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Coal characterization for ECBM recovery: gas sorption under dry and humid conditions, and its effect on displacement dynamics

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Abstract

Enhanced Coal Bed Methane (ECBM) recovery is a technique under investigation as a possible approach to the geological storage of CO_2 in a carbon dioxide capture and storage (CCS) system. This technology allows enhancing the recovery of coal bed methane by injecting CO_2 in the coal seam at supercritical conditions. Through an in situ sorption/desorption process the displaced methane is produced and the adsorbed CO_2 is permanently stored. In the case of coal, the uptake of CO_2, CH_4 and N_2 is a combination of adsorption on its surface and penetration (absorption) into its solid matrix, the latter resulting in coal's swelling. These two processes act simultaneously, making the coal a challenging material to be studied, in particular with respect to the understanding of the fundamental aspects of gas adsorption. High pressure sorption data of CO_2, CH_4 and N_2 on a coal sample from Australia are presented; the interpretation of the experimental data takes into account the dual nature of the sorption process and a Langmuir-like model is applied to the sorption data, by fitting the isotherm parameters to the experimental values. The results confirm that this equation is a valuable option to describe gas sorption on coal. Moreover, a one-dimensional mathematical model previously derived is used to perform numerical simulations on the performance of ECBM recovery in coal beds. Important insights are obtained regarding the gas flow dynamics during displacement and the effects of gas sorption on the ECBM operation.

Keywords: adsorption; ECBM recovery, coal.

1. Introduction

Coal seams have received increasing attention as geological formations for the storage of the captured CO_2, because of the potential to recuperate some of the costs associated with the storage operation. This can be achieved with the so-called Enhanced Coal Bed Methane recovery (ECBM) process, where the recovery of the coal bed methane is enhanced by injecting carbon dioxide in the coal seam at supercritical conditions. Through an in situ adsorption/desorption process the displaced methane is produced and the adsorbed CO_2 is permanently stored. The

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ECBM technology has been implemented in a number of field tests worldwide with different geological characteristics. By testing different gas injection policies, such as CO_2, N_2 or their mixtures, these field tests have shown the potential of coal seams as storage sites for CO_2. However, they also evidenced that many factors affect the success of the ECBM operation, which need to be extensively investigated.

The problems encountered during the field test are largely due to the interactions of the gases with different structural and chemical features of the coal. In fact, coal seams differ from conventional gas reservoirs as their storage capacity is given mainly by the amount of gas that can be adsorbed onto the coal's porous surface and absorbed into the coal's matrix (Figure 1). Moreover, as a consequence of gas sorption the coal swells and the volumetric strain caused by this phenomenon induces a change in the stress field of the coal seam. Fractures (cleats) undertake most of the deformation upon stress changes, being very sensitive to them as compared to the coal matrix. A variation in the cleats opening is definitively affecting the permeability of the coal seam, which is the main petrophysical property controlling the performance of the ECBM operation.

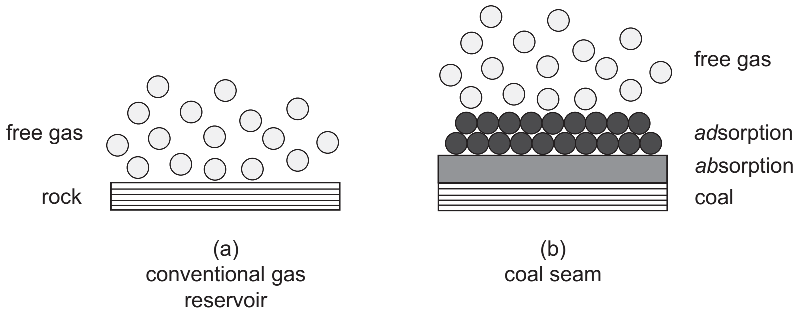

Figure 1 – Simplified schematic for the storage mechanism in a conventional gas reservoir (a) and in a coal seam (b). For the former, the injected gas fills the available pore volume as a compressed fluid, whereas for the latter the gas is additionally adsorbed and absorbed.

A study has been undertaken with the aim of developing experimental and modelling tools that are able to provide a comprehensive characterization of coal required first to understand the mechanisms acting during the process of injection and storage and secondly to assess potential of a given coal for an ECBM operation.

2. Methodology

2.1. Gas sorption

The gas uptake process in coal is most likely a combination of adsorption on its surface and penetration (absorption) into its solid matrix. Moreover, it has been observed that upon exposure to a gas, the coal's volume changes (swelling) and that the causes for this behaviour rely on the dual nature of the sorption process. High pressure adsorption isotherms with pure gases were obtained using a Magnetic Suspension Balance (Rubotherm, Germany) and the details of the adopted measurement procedure are described elsewhere [1]. When working with coal, this technique allows obtaining the so-called excess sorption (m^{eas}), which is the only truly measurable quantity in the high pressure sorption experiment, and it is defined as [2]:

$$m^{eas} = m^{a} + m^{s} - \rho_{m}(V^{a} + \Delta V^{s}) \tag{1}$$

where m^a and m^s are the amount adsorbed and absorbed, respectively; ρ_m is the mass density of the bulk fluid; V^a is the volume of the adsorbed phase and ΔV^s is defined as the difference between the volume of the mixture of coal and dissolved gas, and the initial sample volume. As can be seen from Eq. (1), this technique alone does not allow separating between the effect of adsorption and absorption. This aspect has to be taken into account both when interpreting the experimental sorption data as well as when models are used to describe the experimentally obtained excess sorption isotherms. Unfortunately, this is not always acknowledged in the literature reporting sorption data on coal. We have developed a method that is based on a graphical interpretation of the excess sorption isotherm and that allows obtaining the absolute sorption isotherm (m^{a} + m^{s}) from the measured excess sorption data (m^{eas}) [2]. This method has been applied to several coal samples from different coal mines worldwide, showing that the obtained absolute isotherm is always of Type I, i.e. characterized by a gradual flattening (saturation limit) at large densities, which can therefore be described by a Langmuir-like equation, i.e.

$$m^{eas} = \frac{m^{\infty} b \rho_m}{1 + b \rho_m} - \rho_m (V^{a} + \Delta V^{s}) \tag{2}$$

with m^{∞} and b being the maximum sorption (*ad*- and *ab*sorption) capacity and the Langmuir constant, respectively.

2.2. Displacement dynamics

Gas sorption and coal's swelling have complex effects on the displacement dynamics during an ECBM recovery operation; an accurate description of these processes is essential for the development of reliable reservoir simulators used to history match field test data. Input for these models are the results of laboratory studies, such as the sorption data described above. In a recent paper, a one-dimensional model was derived and successfully applied for describing pure gas injection experiments into coal cores confined under an external hydrostatic pressure and under simulated reservoir temperature and pressure [3]. The model consists of mass balances describing gas flow and sorption, and a geomechanical relationship to account for porosity and permeability changes during injection. This model has now been extended to the multicomponent single-phase (gas) displacement in a coal seam allowing to perform numerical simulations that estimate the performance of CO_2 storage and ECBM recovery in coal beds. In particular, different ECBM scenarios involving the injection of gas mixtures with different composition (from pure N_2 to pure CO_2) into a coal bed previously saturated with methane are investigated.

3. Results and discussion

Single component sorption isotherms of CO_2, CH_4 and N_2 at 55°C on the same Australian coal sample have been measured in two different laboratories, namely at CSIRO (Newcastle, Australia) as reported earlier [4] and in our laboratory at ETH Zurich (Zurich, Switzerland) using the equipment and procedure described above. Each sample was divided in two parts, one of which was analyzed at CSIRO, and one was sent for analysis to ETH Zurich. Sorption data at CSIRO and ETH Zurich have thus been obtained on identical coal. Once received in our lab, care was taken to follow the same procedure as at CSIRO when preparing the sample for the experiments. The sample was kept in a plastic bottle (as received) and was vacuum dried overnight at 60°C prior to the sorption measurements. The techniques to obtain the sorption isotherms are different in the two labs, but they both rely on a gravimetric approach. Moreover, for the experiments reported below, different amounts of sample have been used: 200 g at CSIRO and 3 g at ETH Zurich. Results of these experiments are shown in Figure 2, where the molar excess sorption of CO_2, CH_4 and N_2 at 55°C is plotted as a function of the molar bulk density of the fluid phase. It can be seen that for both samples and for all gases the agreement between the two laboratories over the whole range of densities (which for all gases corresponds to pressures up to 200 bar) is rather satisfactory. Together with the experimental data points are shown the fitting results for the excess (dashed lines) and the corresponding absolute (solid lines) isotherms, which are based on a Langmuir-type equation. It can be seen that this model is able to reproduce well the experimental sorption data.

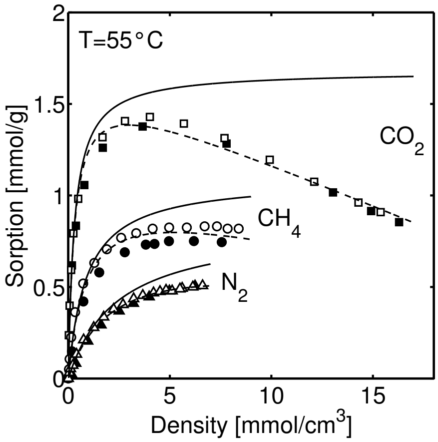

Figure 2 - CO_2, CH_4 and N_2 molar excess sorption on a coal samples from Australia at 55°C as a function of the bulk density measured at CSIRO (Newcastle, Australia) (open symbols) [4] and in our lab (closed symbols). A sorption model based on a Langmuir-like equation has been fitted to the experimental data (dashed lines) allowing to obtain the corresponding absolute sorption curves (solid lines).

Since coal seam may, at least partly, be saturated with water, it is important that sorption experiments aimed at storage capacity estimations are performed on wet coal samples as well. Unfortunately, techniques for the measurement of sorption isotherms on wet samples are not as well established as those on dry samples. All adsorption measurements show that wet coal uptakes CO_2 always less than dry coal, because of competitive water adsorption. However, the quantitative effect of moisture on CO_2 uptake is less certain because of the intrinsic difficulty of the measurements. It has been shown for instance that a coal containing about 3% moisture, corresponding to a relative humidity of 50%, adsorbs 30% less CO_2 than the corresponding dry one [5]. For the coal shown in Figure 2, this would correspond to a decrease of CO_2 maximum sorption capacity per unit mass dry coal from 7% to 5% weight.

The two main peculiarities of the ECBM recovery process are that gas sorption is responsible for gas storage in coal seams and that the adsorption/desorption mechanism taking place controls the displacement dynamics. The latter aspect can be understood at best by performing numerical simulations. Figure 3 shows snapshots of the concentration profiles of CO_2 (solid line), CH_4 (dashed line) and N_2 (dotted line) for three different injection scenarios: pure CO_2 (a), 50:50/CO_2:N_2 (b) and pure N_2 (c). Details about the simulations are given elsewhere [6]. It can be seen that injection of pure CO_2 displaces the CH_4 through a relatively sharp front, due to the higher adsorptivity of the former compared to the latter. Therefore as the preferentially adsorbing CO_2 propagates through the coal bed, no CH_4 is left behind. On the contrary, when pure N_2 is injected the displacement front is much smoother, with the N_2 moving faster than CH_4 and overtaking it. Again, this can be attributed to the adsorption behavior of the gases involved, as in this case the injected component (N_2) adsorbs less than the displaced component (CH_4). Injection of a CO_2/N_2 mixture results in the appearance of both the above mentioned effects, as shown in the central snapshot. It is worth pointing out, that at the CO_2/CH_4 front the N_2 is enriched in the fluid phase, the latter being the least adsorbing component. With respect to the purity of the CH_4 produced, it can be expected that when pure CO_2 is injected, the CH_4 recovery will completed as CO_2 breakthrough takes place, because of the characteristic displacement behavior described above. On the contrary, gas mixtures containing N_2 will show an early breakthrough of N_2. In the case of 50:50/CO_2:N_2 injection, this behavior results in a produced stream of CH_4 polluted with N_2, until CO_2 breakthrough occurs.

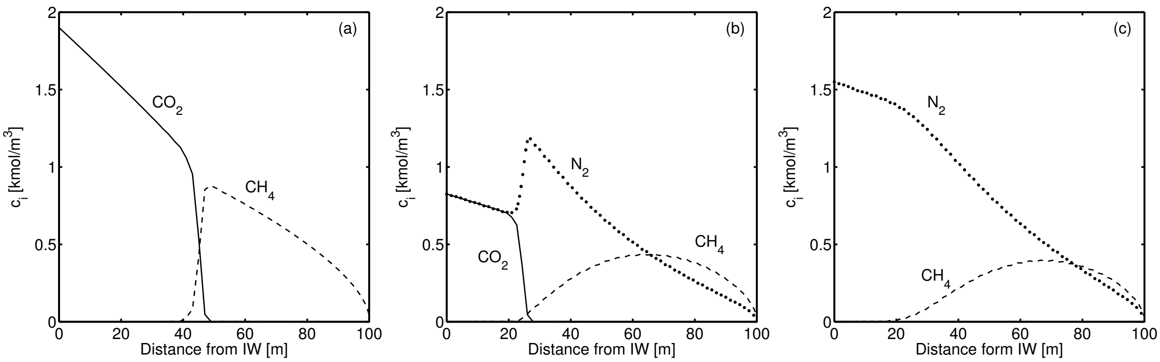

Figure 3 – Concentration profiles of CO_2, CH_4 and N_2 along the coal bed axis for the three different injection scenarios: pure CO_2 (a), 50:50/CO_2:N_2 (b) and pure N_2 (c). Details about the simulations are given elsewhere [6].

4. Concluding remarks

The quantitative estimation of the CO_2 storage capacity of the coal seam is needed to predict the reservoir behavior during an ECBM operation. This estimation has to rely on accurate measurement techniques and on a correct interpretation of the obtained data. In this study, we have presented high pressure sorption data of CO_2, CH_4 and N_2 on an Australian coal sample; the data presented in this study are consistent with those measured at CSIRO on another sample from the same batch. A Langmuir-like model has been applied to the sorption data, by fitting the isotherm parameters to the experimental values. The results confirm that this equation is a valuable option to describe gas sorption on coal.

Gas sorption has complex effects on the displacement dynamics, whose accurate description is essential for the development of reliable reservoir simulators used to history match field test data. The gas flow dynamics during ECBM recovery operations have been studied with the help of a one dimensional mathematical model, consisting of mass balances describing gas flow and sorption, and a geomechanical relationship for the description of porosity and permeability changes during injection. Simulation results show that gas injection can indeed enhance methane recovery. Important insights on the characteristic displacement dynamics could be highlighted and attributed to the specific adsorption behavior of the gases involved in the process.

References

[1] Ottiger S, Pini R, Storti G, Mazzotti M, Bencini R, Quattrocchi F, Sardu G, Deriu G. Adsorption of pure carbon dioxide and methane on dry coal from the Sulcis Coal Province (SW Sardinia, Italy). Env Progress 2006; 25:355-364.

[2] Pini R, Ottiger S, Burlini L, Storti G, Mazzotti M. Sorption of carbon dioxide, methane and nitrogen in dry coals at high pressure and moderate temperature. Int J Greenhouse Gas Control 2010; 4:90-101.

[3] Pini R, Ottiger S, Burlini L, Storti G, Mazzotti M. Role of adsorption and swelling on the dynamics of gas injection in coal. J Geophys Res - Solid Earth 2009; 114:B04203.

[4] Sakurovs R, Day S, Weir S, Duffy G. Application of a Modified Dubinin- Radushkevich Equation to Adsorption of Gases by Coals under Supercritical Conditions. Energy Fuels 2007; 21:992-997.

[5] Day S, Sakurovs R, Weir S. Supercritical gas sorption on moist coals. Int J Coal Geol 2008; 74:203-214.

[6] Pini R, Storti G, Mazzotti M. A model for Enhanced Coal Bed Methane recovery aimed at carbon dioxide storage. Adsorption 2010; submitted.