

Experimental study of CO₂ sequestration by ECBM recovery: the case of Sulcis coal

Ronny Pini¹, Stefan Ottiger¹, Luigi Burlini², Giuseppe Storti³, Marco Mazzotti^{*1}, Roberto Bencini⁴, Fedora Quattrocchi⁴

¹ ETH Zurich, Institute of Process Engineering, Sonneggstrasse 5, CH-8092 Zurich, Switzerland

² ETH Zurich, Geological Institute, Sonneggstrasse 5, CH-8092 Zurich, Switzerland

³ ETH Zurich, Institute for Chemical and Bioengineering, ETH Hönggerberg, CH-8093 Zurich, Switzerland

⁴ National Institute of Geophysics and Volcanology, Via di Vigna Murata 605, 00143 Rome, Italy

Introduction

When coal seams are formed by compaction of plants, gases including methane are generated and accumulated into the coal cleats or adsorbed into the coal micropores. Such coalbed methane is normally recovered by means of reservoir-pressure depletion, i.e. by pumping out water and degassing the reservoir. A more attractive process with higher yields is the so-called Enhanced Coal Bed Methane recovery (ECBM), whereby carbon dioxide is pumped into the coal seam to displace methane thanks to higher CO₂ adsorptivity. Injecting CO₂ in unminable coal seams leads not only to methane recovery but also to CO₂ sequestration. The factors still limiting the implementation of ECBM recovery are economical, i.e. lack of penalties for CO₂ emissions, as well as technological and scientific, i.e. limited understanding of fundamental issues related to ECBM. Therefore, the goal of this study is to combine experimental measurements and modelling to characterize pure and multicomponent competitive adsorption of CO₂ and CH₄ on coal and study the coalbed dynamics using breakthrough experiments, including the effect of the injection of CO₂ on matrix swelling and permeability. Since December 2004, a feasibility study throughout the Sulcis Coal Province in Sardinia [Quattrocchi et al., 2004] is in progress and one of its objectives is to correlate the results of the mentioned experiments with the compositional patterns of the coal, considering its role in the CBM-ECBM exploitation.

Methodology and results

Coal Characterization: First results of the macerals analyses for the Sulcis coal show the composition of a typical low-medium rank, vitrinite-rich coal. These results are clearly suggesting that in the Sulcis Coal Province the ECBM exploitation is promising. The vitrinite reflectance coefficient R_o (around 0.7 and in any case > 0.4), the macerals composition as well as the Volatile Matter (38-55% for Sulcis Coal) allow to locate the coal as *High volatile C bituminous*, which is suitable for CBM and ECBM purposes. Experimental measurements of the Gas-In-Place (GIP) are also planned. Namely, fresh coal samples are transferred from the field to the laboratory using a suitable high-pressure container and analyzed in terms of

* Corresponding Author: mazzotti@ipe.mavt.ethz.ch, +41 44 632 2456

amount and composition of the released gas. According to the Sulcis coal composition, expected total gas generation is about 200 mL/g of CH₄ at atmospheric pressure.

Adsorption: In the case of adsorption, we have been investigating different systems, e.g. CO₂ on silica gel and on 13X zeolite [Rajendran et al., 2002] and on Akabira coal [Toribio et al., 2005]. For the measurement of the excess adsorption, a Rubotherm magnetic suspension balance has been used (Rubotherm, Bochum, Germany). It allows to measure the suspended mass with an accuracy of 0.01 mg and can be operated at pressures up to 450 bar and temperatures up to 250°C. Besides its precision, a key feature of the balance is that it allows for the direct measurement of gas density, thus encompassing the need of an additional pressure measurement to be combined with a reliable equation of state.

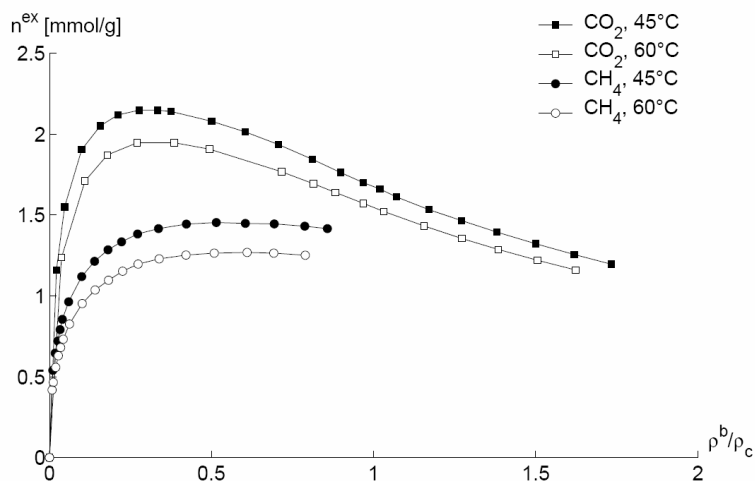


Figure 1. Adsorption of CO₂ and CH₄ on coal from the Sulcis province (Sardinia, Italy)

The adsorption of both pure CO₂ and CH₄ has been measured on Sulcis coal with the magnetic suspension balance. In Figure 1, the excess adsorption is plotted against the reduced density, i.e. the density of the fluid divided by its critical density. The shape of the isotherms exhibits the expected behavior for an excess adsorption isotherm: first the excess rises in the low density range, reaches a maximum and then decreases almost linearly beyond the critical density, as observed for standard adsorbents such as silica gel. As expected, the coal adsorption capacity is larger for CO₂ than for CH₄, an additional advantage when ECBM is primarily used to sequester carbon dioxide.

Coalbed dynamics: Another important feature of the coal under adsorption conditions is represented by the associated change in volume which, in turn, is affecting porosity and permeability of the bed. In considering ECBM production one can distinguish two opposite effects: on the one hand, the desorption of methane causes shrinkage of the coal matrix, improving the permeability, on the other hand the adsorption of carbon dioxide causes the coal matrix to swell, resulting in a reduced permeability of the coal which makes the gas transport slower, thus resulting in a lower injection capacity. For a successful ECBM, the understanding of the actual swelling behavior and of its impact on the permeability is therefore a key factor.

In order to study how the permeability could change, experiments reproducing in situ conditions have to be run under confining pressure. In this study this will be carried out in a setup consisting of a hydrostatic cell, connected to two servo-controlled pumps (NOVA Swiss, Switzerland) that allow controlling the pore and confining pressure, respectively (Figure 2). Carbon dioxide is used as the pore fluid, while oil is used for the confining

pressure system. With this setup, three main types of experiments are usually carried out: the constant-pressure difference, the transient step and the pore pressure oscillation method. Being the last operating mode the most effective in the case of systems exhibiting very small permeability values, such a kind of experiments will be initially performed.

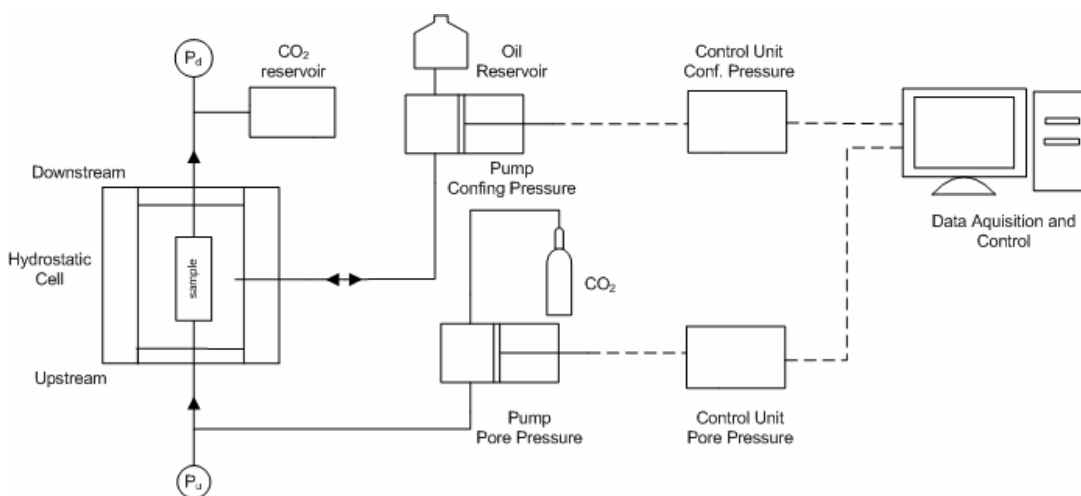


Figure 2. Hydrostatic cell used for dynamic experiments

The results of the dynamic experiments will be simulated by a suitable mathematical model aimed to evaluate the corresponding permeability. Representing the coal seam as a series of matrix blocks separated by fractures (cleats), the coalbed dynamic is described by two mass balances, one for the cleat system and another for the matrix. The flow in the cleats is convective and governed by Darcy's law, while the transport inside the coal micropores is by diffusion. The adsorption into the matrix is described by a suitable equilibrium isotherm evaluated by fitting the experimental data provided by the gravimetric measurements (Figure 1).

Conclusions

Different experimental techniques have been developed and applied to characterize equilibrium and dynamic behaviour of a coal for ECBM application. First results are promising in terms of coal composition and adsorption capacities. Further experiments are in progress to improve our understanding of the process under in-situ conditions. Among the future perspectives of this project, the adsorption of N₂ will be also considered in order to analyze the potential of ECBM when flue-gas instead of CO₂ is used.

References

- Hocker** T., A. Rajendran, M. Mazzotti, 2003: Measuring and modeling supercritical adsorption in porous solids. Carbon dioxide on 13X zeolite and on silica gel. *Langmuir* **19** (2003), pp.1254-1267.
- Quattrocchi** F., R. Bencini, D. Cinti, G. Galli, L. Pizzino, N. Voltattorni, A. Navarra, 2004: CO₂ geological sequestration in Italy: state of art and potential sites. In Proc. WRI-XI, June, 2004, New York, USA, pp. 573-576.
- Toribio** M., Y. Oshima, S. Shimada, R. Pini, S. Ottiger, G. Storti, M. Mazzotti, 2005: Paper presented at ICCS&T, October 9-14, 2005, Okinawa, Japan.