript{2} from CH\textsubscript{4} occurs, while in N\textsubscript{2} injection, the produced gas is a mixture of N\textsubscript{2} and CH\textsubscript{4}, requiring separation of CH\textsubscript{4} from N\textsubscript{2}.

Keywords: ECBM, flue gas, sequestration, analytical solutions

Introduction

Injection of CO\textsubscript{2} into deep, unminable coal seams is an option for geological storage of CO\textsubscript{2}. ECBM is a potentially attractive technology for greenhouse gas mitigation because CO\textsubscript{2} is sequestered, while CH\textsubscript{4} recovery is enhanced, partially offsetting the costs for CO\textsubscript{2} injection and increasing production of a less carbon intensive fuel than coal or oil. Adsorption of gas on the internal surfaces of the coal is the storage mechanism. In many coal beds, large amounts of CH\textsubscript{4} are adsorbed onto the coal. CO\textsubscript{2} adsors more strongly on coal than does CH\textsubscript{4}, and hence injected CO\textsubscript{2} causes CH\textsubscript{4} to desorb as it is replaced by CO\textsubscript{2}. This offers the possibility of desorption and increased recovery of CH\textsubscript{4} from coal while minimizing the pressure reduction in the coal bed. Understanding the complex interplay of adsorption, phase behavior, and convection is important to development of efficient and physically accurate techniques for predicting the fate of injected CO\textsubscript{2} in the subsurface and designing effective CO\textsubscript{2} sequestration schemes.

Coal reservoirs are complex, fractured systems, consisting of a low permeability matrix and a high permeability fracture network. Diffusion and convection play important roles in transport in coals but on different time scales. We show first that diffusion acts rapidly in coal beds with small fracture spacing and that in such settings, an equilibrium assumption is reasonable. The equilibrium assumption permits use of the method of characteristics for obtaining analytical solutions for compositional flow of a coal bed.

New analytical solutions are reported for two-phase, three- and four-component flow with volume change as components adsorb and desorb. Analytical solutions allow isolation of physical mechanisms involved in the displacement and provide useful understanding of the key mechanisms that affect recovery and sequestration.

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Model formulation

To estimate time scales for equilibration, diffusion times were calculated for a sphere of coal. Sphere radius, \( R \), was taken to be an average spacing between fractures. Figure 1 reports the time required for the average \( \text{CO}_2 \) concentration to reach 95\% of the equilibrium value. Diffusion time is a function of diffusion coefficient and length. For coal beds with small diffusion coefficients, \( 5 \times 10^{-9} \) to \( 5 \times 10^{-6} \) \( \text{cm}^2/\text{s} \) [1], equilibration times can range from days to months, depending on fracture spacing. For large diffusion coefficients (\( \sim 10^{-5} \) \( \text{cm}^2/\text{s} \)), equilibration times for all radii of spheres considered are on the order of days.

In systems with small diffusion times, it is reasonable to assume that convection dominates transport. For convection-dominated flows, one-dimensional analytical solutions can be obtained.

![Figure 1: Diffusion times calculated as a function of radius and diffusion coefficient (right) for a simple spherical system (left).](image)

The method of characteristics was used to develop analytical solutions for transport in an ECBM process. This approach has been applied by many [2-5] to solve problems related to adsorption chromatography, enhanced oil recovery, and flow in a coal bed without water present. In this paper, we include the effects of a flowing water phase, and we report the first analytical solutions for four-component flow in a coal bed. In the analysis that follows, flow is assumed to occur in a one-dimensional homogeneous coal bed, and effects of diffusion and dispersion in the flowing fluid are neglected. Adsorption and desorption are assumed to occur quickly enough that phases present at a given location are in equilibrium. An extended Langmuir isotherm [6] is used to represent adsorption, and constant equilibrium K-values are used to represent component partitioning between liquid and vapor phases. Phase molar densities are assumed to be independent of composition. Effects of gravity and capillary pressure are ignored, and water was assumed not to adsorb.

For a multi-component system, there is a conservation equation for each of the \( N_c \) components:

\[
\frac{\partial G_i}{\partial t} + \frac{\partial H_i}{\partial \xi} = 0, \quad i = 1, \ldots, N_c, \tag{1}
\]

where \( G_i \) and \( H_i \) are overall compositions and component fluxes defined as

\[
G_i = \rho_{GD} y_i S_e + \rho_{LD} x_i (1 - S_g) + \frac{1 - \phi}{\phi} a_i D, \tag{2}
\]

\[
H_i = u_D \left( \rho_{GD} y_i f_g + \rho_{LD} x_i (1 - f_g) \right). \tag{3}
\]

Component sorption terms entering Eq. (2) are given by
\[ a_{ID} = \frac{\rho_{i,std} \rho_{coal}}{\rho_{mj}} \times \frac{V_{m,i} b_i P_i y_i}{1 + \sum_{j=1}^{N_{ads}} b_j P_j y_j}. \] (4)

Eqs. 2 to 4 are given in dimensionless form with \( \rho_{GD}, \rho_{LD} \) and \( a_{ID} \) denoting the dimensionless molar density of gas, liquid and adsorbed phases respectively. These terms are scaled by the molar density of the injected fluid (\( \rho_{inj} \)). Time and space are made dimensionless using the following relations:

\[ \tau = \frac{v_{inj} t}{\phi L}, \quad \xi = \frac{x}{L}. \] (5)

\( u_D \) is the dimensionless total velocity, scaled with respect to the injection velocity and \( f_g \) is the fractional flow of gas given, as a function of the gas saturation (\( S_g \)), by the Corey type relation

\[ f_g = \frac{\hat{S}^2}{\hat{S}^2 + M(1 - \hat{S})^2}, \quad M = \frac{\mu_g}{\mu_w}, \quad \hat{S} = \frac{S_g - S_{gc} - S_{wc}}{1 - S_{gc} - S_{wc}}. \] (6)

In the examples that follow, the critical gas saturation (\( S_{gc} \)) and the connate water saturation (\( S_{wc} \)) were set to zero. The viscosity of gas and liquid phases entering Eq. (6) are given by \( \mu_g \) and \( \mu_w \) respectively. The amount of component \( i \) adsorbed depends on the partial pressure (\( P_i y_i \)) of the equilibrium gas composition (\( y_i \)) and the Langmuir parameters \( V_{m,i} \) and \( b_i \). The adsorbed amount (scf/ton of coal) is converted to moles/m\(^3\) using pure component molar density at standard conditions (\( \rho_{i,std} \)) and the coal mass density (\( \rho_{coal} \)).

Equilibrium phase compositions are given by

\[ y_i = K_j x_i, \] (7)

where \( y_i \) and \( x_i \) are mole fractions of component \( i \), and \( K_j \) is an equilibrium K-value. This set of equations can be written as an eigenvalue problem in which the eigenvalues represent velocities at which compositions propagate. The corresponding eigenvectors represent the directions of variation in composition space. Solutions connecting initial and injection conditions can consist of segments of continuous variation, discontinuous variation and zones of constant state. Unique physical solutions are constructed by applying a velocity rule that requires fast-moving compositions to lie downstream of slow ones and an entropy condition that requires shocks to be stable to perturbations.

For clear display of the solution structure, model flow parameters were used (Tables 1 and 2).

<table>
<thead>
<tr>
<th>( \rho_{inj} ) (mol/m(^3))</th>
<th>1</th>
<th>component</th>
<th>( K_i ) (m(^3)/ton)</th>
<th>( V_{mi} ) (m(^3)/ton)</th>
<th>( b_i ) (MPa(^{-1}))</th>
<th>( \rho_{i,std} ) (mol/m(^3))</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \rho_{GD} )</td>
<td>1</td>
<td>( N_2 )</td>
<td>5</td>
<td>0.0069</td>
<td>0.2466</td>
<td>44.051</td>
</tr>
<tr>
<td>( \rho_{LD} )</td>
<td>2</td>
<td>( CH_4 )</td>
<td>3</td>
<td>0.0138</td>
<td>0.4931</td>
<td>44.135</td>
</tr>
<tr>
<td>( \rho_{coal} )</td>
<td>1536</td>
<td>( CO_2 )</td>
<td>1.2</td>
<td>0.0221</td>
<td>0.9573</td>
<td>44.326</td>
</tr>
<tr>
<td>( M )</td>
<td>10</td>
<td>( H_2O )</td>
<td>0.1</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

In actual \( N_2\)-\( CH_4\)-\( CO_2\)-\( H_2O \) systems, K-values of \( N_2 \), \( CO_2 \) and \( CH_4 \) are much higher than the values used here, while that of \( H_2O \) is much lower. The more realistic K-values result in even lower solubilities of \( N_2 \), \( CO_2 \) and \( CH_4 \) in the aqueous phase, increasing the size of the two phase region. In addition, the phase densities used exaggerate the effects of sorption, so that they can be seen more clearly in the solutions presented. When the more realistic parameters are used, the features of the displacement (shocks, rarefactions and zones of constant state) remain the same.
Ternary displacements

Solutions in three-component flows display two types. One type (Figure 2) occurs when the injection gas adsorbs more strongly than the initial gas composition. Preferential adsorption of injection gas removes gas molecules from the mobile phases as they propagate, resulting in self-sharpening behavior. Injection of a CO$_2$-rich mixture into a coal bed with adsorbed CH$_4$ displays such behavior. Because adsorbed CO$_2$ occupies less volume than CO$_2$ in the free gas phase, flow velocity decreases, resulting in a delay of breakthrough time of CO$_2$ at the outlet.

The second type of solution (Figure 3) occurs when a component that adsorbs more weakly displaces a more strongly adsorbing component. N$_2$ injection to displace adsorbed CH$_4$ is an example. As CH$_4$ desorbs, more volume is added to the flowing vapor phase, which causes an increase in flow velocity and earlier production of CH$_4$. Recovery curves for pure CO$_2$ and pure N$_2$ injection are compared in Figure 4. The additional volume change associated with N$_2$ injection results in earlier recovery of CH$_4$. For pure N$_2$ injection, the produced gas stream is a mixture of resident gas and injection gas, whereas in pure CO$_2$ injection, more strongly adsorbed component (CO$_2$) is separated completely from the less strongly adsorbed component (CH$_4$). The volume reduction associated with pure CO$_2$ injection reduces flow velocity, which delays injection gas breakthrough.

In displacements in which the coal is only partially saturated initially with adsorbed components, an additional volume change occurs across the shock from the two-phase region to the initial composition, delaying the breakthrough time of injection gas. At early times, CH$_4$ recovery in undersaturated systems is slower than in saturated ones as additional injection gas is required to saturate the system.
Figure 3: Composition profile (left) and corresponding path in composition space (right) for displacement by pure $N_2$ of an initial composition of 0.31 $CH_4$ and 0.69 $H_2O$.

Figure 4: Recovery curves for pure $N_2$ injection, pure $CO_2$ injection and flue gas injection into a system with an initial composition of 0.31 $CH_4$ and 0.69 $H_2O$.

**Quaternary Displacements**

Separation of flue gases from a combustion process to obtain a pure $CO_2$ stream can be expensive, and hence, it may be desirable to inject a mixture of $CO_2$ and $N_2$ that results from a partial separation. Solutions for injection of $N_2$-$CO_2$ mixtures display a combination of the shock and rarefaction features that appear in the displacements by pure $CO_2$ and $N_2$ (Figure 5). Preferential adsorption of $CO_2$ causes it to propagate more slowly than $CH_4$ and $N_2$. The narrow bank of $CO_2$ in the solution profile demonstrates efficient displacement of $CH_4$ from the coal bed. $N_2$ propagates most rapidly and is present at the leading edge of the displacement. When a mixed gas is injected, $N_2$ and $CH_4$ are produced simultaneously, and subsequent separation of $CH_4$ from produced $N_2$ would be required. The effect of $N_2$-$CO_2$ injection on $CH_4$ recovery as a function of initial reservoir composition is shown in Figure 4. As the $CO_2$ concentration in the flue gas increases, recovery becomes slower. A larger proportion of the injected gas is removed from the flowing gas because $CO_2$ is preferentially adsorbed onto the coal, which slows the flow. Earlier recovery is achieved with mixed gas injection.
Conclusions

For convection-dominated systems, 1D analytical models provide insight into the interplay of phase behavior, adsorption, and convection in ECBM processes. These simple ternary and quaternary solutions establish the basis for understanding more complex multicomponent, two-phase flow with sorption. Strong adsorption of CO$_2$ onto the coal reduces the propagation velocity of CO$_2$, delaying breakthrough time of CO$_2$ at the outlet. If pure CO$_2$ is injected, pure CH$_4$ is produced until breakthrough of the injected CO$_2$. In displacements of CH$_4$ by N$_2$, an increase in flowing volume occurs as CH$_4$ desorbs, which results in earlier recovery of CH$_4$, though the produced gas is a mixture of N$_2$ and CH$_4$. When mixtures of N$_2$ and CO$_2$ are injected, elements of self-sharpening and spreading composition waves are present. Preferential adsorption of CO$_2$ causes it to propagate most slowly, and it is separated completely from the other gas components. N$_2$ propagates most rapidly and is co-produced with CH$_4$.

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References


