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Kinetics of coal swelling in gases: Influence of gas pressure, gas type and coal type

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ABSTRACT

The increasing interest in enhanced coal bed methane (ECBM) production and reporting of greenhouse gas emissions from mines has spotlighted gas diffusion and gas-induced swelling in coals. Although it is generally agreed that carbon dioxide (CO_2) diffuses faster than methane (CH_4) in coal and that diffusion rates decline as the pressure is increased, there is no general agreement on the physical mechanism responsible. In this paper kinetic data for sorption-induced swelling for five Australian bituminous coals have been analysed. Gases investigated included CO_2 and CH_4, Xe and ethane. It was found that swelling rates are influenced by gas type. CH_4 swelled coal more slowly than CO_2 at the same pressures. Other swelling kinetic data showed that, of the gases investigated, xenon was the slowest, followed by ethane. CO_2 swelling was found to be the fastest overall. The swelling rate was found to depend on maceral composition, with the slower-swelling coals being vitrinite-rich. However, the swelling rate of different coals varied differently with increasing pressure: the slowest swelling coal showed an increase in swelling rates with an increase in system pressure, whereas the fastest swelling coals showed a decrease in the swelling rates at high pressures.

1. Introduction

The diffusion of gas in coal and the swelling of coal by gases have been subject to intense investigation recently due to an increasing interest in enhanced coal bed methane (ECBM) and of reporting greenhouse gas emissions from mines. Though it is generally agreed that carbon dioxide (CO_2) diffuses faster than methane (CH_4) through coal (Busch and Gensterblum, 2011) and that diffusion rates decrease as the pressure is increased, there is no general agreement on the physical mechanism for, nor physical model of, diffusion in coals. Cui et al. (2004) have suggested that swelling-induced constrictions could explain the decline of the measured micropore diffusion coefficient with pressure. Pone et al. (2009) have also discussed the possibility that non-uniform swelling and the subsequent visco-elastic response of the coal structure could influence transport of the gas in the coal.

There have been only a handful of studies investigating the kinetics of gas sorption-induced coal swelling, notably those of Reucroft and Sethuraman (1987), Kelemen and Kwiatek (2009) and Mazumder et al. (2006). More attention has been devoted to the kinetics of coal swelling with organic solvents such as pyridine (Hall et al., 1992; Milligan et al., 1997; Murata et al., 2008; Ndaji and Thomas, 1993; Otake and Suuberg, 1997; Peppas and Lucht, 1985).

Reucroft and Sethuraman (1987) compared CO_2 swelling kinetics on three cylinders of coals (1 cm in length and 0.4 cm in diameter) at three different integral (vacuum to pressure) steps 5, 10 and 15 atm at 25 °C. They found that swelling was faster at higher pressures. They concluded that swelling extent increases with the rank of the coals (although only three samples were compared). The swelling rates appeared to be different between the coals, but this was not analysed.

More recently, Kelemen and Kwiatek (2009) made strain and kinetic measurements on coals using different gases and at different temperatures. They found that the rate of sorption (as determined by an approximation of the unipore diffusion model) and rate of swelling for integral steps 0 − 1.8 MPa were similar. Both followed the same order for gas type, with the rates decreasing in the order CO_2, N_2 and CH_4.

The kinetics of sorption-induced swelling of coal thus remains an unexplored area of research. Day et al. (2008b, 2010) have published data for the swelling extent of different coals and the pressure for various gases, but not on the kinetics of swelling. There is an additional motivation to utilise these data not only to search for possible links with diffusion behaviour but to identify key coal properties that may influence its swelling rate.

In this paper, sorption-induced swelling kinetic data are examined. At equilibrium, the extent of swelling in a coal is proportional to the van der Waals volume of gas sorbed, independently of the nature of the gas (Day et al., 2010), but it does not follow that swelling only happens immediately when the gas is sorbed; the coal may show viscoelastic behaviour. However, Mazumder et al. (2006) have found in their

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study that the CO_2 sorption rate and CO_2-induced swelling rate occurred on similar time scales. If mechanical deformation of the coal due to the absorption of gas occurs on the same time scale as gas diffusion into the coal, it would indicate that absorption is a limiting factor for gas uptake rates in coal. The swelling rate of coal is here examined as a function of coal type, gas type and pressure.

2. Materials and methods

A brief description is given here of the procedure used to determine the amount of swelling by the coal, following Day et al. (2008b, 2010) and of the calculations used in this paper for the analysis of time-based swelling data. The data collected were for five coal blocks: Coal-C, Coal-D, Coal-E, Coal-F and Coal-G (refer to Table 1 for key coal properties).

All five samples were machine-cut bulk specimens (nominal dimensions: 30 mm × 10 mm × 10 mm) from run-of-mine samples, from the Bowen basin (Coal-C and Coal-D) and Sydney basin (Coal-E, Coal-F and Coal-G), Australia. Since crushed and screened samples were used for isotherm measurements and coal characterisation (Day et al., 2008a) there may have been some sample bias, as the blocks were only selected from parts of the batch that did not break when cut. Also, to avoid potential fracturing during high pressure gas loading, for CO_2 Coal-C and Coal-E were first dosed very slowly, over a half hour period The subsequent pressure increments were similar to those applied in isotherm measurements. No fractures were reported from these large pressure increments (Day et al., 2008b).

The swelling was recorded using an optical dilatometer. Digital cameras were positioned to observe dimensional changes in the blocks coal inside a pressure cell with glass windows. The visually reported swelling was compared to that of a steel block of the same dimensions to correct for image changes due to the refractive index of the gas. The reader is referred to Day et al. (2008b) for a detailed summary of the experimental setup. A brief description is provided below.

The swelling measurements were performed at 55 °C, with the swelling enclosure submerged in a bath of stability ±0.5 °C. Samples were held under vacuum for at least 24 h before being dosed with gas. Percentage length changes were measured relative to the size of the block at vacuum (initial volume $V0$) as the pressure was increased in steps, to a maximum of 15 MPa. Two lengths were measured for two blocks of the same coal: one positioned perpendicular to its bedding plane (l_{per}), one parallel (l_{par}). It was assumed that expansions of the parallel lengths of each block were equal and the perpendicular lengths of each block were equal. The volume of the coal as a function of time ($V(t)$) can be defined as:

$$V(t) = V0 + \Delta V(t) = \left(l_{per} + \Delta l_{per}(\mathrm{t})\right) \times \left(l_{par} + \Delta l_{par}(\mathrm{t})\right)^2 \tag{1}$$

The swelling, as a function of time ($Q(t)$), is:

$$Q(t) = \frac{V(t) - V0}{V0} \tag{2}$$

At equilibrium the swelling is simply $Q = (V - V0)/V0$. At each pressure step if Q_i is the swelling at the end of the time the sample is exposed to pressure P_i (i.e. the i^{th} Pressure step) and Q_{i-1} is the swelling at the start of exposure to P_i then the fractional swelling change during the exposure to gas, $q_i(t)$, is:

$$q_i(t) = \frac{(V(t) - V0)/V0 - Q_{i-1}}{Q_i - Q_{i-1}} \tag{3}$$

This form (where $q_i(t)$ is 0 at the start of each gas addition and 1 when the swelling has reached equilibrium at the given gas pressure) is analogous to the calculation of the excess sorption at any time t when expressed as a fraction of the value at equilibrium (Li et al., 2010; Pone et al., 2009; Staib et al., 2013). Whilst fractional extents of swelling over time have been studied before (Hall et al., 1992; Kelemen and Kwiatek, 2009; Otake and Suuberg, 1997) it has only been in single *integral* steps (from vacuum to the desired pressure) not for *incremental* swelling points where the coal has already had prior swelling. As far as can be determined Eq. (3) has not been used before to describe swelling kinetics in coal.

The swelling kinetic data was of sufficient resolution to examine the following:

1. Comparison of CO_2 swelling kinetics at 1 MPa and 2 MPa, for all five coals.
2. Possible pressure dependence of swelling kinetics for Coal-D at 1, 2, 4, and 8 MPa.
3. Comparison between CH_4 and CO_2 swelling kinetics at 2 MPa and 4 MPa for Coal-D.
4. Comparison of CO_2, ethane and xenon swelling kinetics at 1 MPa and 4 MPa for Coal-C and Coal-E.

3. Results

3.1. Swelling kinetics for CO_2

CO_2 swelling isotherms presented in Fig. 1 show that Coal-C had the largest volumetric expansion, with Coal-E only slightly less, followed by Coal-D, with Coal-F and Coal-G showing the least, over the range of pressures applied, at 55 °C.

Using Eq. (3) the swelling kinetics could be described as a fractional swelling extent for a given sorbate loading in a pressure step. The fractional swelling extent for all coals at 1 MPa pressure is presented in Fig. 2. There are differences in the kinetics of swelling for the five coals. Coal-C, Coal-E and Coal-G have the highest swelling rates with the swelling rate of Coal-F slightly less and Coal-D displaying a distinctly lower swelling rate. Similarly, at a higher pressure (2 MPa), Fig. 3 shows that the differences in swelling rates between the coals remain the same.

The ocular technique for detecting volumetric changes in the coal blocks was optimised for equilibrium measurements where an average could be obtained over several instrument recordings. The error in each length measurement was estimated to be 0.01 mm; hence, the absolute error in an individual swelling ratio value was about 0.1%. If the

Table 1
Coal properties. ar = as received, db = dry basis, daf = dry ash free basis. mmf = mineral matter free, VM = volatile matter. W_0 = sorption capacity, as presented in Day et al. (2008a, 2008c).

Coal	Moisture	Ash	VM	C	H	N	Vitrinite	Liptinite	Inertinite	W_0 (CO_2)
ID	% ar	%db	daf %	daf %	daf %	daf %	vol.% mmf	vol.% mmf	vol.% mmf	kg/t
Coal-C	9.3	20.3	31.2	80.7	3.9	1.1	23.9	1.6	74.5	135
Coal-D	1.5	5.6	36.1	84.1	5.7	2.3	82.7	4.1	13.2	68
Coal-E	8.5	7.7	31.7	83.0	4.7	1.8	29.7	3.9	66.4	119
Coal-F	2.4	7.4	37.3	83.6	5.4	2.1	88.7	2.2	9.1	73
Coal-G	1.1	16.9	21.7	88.9	4.6	1.6	28.1	0.0	64.6	65