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Conference Paper

Author(s):
Pini, Ronny; Ottiger, Stefan; Burlini, Luigi; Storti, Giuseppe; Mazzotti, Marco

Publication date:
2009-02

Permanent link:
https://doi.org/10.3929/ethz-b-000019366

Originally published in:
Energy Procedia 1(1), https://doi.org/10.1016/j.egypro.2009.01.224

Available online at www.sciencedirect.com

ScienceDirect

Energy Procedia 1 (2009) 1711–1717

Energy Procedia

www.elsevier.com/locate/procedia

GHGT-9

CO_2 storage through ECBM recovery: an experimental and modeling study

Ronny Pini[a], Stefan Ottiger[a], Luigi Burlini[b], Giuseppe Storti[c], Marco Mazzotti[a]*

[a]*ETH Zurich, Institute of Process Engineering, Sonneggstrasse 3, CH-8092 Zurich, Switzerland*
[b]*ETH Zurich, Geological Institute, Leonhardstrasse 19, CH-8092 Zurich, Switzerland*
[c]*ETH Zurich, Institute for Chemical and Bioengineering, Wolfgang-Pauli-Str. 10, CH-8093 Zurich, Switzerland*

Abstract

The permeability of the coal seam is the main petrophysical property controlling the performance of the ECBM operation, since it affects both the CO_2 injection and CH_4 recovery. In the present paper, coal swelling of intact coal samples is studied both under unconstrained and constrained conditions. Unconstrained swelling experiments are performed in a view cell under a static high pressure gas atmosphere, whereas gas injection experiments are carried out in a flow cell, where the sample is subjected to a given hydrostatic confinement. Both experiments are performed by using different gases, namely He, CO_2, CH_4 and N_2, and under typical coal seam conditions, i.e. at high pressure and at 45°C. The results of the unconstrained coal sample showed that swelling increases monotonically with pressure up to a few percents for adsorbing gases, with CO_2 swelling coal more than CH_4 that swells more than N_2, whereas for helium, a non-adsorbing gas, volume changes are negligible. The results of the flow experiments were successfully described using a mathematical model consisting of mass balances accounting for gas flow and adsorption, and mechanical constitutive equations for the description of porosity and permeability changes during injection. Results showed increase in permeability with decreasing effective pressure on the sample. Moreover, when CO_2 is used a permeability reduction was observed compared to Helium, which can be explained by taking into account the effects of swelling on the flow dynamics.

Keywords: ECBM; swelling; permeability; adsorption; modeling;

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1. Introduction

Research in the laboratory is very active to improve the understanding of the different mechanisms acting during the ECBM process, and in particular those related to permeability [1]. The ECBM field tests that have been carried out worldwide were only partly able to show that the CO_2/CH_4 displacement mechanism is actually taking place, that CO_2 can be stored and that the production of CH_4 can be enhanced. As an example, the observed decrease in permeability caused by swelling forces a reduction of the injection pressure thus lowering the overall process

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* Corresponding author. Tel.: +41 44 6322456; fax: +41 44 6321141.
E-mail address: marco.mazzotti@ipe.mavt.ethz.ch

doi:10.1016/j.egypro.2009.01.224

performance. Being at several hundred meters of depth, the coal seam is subjected to a specific stress situation, consisting of both overburden and lateral stresses. Both fluid pressure and volumetric strain (swelling/shrinkage) induced by gas adsorption/desorption induce changes 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 [2]. A variation in the cleats opening is definitively affecting the permeability of the coal seam. This phenomenon needs to be quantified, since, besides affecting CO_2 injectivity and CH_4 recovery, it hinders an optimal exploitation of the coal seam.

In the present paper, coal swelling of intact coal samples is studied both under un- and costrained conditions. Unconstrained swelling experiments are performed in a view cell under static conditions, whereas gas injection experiments are carried out in a flow cell, where the sample is subjected to a given hydrostatic confinement. Both experiments are performed by using different gases, namely He, CO_2, CH_4 and N_2, and under typical coal seam conditions, i.e. at high pressure and at 45°C. A one-dimensional model is used to describe the flow experiments, which consists of mass balances accounting for gas flow, adsorption and swelling combined with mechanical constitutive equations for the description of porosity and permeability changes during injection.

2. Experimental Section

A coal core from the Monte Sinni coal mine in the Sulcis Coal Province (Sardinia, Italy) was obtained. The sample was drilled in December 2004 at a depth of about 500 m and preserved in a plastic box in air. Results of a thermo-gravimetric analysis (TGA) give a coal composition of 49.4% in fixed carbon content, 41.2% in volatile matter, 2.1% in ash and 7.3% in moisture. These values, together with a vitrinite reflectance coefficient (R_0=0.7), allow classifying the coal as high volatile C bituminous [3]. For the unconstrained swelling experiments a coal disc of about 2 cm diameter was drilled form the coal block, whereas for the flow measurements a coal core of 2.54 cm (1 inch) in diameter and 3.6 cm in length was used. Prior to the experiments, the samples were vacuum dried in an oven at 70°C for at least 2 days. The following pure gases obtained from PanGas (Dagmersellen, Switzerland) were used in this study, namely, CO_2 and CH_4 at purities of 99.995% and N_2 and He at purities of 99.999%.

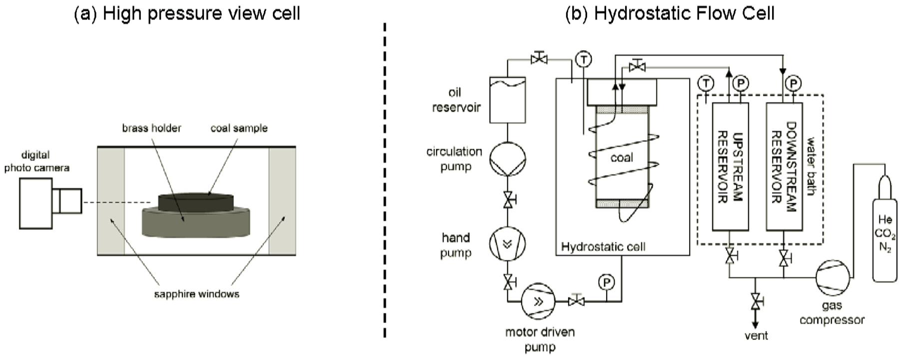

Figure 1 - Simplified schematics of the experimental equipments used in this study: (a) high pressure view cell, for performing free swelling experiments, and (b) hydrostatic flow cell, for performing gas injection experiments under constrained conditions.

2.1. Free swelling experiments

The swelling experiments were performed in a view cell, consisting of a cylindrical vessel with a volume of 50 cm^3 equipped with circular sapphire windows, which are orthogonal to the axis of the cylinder (Figure 1a) [4]. The view cell is immersed in a water bath and is equipped with a pressure transducer. The coal disc is placed on a brass holder, which is then positioned inside the view cell. The role of the brass holder is two-fold: first, it ensures to keep the coal disc in a horizontal position and secondly, it is taken as a reference in the evaluation of the coal diameter from the digital picture since its diameter will not be influenced by the fluid pressure. Then, the view cell is brought to the desired temperature and filled up to a certain pressure with the fluid to be measured. The coal disc is allowed to expand for two days to reach equilibrium conditions before a picture is taken and the diameter of the disc is determined using commercial image analysis software.

In these experiments, the coal disc expansion is unconstrained, being the effective pressure on the sample zero (defined as the difference between confining and pore pressure, $P_\mathrm{e} = P_\mathrm{c} - P$). Moreover, if isotropic behavior is assumed, the volumetric sample expansion s is obtained by the change in sample diameter only, as

$$s(P,T) = \frac{V - V_0}{V_0} = \frac{d^3 - d_0^3}{d_0^3} \tag{1}$$

where V and d are the volume and the diameter of the coal disc, respectively, and subscript 0 refers to the initial (vacuum) condition.

2.2. Flow experiments under constrained conditions

The flow experiments were conducted in a high pressure hydrostatic cell capable of withstanding confining pressures up to 1000 bar and modified to allow for gas flow through the sample [5]. A simplified schematic of this equipment is shown in Figure 1b. It is worth noting that in these experiments, the confining pressure P_c and the pore pressure P can be controlled separately, allowing imposing the desired effective pressure on the sample. The cylindrical sample is isolated from the confining fluid with a 4 mm thick polyvinyl chloride rubber jacket and is placed between two stainless steel disks connected to the tubing system and finally to two reservoirs: the upstream reservoir, which can be pressurized with the gas to be injected, and the downstream reservoir used to collect the gas which leaves the sample. The hydrostatic cell is kept at the desired temperature with a heating jacket, whereas the reservoirs are placed in a water bath which is maintained at the same temperature as the hydrostatic cell.

The transient step method was used to carry out the flow experiments. After having placed the sample into the hydrostatic cell, a confining pressure is applied and held constant. The sample is then flushed with the fluid to be used and, as an initial condition, reservoirs and sample are equilibrated with a fluid at the same pressure. A pressure change is then imposed at the upstream end of the sample and the system is allowed to equilibrate at a new pressure level. After reaching pressure equilibrium and allowing for at least two days for the fluid to adsorb, the pressure in the upstream reservoir is raised again to a new level, and a new measurement is carried out.

3. Modeling

A model that describes the dynamics of the system upstream reservoir-coal core-downstream reservoir has been applied to describe the behavior of the pressure in the two reservoirs observed during the flow experiments. This comparison allows obtaining insights on the dynamic behavior of important parameters such as the permeability. The model consists of one-dimensional mass balances accounting for gas flow, adsorption and swelling combined with mechanical constitutive equations for the description of porosity and permeability changes during injection.

The overall mass balance equation for the fluid in the coal core and the material balance for the adsorbed phase take the following form:

$$\frac{\partial(\varepsilon_\mathrm{t} c)}{\partial t} + \frac{\partial\left[(1-\varepsilon_\mathrm{t})q\right]}{\partial t} + \frac{\partial(uc)}{\partial z} = 0 \tag{2}$$

$$\frac{\partial\left[(1-\varepsilon_\mathrm{t})q\right]}{\partial t} = k_\mathrm{m}(1-\varepsilon_\mathrm{t})(q^* - q) \tag{3}$$

where c and q are the gas and adsorbed phase concentration, respectively; q^* is its equilibrium concentration in the adsorbed phase; k_m is its mass transfer coefficient; ε_t is the total interconnected porosity consisting of both macropores and cleats ; u is the superficial velocity; t and z are time and space coordinates. Note that since Helium is considered to be an inert, both adsorption on coal and mass transfer in the adsorbed phase are absent, and therefore Eq. (3) is considered for CO_2 only.

Cleat porosity, ε, and permeability, k, in the coalbed can then be predicted as [2]:

$$\frac{\varepsilon}{\varepsilon_0}=\left(\frac{k}{k_0}\right)^{1/3}=\exp\left[\frac{-C_1(P_c-P)-C_2E_Ys}{K\varepsilon_0}\right] \tag{4}$$

where P and P_c are fluid and confining pressure, respectively; s is the pressure dependent swelling; C_1 and C_2 are coefficients depending on the coal properties. In the above equation, the reference values of porosity and permeability apply to an unstressed coal in contact with a non-swelling gas at atmospheric pressure.

These equations are completed by several constitutive equations:

- Darcy's law, expressing velocity as a function of pressure gradient and permeability:

  $$u=v\varepsilon=-\frac{k}{\mu}\left[\frac{\partial P}{\partial z}-gM_\text{m}\left(z\frac{\partial c}{\partial z}+c\right)\right] \tag{4}$$

  where v is the interstitial velocity and ε the cleat porosity, P the pore pressure, k the permeability, μ the dynamic viscosity, g the gravitational acceleration and M_m the molecular weight of the fluid.
- An equation of state: data from the National Institute of Standards and Technology (NIST) have been interpolated with polynomials in the temperature and pressure range used in this study to relate pressure and viscosity to the fluid concentration [6].
- The adsorption and the swelling isotherms, giving q^* and s as a function of pressure and temperature:

  $$q^*=\frac{\rho_\text{ads}bq_mc}{1+bc} \tag{5}$$

  $$s=\frac{b_ss_mP}{1+b_sP} \tag{6}$$

  where ρ_ads is the coal bulk density; the saturation capacity per unit mass adsorbent and the Langmuir equilibrium constant are given by q_m and b for the adsorption isotherm and by s_m and b_s for the swelling isotherm, respectively.

Finally, upstream and downstream boundary conditions are given by simple reservoir material balances:

$$\left(\frac{\partial\rho}{\partial t}\right)_{z=0}=-\frac{A}{V_{US}}u\rho \qquad \left(\frac{\partial\rho}{\partial t}\right)_{z=L}=\frac{A}{V_{DS}}u\rho \tag{7}$$

where A and L are cross-sectional area and length of the sample, and V_US and V_DS are the volumes of the upstream and downstream reservoirs, respectively.

3.1. Solution procedure

The orthogonal collocation method has been applied to discretize in space the partial differential equations and the resulting system of ordinary differential equations has then been solved numerically using a commercial ODEs solver (in Fortran). The input parameters used for the model calculations are summarized in Table 1. The set of parameters to be estimated consists of the coefficients C_1 and the C_2 accounting respectively for changes in the porosity due to effective pressure and swelling, the absolute (unstressed sample) permeability k_0 and the mass transfer coefficients k_m. All parameters were estimated by reproducing the experimentally obtained transient steps using the model described above. In particular, the experiments with helium, i.e. the non-adsorbing (and non-swelling) gas, were used to obtain values for C_1 and k_0, and therefore are needed to study the effect of the confining pressure on the sample permeability. The experiments with the adsorbing CO_2 were used to estimate the value for the coefficients C_2 and of the mass transfer coefficient k_m, and therefore are needed to quantify the effects of adsorption and swelling on the flow dynamics.

Table 1 – Model input parameters and coal properties.

System properties			**Fluid properties**		
initial total porosity [-]	ε_t^0	0.051	Saturation capacity (adsorption) [mol/g]	q_m	2.38×10^{-3}
initial cleat porosity [-]	ε^0	0.031	Langmuir constant (adsorption) [m^3/mol]	b	3.55×10^{-3}
Poisson's ratio [-]	ν	0.26	Saturation capacity (swelling) [-]	s_m	4.90×10^{-2}
Young's elastic modulus [Pa]	E_Y	1.119×10^9	Langmuir constant (swelling) [Pa^{-1}]	b_s	3.80×10^{-7}
coal bulk density [g/cm^3]	ρ_{ads}	1.36	Molar Mass of adsorbate [g/mol]	M_{mCO2}	44.01
cross sectional area [cm^2]	A	4.73	Molar Mass of adsorbate [g/mol]	M_{mHe}	4.00
upstream reservoir volume [cm^3]	V_{US}	50.4			
downstream reservor volume [cm^3]	V_{DS}	15.2			

4. Results and Discussion

4.1. Free swelling experiments

Using the view cell, measurements of swelling were carried out using the three pure fluids CO_2, CH_4, N_2 and He at a temperature of 45°C and up to a pressure of 130 bar. In Figure 2 the isotropic swelling s is shown as a function of the pressure P for the different gases at a temperature of 45°C. In the figure, likewise all other studies [7], the extent of swelling increases monotonically with pressure up to a few percents for adsorbing gases, with CO_2 swelling coal more than CH_4 that swells more than N_2, whereas for helium, a non-adsorbing gas, volume changes are negligible. Therefore, in view of an ECBM operation, the displacement of CH_4 by CO_2 would lead to a net coal swelling, whereas its displacement by N_2 to a net shrinking. As shown in Figure 2 swelling isotherms can be effectively described with Langmuir-like equations.

In a recent study, repeated CO_2 swelling measurements on coal showed that changes are greater in the direction perpendicular to the bedding plane than in that parallel to it [8,9]. In another study, the observed differences between the two directions were very limited [9]. The results from the first case suggest that the anisotropic nature of the coal remains unchanged upon repeated exposure to the high-pressure gas, which is different from the behavior observed when organic solvents are used, where after the first exposure the coal behaved isotropically [10]. It is clear that further measurements are needed to clarify whether one or the other conclusion can be drawn. However, we believe that the data presented in Figure 2 are useful and the assumption of isotropic expansion acceptable, being the error associated to the experimental technique similar to the difference due to assuming in the two mentioned works either isotropic or anisotropic behavior.

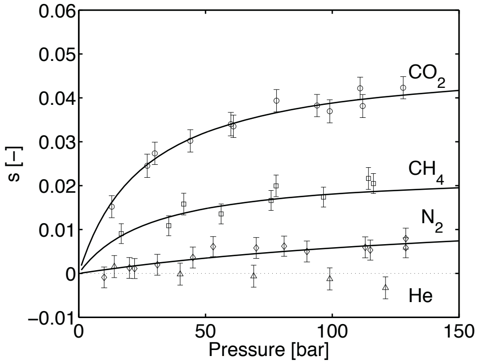

Figure 2 - Swelling of an unconstrained dry disc of the same coal from the Sulcis Coal Province as a function of the pressure, P, of CO_2, CH_4, N_2 and He at 45°C. Swelling is assumed to be isotropic. The experimental data (symbols) are well described as a function of pressure by Langmuir-like equations (solid lines), i.e. Eq. (6).

4.2. Flow experiments under constrained conditions

As it can be seen from Eq. (4), under constrained conditions the volume changes of the coal affect the porosity and permeability of the coal. Permeability is a parameter of key importance for the ECBM operation, since it controls injection pressure and gas production. In order to quantifiy this phenomenon, flow experiments under constrained conditions using the hydrostatic cell described above were conducted [5]. A selection of these experiments are presented here. In particular, experiments with Helium and CO_2 are shown, allowing to compare the coal sample behavior under injection of an inert gas like helium and an adsorbing (and swelling gas) like CO_2. All the experiments were carried out at 45°C, a temperature which is representative of the conditions of the coal seam in the Sulcis Coal Province in Sardinia, Italy.

In Figure 3 the transient steps obtained with Helium are shown, which were carried out by keeping the confining pressure constant at 100 bar and increasing the fluid pressure. The symbols are the experimental data whereas the solid lines correspond to the model results. A good agreement is observed between experiments and simulated transient steps, which were obtained by fitting the initial (unstressed) permeability k_0 (0.049 mD) and the effective pressure coefficient C_1 (4.676) to the experimental data. It is worth noting, that the obtained permeability k_0 is smaller than that of typical coalbeds, which ranges between 1 and 10 mD [11,12], but it is similar to the permeability values obtained in other laboratory studies [12,13]. This discrepancy between laboratory and results from the field can be attributed to the absence of the large fractures in the small samples which on the contrary are present in the coal seam and represent an important contribution as far as the gas flow is concerned. It can also be seen in Figure 3, that the transient step equilibration time increases with increasing effective pressure on the sample, i.e. $P_e = P_c - P_{eq}$. As given in Eq. (3), the permeability and the porosity decrease with increasing effective pressure, thus slowing down the flow process. This phenomenon corresponds in practice to the compression of the cleats under an external stress.

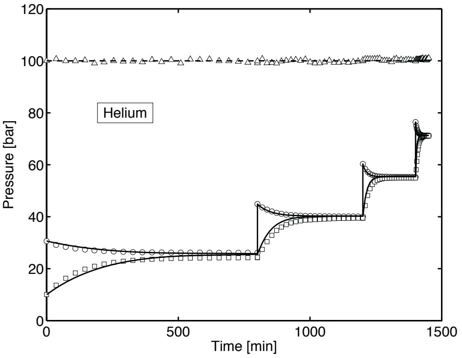

Figure 3 – Transient steps measurements at 45°C when Helium is injected. The confining pressure is kept constant at 100 bar. Symbols: Confining pressure P_c (△), upstream P_{US} (○) and downstream P_{DS} (□) reservoir pressures as a function of time. Solid lines correspond to model results.

Since the coal sample specific parameters are already known from the Helium experiments, the injection experiments performed with an adsorbing fluid allow investigating the effects of adsorption and swelling on the flow dynamics. Figure 4 reports an example of transient steps obtained with CO_2 when the confining pressure was kept constant at 100 bar. The symbols are the experimental values whereas the solid lines correspond to the model results, obtained by fitting the mass transfer coefficient, k_m (3.878×10^{-6} s^{-1}) and the swelling coefficient, C_2 (0.622) to the experimental data. A good agreement is achieved between experiments and simulated transient steps. The mass transfer coefficient is a lumped parameter which combines all the kinetic factors related to the gas diffusion in the coal matrix. Its reciprocal value corresponds to the sorption time constant used in other studies [14,15], whose values are in agreement with those found in this work, i.e. in the order of a few days.

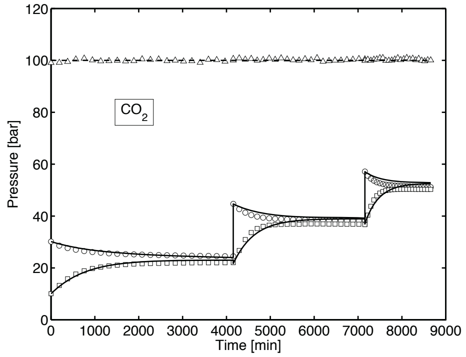

Figure 4 - Transient steps measurements at 45°C when CO_2 is injected. The confining pressure is kept constant at 100 bar. Symbols: Confining pressure P_c (△), upstream P_{US} (○) downstream P_{DS} (□) reservoir pressures as a function of time. Solid lines correspond to model results.

5. Conclusions

The study of the aspects related to the swelling of coal and their effects on the permeability has a specific practical aspect, namely the assessment of coal seam behavior during an ECBM operation. Laboratory measurements allow in fact to reproduce, or at least to approach, the conditions present in the coal seam. In this study, two techniques have been presented to investigate coal swelling both under un- and constrained conditions. The former is used to obtain swelling data under a static high pressure gas atmosphere, whereas the latter is used to perform flow experiments on coal cores confined under an external hydrostatic pressure. Moreover, a model describing the fluid flow trough the coal core has been derived, which includes mass balances accounting for gas flow, adsorption and swelling, and mechanical constitutive equations for the description of porosity and permeability changes during injection. The combination of the experimental data with the model predictions allowed to highlight and to understand the different fundamental aspects of the process dynamics related to effective pressure, adsorption and swelling phenomena.

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