



Thermodynamic models for swelling of unconfined coal due to adsorption of mixed gases



Jinfeng Liu ^{a,*}, Colin J. Peach ^a, Hongwei Zhou ^b, Christopher J. Spiers ^a

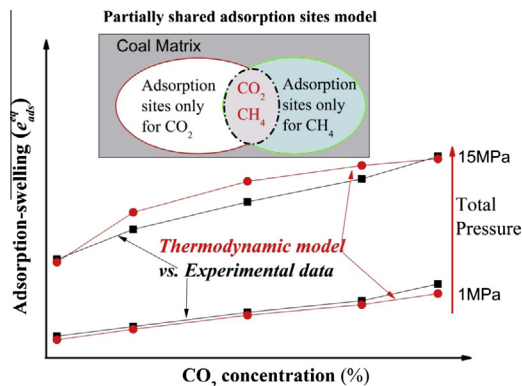
^a Department of Earth Sciences, Faculty of Geosciences, Utrecht University, 3584CD Utrecht, The Netherlands

^b State Key Laboratory of Coal Resources and Safe Mining, China University of Mining and Technology, Beijing 100083, China

HIGHLIGHTS

- Four swelling models derived for adsorption of binary gas mixture by coal.
- Sub-bituminous coal swelling best explained by partial pressure plus selective sorption model.
- Generalized model applicable to any coal and any binary gas mixture.
- Better basis for modeling coal seam permeability during ECBM.

GRAPHICAL ABSTRACT



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ABSTRACT

Permeability evolution in coal seams during CO₂-Enhanced Coalbed Methane (ECBM) production is strongly influenced by swelling/shrinkage effects related to sorption and desorption of both CO₂ and CH₄. Other gases, such as N₂, have also been proposed for injection in ECBM operations. In addition, water vapour will almost always be naturally present. Much work has been done on the sorption/swelling response of coal exposed to pure gases. However, to address in-situ coal-seam conditions realistically, an understanding of the swelling behaviour of coal matrix material due to multiple-species adsorption is needed. Here, we construct new thermodynamic models for swelling of unconfined coal due to adsorption of multiple gas/fluid species, addressing the equilibrium state and focusing on a binary gas mixture. Four models are derived, covering three possible end-member interactions, plus a generalized case. The end-member models considered correspond to adsorption of gases α and β at fully separate sites, at fully shared sites and at partially shared sites (Models 1–3, respectively). We compare our model predictions with literature data on the swelling behaviour of Bowen Basin coal exposed to CH₄/CO₂ mixtures at total pressures up to 15 MPa and at 55 °C. The results show that swelling of the Bowen Basin coal exposed to CO₂/CH₄ mixtures is best explained by Model 3, which in turn implies that adsorption and swelling is in this case determined by both partial pressure and selective adsorption (affinity) effects. Model 3 is easy to parameterize and its applicability to specific coals and gas mixtures is easily evaluated. Though more difficult to parameterize, our generalized swelling model (Model 4) can be applied to any coal rank, and to any binary gas mixture. It therefore offers an important tool for modeling swelling and permeability evolution during ECBM operations.

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* Corresponding author.

1. Introduction

The laboratory finding that coal matrix material has higher sorption capacity for pure CO₂ than for pure CH₄ at given pressure and temperature (*PT*) conditions has led to the now well-known concept of enhanced coalbed methane (ECBM) production coupled with storage of CO₂ in underground coal seams [1]. In this concept, CO₂ injection into a coal seam displaces the coalbed methane, which is then recovered and used for energy production, while the CO₂ generated is, in effect, re-injected [2]. Several field tests (e.g. in Canada, U.S.A., Poland, Japan and China) have confirmed the potential of CO₂-ECBM production [3–5]. However, most sites investigated to date have shown reduced CO₂ injectivity with time due to permeability reduction in the coal seam [3,4,6]. These effects are generally recognized to be caused by sorption-induced swelling of the coal as a result of CO₂ injection and sorption, while CH₄ is desorbed [7–13]. This exchange, or preferential adsorption effect, is considered to be due to either the reduction of CH₄ partial pressure as CO₂ penetrates the system or to truly selective adsorption of CO₂ over CH₄ [2]. The resulting partial replacement of CH₄ by CO₂ leads to net swelling, reflecting equilibration with respect to the new composition of the gas/fluid mixture present in the coal seam [2,14].

However, most experimental measurements and theoretical models for adsorption and swelling of coal focus on a single, pure gas, and not on mixed gases. Much less is known about competitive adsorption between CH₄ and CO₂, and there is insufficient evidence to resolve whether the controlling factor is a partial pressure or a selective sorption effect. To avoid confusion about the meaning of these terms that has crept into the literature in recent years, in this paper we use the term “preferential adsorption” of species α to mean that the number of adsorption sites occupied by α , in a given coal mass at equilibrium with a gas/fluid mixture containing species α plus β , is greater than the number of sites occupied by β , regardless of the reason. We use the term “selective adsorption” of species α to mean that the number of adsorption sites occupied by α is greater than occupied by β , due to different site accessibilities (i.e. different affinities), even when α and β are present in the gas/fluid phase at equal activities (see Table 1 for definition of terms).

We return now to the lack of evidence on whether preferential adsorption of CH₄ and CO₂ in coal is controlled by partial pressure or selective sorption effects. To date, strongly conflicting experimental results have been reported. For example, in experiments on high volatile bituminous coal performed by using mixtures containing 25–75% CO₂ at 25 °C and 0.1–4 MPa total pressure, Majewska et al. [15] observed that CH₄ preferentially adsorbed

with respect to CO₂. Czerw [16] repeated the experiments of Majewska et al., using the same apparatus, similar coals and a 75:25 CO₂-CH₄ mixture at 25 °C and 4 MPa total pressure, but reported that CO₂ preferentially adsorbed with respect to CH₄. Similarly, Busch et al. [13,17] reported that whereas preferential adsorption of CO₂ and preferential desorption of CH₄ were observed in high rank coals at all total pressures investigated (1–23 MPa), low rank samples showed preferential adsorption of CH₄ at low pressures and preferential desorption of CO₂ at all pressures. These authors [17] further reported that preferential sorption is independent of CO₂-CH₄ mixture composition, (i.e. of partial pressure), and proposed that preferential sorption is determined purely by selective sorption. On the other hand, in experiments on the effect of moisture on the (otherwise) pure CO₂ and pure CH₄ sorption capacity of bituminous coals at 55 °C and total pressures up to 20 MPa, Day et al. [8] found that the heat of adsorption (i.e. hence K) for pure CO₂ and CH₄ are similar when drying, falling only slightly when water is present. Their calculations were done based on Dubinin-Radushkevich model applied for pure CO₂ and pure CH₄, ignoring potential effects of water in this model. They therefore proposed that CO₂ and CH₄ have about the same access to all adsorption sites, i.e. that selective adsorption effects are negligible. The important question thus remains as to whether sorption by coal of a given rank exposed to mixed gases is solely determined by partial pressure or also by selective adsorption. Understanding the relative importance of these effects, via a physically correct sorption model, clearly plays a crucial role in assessing whether CO₂-ECBM operations are feasible in practice.

Focusing on the swelling response to mixed gas sorption, Day et al. [14] recently reported measurements performed on four Australian coals, exposed to CO₂, methane and mixtures of the two. Measurements were made on laterally unconstrained (i.e. mechanically unconfined), monolithic samples (30 × 9 × 9 mm) at 55 °C, applying total gas pressures up to ~15 MPa. Volumetric swelling strain ranged from about 1.0% to 5.5%, depending on coal rank and the proportion of CO₂ in the CH₄-CO₂ mixture. These authors found that the swelling observed using mixed gases lay between the values obtained for the pure end members. They also performed experiments in which CO₂ was injected in an attempt to displace CH₄ from the same coals. In these runs, coal samples were first equilibrated with CH₄ at a pressure of 15 MPa at 55 °C. CO₂ was then injected, keeping the total pressure constant. It was found that CH₄ was completely displaced from the coal, causing it to swell to the same level as if exposed to pure CO₂ at the same *PT* conditions. Upon subsequent injection of helium, which does

Table 1
Definition of terms used in this paper for the case of adsorption of gas/fluid species α and β , i.e. for adsorption from a binary gas mixture. Note that $n_s^{i\beta}$ is the number out of all adsorption sites fully accessible to α and β , C_s^i is the concentration of available adsorption sites accessible to component i in mole per kg coal, K^i is the equilibrium constant for the adsorption process of species i , a_g^i is the activity of the free gas/fluid species i .

Term	Definition	Controlling factors	Model developed in this study
Preferential adsorption	The number of adsorption sites occupied by one component is greater than that occupied by the other component, regardless of the reason	C_s^i, K^i, ϕ, a_g^i	
Selective adsorption	The number of adsorption sites occupied by one component is greater than that occupied by the other component even when each gas component has equal activity, due to different site accessibilities	C_s^i, K^i, ϕ	
Site-sharing factor ϕ	The fraction of the total adsorption sites n_s^{total} present in the coal matrix material that is accessible to gas α and to gas β . $\phi = n_s^{\alpha\beta} / n_s^{total}$, $0 \leq \phi \leq n_s^\alpha / n_s^\beta \leq 1$	K^i	
Independent adsorption sites	The adsorption sites accessible to each gas component (α, β) are fully separate and independent, such that $\phi = 0$	K^i	Model 1
Fully shared adsorption sites	All adsorption sites are fully accessible to (i.e. can take up) both gas components (α, β), so that $\phi = 1$	K^i	Model 2
Partially shared adsorption sites: Type I	All adsorption sites accessible to gas α are also accessible to gas β , while all remaining adsorption sites are accessible only to gas β . ($\phi = n_s^\alpha / n_s^\beta < 1$)	K^i	Model 3
Partially shared adsorption sites: Type II	$n_s^{\alpha\beta} = \phi n_s^{total}$ sites are accessible to both gases (cf. Model 2). The remaining adsorption sites can only take up one component (cf. Model 1), such that $(n_s^\alpha - \phi n_s^{total})$ adsorption sites are accessible only to gas α , while $(n_s^\beta - \phi n_s^{total})$ adsorption sites are accessible only to gas β	K^i	Model 4

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