



Report on the relation between uptake, swelling behaviour, stress state and injectivity in coal: injection of CO₂ in 'dry' coal and in coal with water and methane present

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1 Executive Summary (restricted)

The injection of CO₂ in coal seams while simultaneously extracting methane is a promising CO₂ storage method. This is referred to as enhanced coal-bed methane (ECMB). Recent research, field trials and commercial projects demonstrated the feasibility of ECMB. However, the detailed design, planning and execution of any new ECBM project requires extensive knowledge about the site characteristics and about the site-specific coal-CO₂ interactions. The Chemelot site of DSM was initially proposed as a demonstration site for the CATO 2 project but was later withdrawn. Several studies of CO₂ sorption in coal were conducted as a detailed extension of the research started under the first CATO project. Extensive data on the CO₂ sorption and coal swelling were determined for several coal samples. It has conclude that permeability reducing due to coal swelling induced by CO₂ sorption could complicate the application of ECBM in European coal seams because of their low permeability.

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2 Applicable/Reference documents and Abbreviations

2.1 Applicable Documents

(Applicable Documents, including their version, are documents that are the “legal” basis to the work performed)

	Title	Doc nr	Version
AD-01a	Beschikking (Subsidieverlening CATO-2 programma verplichtingnummer 1-6843	ET/ED/9078040	2009.07.09
AD-01b	Wijzigingsaanvraag op subsidieverlening CATO-2 programma verplichtingennr. 1-6843	CCS/10066253	2010.05.11
AD-01c	Aanvraag uitstel CATO-2a verplichtingennr. 1-6843	ETM/10128722	2010.09.02
AD-01d	Toezegging CATO-2b	FES10036GXDU	2010.08.05
AD-01f	Besluit wijziging project CATO2b	FES1003AQ1FU	2010.09.21
AD-02a	Consortium Agreement	CATO-2-CA	2009.09.07
AD-02b	CATO-2 Consortium Agreement	CATO-2-CA	2010.09.09
AD-03a	Program Plan 2009	CATO2-WP0.A-D.03	2009.09.17
AD-03b	Program Plan 2010	CATO2-WP0.A-D.03	2010.09.30
AD-03c	Program Plan 2011	CATO2-WP0.A-D.03	2010.12.07

2.2 Reference Documents

(Reference Documents are referred to in the document)

	Title	Doc nr	Version/issue	Date
	Calibration and testing of models to experimental data			

2.3 Abbreviations

(this refers to abbreviations used in this document)

ECBM	Enhanced coal-bed methane

3 Progress report

3.1 Introduction

The structure, composition and physical properties of natural coals vary with the depositional conditions, burial history and geographic origin of the coal. Nevertheless, coals have several generic characteristics that are important when considering CO₂ storage combined with the extraction of CH₄, the enhanced coal-bed methane (ECBM) process.

A natural coal matrix is characterised by a macro- and micro-porosity which contains sorbed and free methane. Furthermore, the coal is characterized by micro-fractures (cleats) and by larger scale fractures. These cleats and fractures function as low resistance flow paths for the fluids.

The ECBM process involves several phenomena, of which the competitive adsorption of CO₂ and CH₄ is one of the most important. Despite the extensive work done to date, there is still a need of experimental data on these sorption phenomena under in-situ conditions. For each storage site, data gathering under the specific site conditions is an essential part of the design of the CO₂ storage and CH₄ production operation.

3.2 Results and discussion

3.2.1 CO₂ sorption on coal

High pressure isotherms are critically important but their measurement proved to be rather challenging: available data suffers from large scatter. To address these issues, a collaborative research project involving several European research institutions was undertaken (details can be found in Gensterblum et al., 2009 and 2010). The specific goal of the inter-laboratory collaborative research was to assess existing high-pressure CO₂ sorption data on coals and to identify ways to improve their quality. In the first phase of the project, the sorption isotherms of CO₂ on coal Filtrasorb 400 (F400) were measured. The data reported by the participant laboratories showed excellent agreement. In the second phase of the project the excess sorption isotherms for the adsorption of CO₂ on three coal types with increasing maturity, namely lignite, bituminous coal and semi-anthracite, were measured at 318 K and at pressures up to 16 MPa, using either the manometric method or the gravimetric method. The manometric method was used at the TU Delft (Netherlands) and the RWTH Aachen University (Germany) while the gravimetric method was used at the University of Mons (Belgium). The measured sorption isotherms showed a maximum for the three coal types. The maximum was nearly 1.77 mole of CO₂/kg of coal for lignite and around 1.37 mole of CO₂/kg of coal for both bituminous coal and semi-anthracite. The pressures at which the maximum excess sorption occurred decreased with increasing coal maturity: 6.89 MPa for the lignite, 6.68 MPa for the bituminous coal and 5.89 MPa for the semi-anthracite. Through this study, it was established that the excess isotherms for CO₂ sorption on natural coals, can be measured accurately at high pressures, under supercritical conditions for the CO₂.

3.2.2 CO₂ sorption and coal swelling

The excess isotherms for the adsorption and desorption of He, N₂, CH₄ and CO₂ on dry Selar Cornish coal were studied by the manometric method at two temperatures (318 and 338 K) at pressured up to 16 MPa (Van Hemert, 2009). The sorption-desorption isotherms for N₂, CH₄ did not show any hysteresis, whereas the CO₂ shows some hysteresis attributed to experimental artefacts.

The equilibration time was found to increase according to increasing molecular weight of the gases, i.e. He, N₂, CH₄, and CO₂, suggesting that the specific diffusivities of the gases play a significant role in the sorption process. Furthermore, the measured data showed that maximum of the excess sorptions N₂:CH₄:CO₂ are in good agreement with literature (e.g., Busch et al., 2003). This ratio confirms that, theoretically, considerably more CO₂ can be stored in Selar Cornish coal than the amount of CH₄ present.

The swelling and shrinkage of the Selar Cornish coal during sorption and desorption of CO₂, respectively was also investigated (Battistutta et al., 2010). The swelling and shrinkage ratios were measured using strain gauges during injection or extracting CO₂ from unconfined cubic samples. Experiments were done at 321 K and at pressures up to 4.1 MPa. The induced swelling and shrinkage resulted in a fully reversible mechanical deformation of the samples.

The density of the sorbed CO₂, extrapolated from the excess sorption isotherms calculated including the swelling, was unrealistically high. This was due to the assumption that the porosity of coal ($\phi = 0.03$) does not change with swelling. Later it was recognized that swelling induced by CO₂ sorption leads to much larger changes of the porosity. This improved considerably the sorption isotherms taking into account swelling.

3.3 Conclusions and outlook

Inter-laboratory studies of the sorption showed that the manometric and gravimetric method are adequately accurate for determining sorption isotherms of coal. It is recommended to further evaluate these methods with other coal types.

Sorption and extraction of CO₂ for Selar Cornish coal correlate well with reversible swelling and shrinkage behavior. More research is needed to understand the swelling behavior and its effect on CO₂ storage.

3.4 References

Gensterblum, Y., P. van Hemert, et al. (2009). European inter-laboratory comparison of high pressure CO₂ sorption isotherms. I: Activated carbon. *Carbon* **47**(13): 2958-2969.

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