



HAL
open science

Simulation of CO₂ storage in coal seams: Coupling of TOUGH2 with the solver for mechanics CODE_ASTER®

Annick Loschetter, Farid F. Smaï, Serigne Sy, André Burnol, Aurélien Leynet,
Stéphane Lafortune, Alain Thoraval

► To cite this version:

Annick Loschetter, Farid F. Smaï, Serigne Sy, André Burnol, Aurélien Leynet, et al.. Simulation of CO₂ storage in coal seams: Coupling of TOUGH2 with the solver for mechanics CODE_ASTER®. TOUGH symposium 2012, Sep 2012, Berkeley, United States. pp.Session IX: Carbon Dioxide Storage III. hal-00722452

HAL Id: hal-00722452

<https://brgm.hal.science/hal-00722452>

Submitted on 19 Nov 2012

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

SIMULATION OF CO₂ STORAGE IN COAL SEAMS: COUPLING OF TOUGH2 WITH THE SOLVER FOR MECHANICS CODE_ASTER®

A. Loschetter¹, F. Smai¹, S. Sy¹, A. Burnol¹, A. Leynet¹, S. Lafortune² and A. Thoraval²

¹French Geological Survey (BRGM)

BP 6009 45060 Orléans, France

e-mail: a.loschetter@brgm.fr

²French National Institute for Industrial Environment and Risks (INERIS)

BP 2, 60550 Verneuil-en-Halatte, France

ABSTRACT

Amongst the various geological storage options currently under consideration, CO₂ storage in coal formations presents the most economic potential for middle-term spreading but also the most uncertainties and technical difficulties. Indeed, the profit from methane recovery could cover part of the costs, but little testing for this technique has been conducted in European coal deposits. Therefore, there is a need for an in situ injection study at an intermediate scale between a laboratory and an industrial pilot. This is the aim of the CARBOLAB project.

This study investigates coupled flow and mechanical processes that will take place around the injection point at the bottom of the Montsacro mine in Asturias, Spain. Reservoir simulators, such as TOUGH2, have been used for CO₂ storage in saline aquifers and oil and gas reservoirs (Oldenburg et al., 2004; Xu and Pruess, 2001). However, until now, this tool did not represent the adsorption process, a key point for explaining the gas storage and gas migration through coal seams. Moreover, CO₂ sorption and exchange with adsorbed CH₄ are associated with mechanical processes like swelling/shrinkage. In order to quantify the strain and stress fields due to changes in the fluid pressure field and to account for stress/sorption dependent porosity/permeability effects, an efficient coupling between TOUGH2/EOS7C, a special module with an Extended Langmuir Sorption model, and the solver for mechanics CODE_ASTER® has been developed by BRGM. This new tool will be first verified by intercomparison with the COMSOL-based tool developed by INERIS and, afterwards, validated with in situ data acquired during the ongoing injection at the Montsacro mine.

INTRODUCTION

The injection of CO₂ in deep unmineable coal seams is one way of reducing greenhouse gas emissions, whilst at the same time it enables recovering a high value gas — methane. The potential feasibility and economic interest of this option depend essentially on the coal permeability and the quantity of recoverable methane. This option for CO₂ storage is the one that presents the more uncertainties and technical difficulties. Understanding thermo-hydrological as well as mechanical processes is a serious prerequisite to demonstrate the feasibility of injecting CO₂ in coal seams and is a major handicap for identifying the reliability and safety of this sequestration option. Little testing for this technique has been conducted on European coal deposits. Therefore, there is a need for an in situ injection study at an intermediate scale between a laboratory and an industrial pilot.

The CARBOLAB project intends to advance in this specific target, by performing underground tests of CO₂ injection and Coal Bed Methane (CBM) production in a specially conditioned panel of a coal mine in Spain, and also by developing adequate modeling tools. These developments are necessary for sizing the experiments through preliminary simulations; then the experimental results will be compared to simulations of experimental data in order to validate and improve the models. Modeling developments aim at providing a thermo-hydro-mechanical reservoir simulator taking into account the adsorption process, the mechanical processes like swelling/shrinkage, and stress/sorption dependent porosity/permeability effects. These effects may indeed play a significant role for simulating CO₂ injection in coal formations.

We first present the implementation of the thermo-hydro-mechanical simulator into a sequential coupling of CODE_ASTER® for mechanics and TOUGH2 for multiphase flow. Then the simulation scenarios are introduced and the results are discussed.

MODEL DESCRIPTION

The modeling of CO₂ injection in a coal seam copes with significant issues which are linked with the specificity of coal characteristics: the adsorption of CO₂ and CH₄ is a predominant phenomenon; effective stresses are significantly impacted by adsorption processes; porosity and permeability depend on stress and adsorption features. In order to consistently model these processes, we propose to couple TOUGH2 including a modified version of module EOS7C with an Extended Langmuir Sorption model, and the solver for mechanics CODE_ASTER®.

Thermo-Hydrological Processes

For modeling the thermo-hydrological processes, the multiphase non-isothermal transport code TOUGH/EOS7C provides a reliable and efficient base. However, until now, this tool did not represent the adsorption process. The developments in this code thus focused on introducing adsorption processes in the module “EOS7CS” based on module “EOS7C”. The conservation equation was modified by introducing a new term that corresponds to the subtraction of adsorbed quantities to the free flow:

$$\partial_t \left((1 - \Phi) \rho_m M^i Q_{sorb}^i + \sum_{\alpha} \Phi S_{\alpha} \rho_{\alpha}^i \right) - \text{div} \left(\sum_{\alpha} \rho_{\alpha}^i q_{\alpha} + d_{\alpha}^i \right) = 0$$

with Φ the porosity, ρ_m the matrix density (kg/m³), M^i the molar mass of component i (kg/mol), S_{α} the saturation of phase α , Q_{sorb}^i the molar adsorbed quantity of component i (mol/kg), ρ_{α}^i the density of component i in phase α (kg/m³), d_{α}^i the diffusion flux of component i in phase α (kg/m²/s), q_{α} the flow velocity of phase α (m/s). d_{α}^i is obtained by the Fick law: $d_{\alpha}^i = \rho_{\alpha} D_{\alpha,eff}^i \nabla x_{\alpha}^i$ with x_{α}^i the mass

fraction of component i in phase α and $D_{\alpha,eff}^i = \tau \phi D_{\alpha}^i$ the effective diffusion coefficient of component i in phase α (m²/s), that includes tortuosity coefficient τ , porosity and the diffusion coefficient D_{α}^i (m²/s). The Darcy law is used for computing $q_{\alpha} = \lambda_{\alpha} k \nabla p_{\alpha}$, with p_{α} the total pressure of phase α , λ_{α} the mobility of phase α (that depends on saturation in phase α), and k the permeability.

This conservation equation requires implementing a relation that links Q_{sorb}^i with the parameters that control adsorption. A literature review shows that the adsorption is mainly controlled by the values of partial pressures of the components:

$$Q_{sorb}^i(p_g^1, p_g^2, \dots)$$

with p_g^i the partial pressure of component i in the gas phase (Pa). A first relation (between the adsorbed quantity and the partial pressure for each component) was implemented with a Langmuir Sorption model. However, this model is not consistent for representing a competitive sorption between different gas species onto the coal surface sites. Therefore an Extended Langmuir Sorption model was implemented and the adsorbed quantity for CO₂ is computed as (and symmetrically for CH₄):

$$Q_{sorb}^{CO_2} = Q_{max}^{CO_2} \times \frac{\alpha^{CO_2} p_g^{CO_2}}{1 + \alpha^{CO_2} p_g^{CO_2} + \alpha^{CH_4} p_g^{CH_4}}$$

with $Q_{max}^{CO_2}$ the constant of maximal adsorbed quantity for CO₂ and α^i the parameters of the Extended Langmuir Sorption Model. In addition to the integration of sorption process, the variation of permeability depending on porosity was also implemented in the new module. The relation currently used is a cubic relation:

$$\frac{k}{k_0} = \left(\frac{\Phi}{\Phi_0} \right)^3$$

with k the permeability, k_0 the reference permeability, Φ the porosity, Φ_0 the reference porosity.

Mechanical Processes

For modeling the mechanical processes, the open CODE_ASTER® (provided by EDF) was

used. CODE_ASTER® offers a full range of multiphysical analyses and modeling methods and enables to model geomaterials. The aim is to simulate with CODE_ASTER® the significant impact of adsorption processes on effective stresses.

The equilibrium equation is written as:

$$\text{div}(\sigma_{ij}) + f_i = 0$$

with σ_{ij} the component of the total strain tensor and f_i the component of the body force. The coal is considered as a homogeneous and elastic continuum, and the system is isothermal, so that the strain-displacement relation can be expressed as:

$$\varepsilon_{ij} = \frac{1}{2} (\nabla u_i + \nabla u_j)$$

with ε_{ij} the component of the strain tensor and u_i the component of the displacement. The effects of gas-sorption on the strain can be introduced analogously as a thermal contraction in the stress-strain relationship (Shi and Durucan, 2004):

$$\varepsilon_{ij} = \frac{1}{2G} \sigma_{ij} - \left(\frac{1}{6G} - \frac{1}{9K} \right) \sigma_{kk} \delta_{ij} + \frac{b}{3K} p_{\text{pores}} \delta_{ij} + \frac{\varepsilon_{\text{sorb}}}{3} \delta_{ij}$$

with G the shear modulus of coal, K the bulk modulus of coal matrixes, b the Biot's coefficient, p_{pores} the gas pressure in the pores, $\varepsilon_{\text{sorb}}$ the sorption-induced strain and δ_{ij} the Kronecker delta. Combining these equations yields:

$$G \nabla^2 (u_i) + \frac{G}{1-2\nu} \nabla (\varepsilon_v) - b \nabla (p_{\text{pores}}) - K \nabla (\varepsilon_{\text{sorb}}) + f_i = 0$$

with ν the Poisson's ratio of the coal-fracture assemblage, ε_v the volumetric strain. Owing to pore pressure and sorption induced strain, the term $(b p_{\text{pores}} - K \varepsilon_{\text{sorb}})$ is implemented in our model as a load, because of the chaining strategy chosen as "fixed strain split" (Settari and Mourits, 1998).

Coupling between TOUGH2/EOS7CS and CODE_ASTER®

In order to couple both codes, a supervisor was elaborated in python language. This supervisor manages the launch of both codes and ensures the computing of input parameters for each code. As illustrated on Figure 1, the dialogue between the mechanic and thermo-hydrodynamic codes requires the implementation of three main relations:

- i. for updating porosity that depends on stress and adsorption features (permeability is updated within TOUGH2 through the relation porosity-permeability);
- ii. for computing the gas pressure in pores;
- iii. for computing the sorption-induced strain.

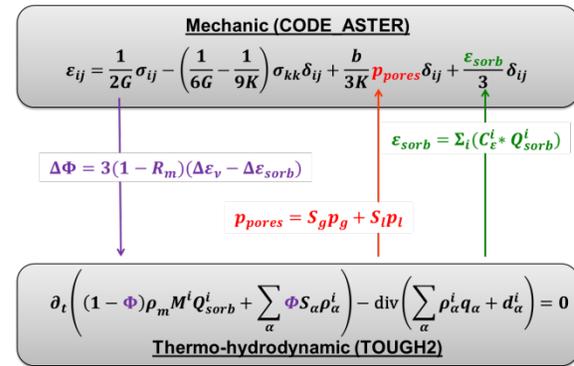


Figure 1. Illustration of relations that enable coupling mechanic with thermo-hydrodynamic equations

For the first point (i), coal matrix porosity (and permeability) changes are controlled by the mechanical effective stress change (Liu et al., 2010):

$$\Delta \Phi = 3(1 - R_m)(\Delta \varepsilon_v - \Delta \varepsilon_{\text{sorb}})$$

with $R_m = \frac{E}{E_m}$ the elastic modulus reduction ratio, E the equivalent Young's modulus of the coal-fracture assemblage and E_m the Young's modulus of the coal matrix. In this relation, the change in porosity and permeability due to coal matrix is neglected, only the change from the coal fractures is considered. The porosity changes are computed from the outputs of CODE_ASTER® and are integrated in the inputs of TOUGH2.

Concerning (ii), p_{pores} is the average pore pressure and is calculated as follows:

$$p_{\text{pores}} = S_g p_g + S_l p_l$$

with S_g the gas saturation, p_g the total gas pressure, S_l the liquid saturation, p_l the total liquid pressure. Liquid pressure and gas pressure are extracted from the outputs of the hydraulic module TOUGH2 and used in CODE_ASTER® for determining the pore pressure.

Concerning (iii), the gas sorption-induced strain $\varepsilon_{\text{sorb}}$ is presumed to result in volumetric strain only. According to Chen et al. (2010), experimental evidence supports the use of the extended Langmuir isotherm equation for representing the adsorption of gas mixtures:

$$\varepsilon_{\text{sorb}} = \sum_i (C_\varepsilon^i Q_{\text{sorb}}^i)$$

where C_ε^i is the gas volumetric strain of component i at infinite pressure. The adsorbed quantities are extracted from the outputs of TOUGH2 and used in CODE_ASTER® for determining the gas-sorption induced strain.

For efficient coupling the supervisor works with a parallel algorithm, which means that at time n , TOUGH2 calculates the total pressure and adsorbed quantities for time $n + 1$ while CODE_ASTER® calculates the induced porosity changes that enable updating the porosity and permeability for time $n + 1$.

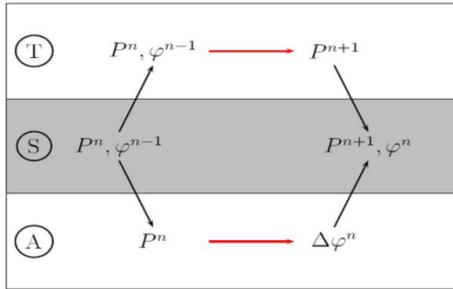


Figure 2. Illustration of the parallel algorithm between TOUGH2 (T), CODE_ASTER® (A) and the supervisor (S).

SIMULATION SCENARIOS

The simulations presented in this paper aim at testing the thermo-hydro-mechanical simulator presented above. The parameters are chosen in

order to represent realistic values, but the modeled scenario is purely imaginative. It is planned to perform a second set of simulations that will be based on the experimental scenario. The following choices (for input parameters as well as for boundary and initial conditions) have been made consensually by all attendees using data obtained in the CARBOLAB research projects and other research projects (especially the CHARCO project), data from scientific literature (especially Defossez, 2011) and expert opinion if necessary.

Data and input parameters used

The properties that are used for the coal seam and the shale are presented in Table 1.

Table 1. Properties of rocks

Properties	Coal	Shale
Density ρ (kg/m ³)	1 400	2 300
Porosity Φ	0.086	0.042
Young modulus E (GPa)	2	5.6
of the coal-fracture system		
Poisson coefficient ν	0.3	0.26
Biot coefficient b	0.9	0.8
Permeability k (m ²)	$1.8 \cdot 10^{-16}$	$8.1 \cdot 10^{-18}$
Gas diffusion (cm ² /s)	$1.6 \cdot 10^{-6}$	$1.6 \cdot 10^{-6}$

Shi and Durucan (2003) reported CO₂ diffusion coefficient values, estimated by fitting effluent composition data for pulverized coals, of 5×10^{-8} to 5×10^{-6} cm²/s for micropores and 2×10^{-5} to 7×10^{-4} cm²/s for macropores. It has been found experimentally that the matrix of the coals from the Montsacro Mine is composed of approximately 29% micropores and 68% macropores (P. Defossez, personal communication), so a representative value was used (1.6×10^{-6} cm²/s). For simplicity (and lack of data) the coefficients were made the same for CO₂ and CH₄.

The bulk modulus K is obtained by:

$$K = \frac{E}{3 \times (1 - 2\nu)}$$

For obtaining the Young modulus of the coal matrix E_m , we use the formula:

$$K_m = \frac{K}{1 - b}$$

By assuming that the Poisson coefficient for the coal matrix is the same as for the coal-fracture assemblage, we obtain:

$$E_m = 3K_m \times (1 - 2\nu) = \frac{3K \times (1 - 2\nu)}{1 - b}$$

The thermo-hydrodynamic model requires noticing the van Genuchten parameters (cf. Table 2) for relative permeability and capillary pressure functions.

Table 2. van Genuchten properties

Properties	Coal	Shale
van Genuchten P_0 (Pa)	1×10^5	1×10^6
van Genuchten P_{max} (Pa)	1×10^7	1×10^7
van Genuchten m	0.5	0.5
S_{lr}	0.003	0.003
S_{gr}	0.	0.
S_{ls}	1.	1.

For the dissolution of CO_2 , the coefficient of the Henry law is set to $1.53 \cdot 10^8$ Pa at $25^\circ C$. The chosen parameters for the extended Langmuir Sorption model are presented in Table 3:

Table 3. Extended Langmuir Sorption Model parameters

Properties	CO_2	CH_4
Q_{max}^i (mol/kg)	$1.69 \cdot 10^{-3}$	$8.1 \cdot 10^{-4}$
α^i (Pa^{-1})	$1.12 \cdot 10^{-6}$	$1.57 \cdot 10^{-6}$

Concerning the gas volumetric strains $C_\epsilon^{CO_2}$ and $C_\epsilon^{CH_4}$, the references are very scarce. Consequently, it was chosen to perform a sensitivity analysis on these parameters.

Reference Case

A first simplified base case has been elaborated to test the developed simulator. It consists in injecting CO_2 in a non-saturated porous coal seam with a pressure controlled injection of 4.5 MPa, during 24 hours, and then in relaxing during 96 hours (total simulated duration is therefore 120 hours). In addition to the coal layer, two surrounding rocks (shale) are considered in the model. We consider a 2D-axisymmetric model whose geometry is represented on Figure 3. The model length is 25 m.

The boundary conditions for mechanics are represented on Figure 3: a no-displacement condition is imposed on the upper horizontal boundary and on the injection axis; the stress field is imposed on the lower horizontal boundary (set to 18.5 MPa) and on the lateral boundary for $x = 25$ m (set to 12.25 MPa).

The boundary conditions for thermo-hydrodynamics are also represented on Figure 3: a no-flow condition is imposed on the upper and lower horizontal bounds; the gas composition and pressure are imposed on the lateral bound at $x = 25$ m (equal to the initial composition and pressure); the injection axis corresponds to a no-flow condition, except in the coal. The initial conditions are presented in Table 4.

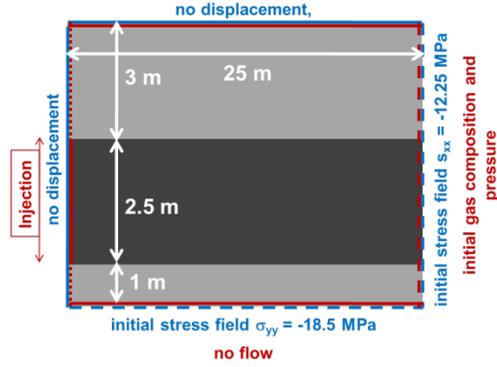


Figure 3. Geometry and boundary conditions (color red: hydrodynamic boundary conditions; color blue: mechanical boundary conditions)

Table 4. Initial conditions

Properties	
Initial water saturation	0.046
Initial gas saturation	0.954
Initial CH_4 pressure (MPa)	1.4955
Initial CO_2 pressure (MPa)	0.0045

In the reference case, the gas volumetric strains $C_\epsilon^{CO_2}$ and $C_\epsilon^{CH_4}$ are given in the Table 5.

A first set of simulations has been performed in order to test the influence of the mesh on results. To obtain a satisfying convergence, it is necessary to refine the mesh in the first meter (mesh length of 0.02 m) and to keep mesh not distorted, which imposes using the same order of

magnitude for the mesh height as for the mesh length. For computing time reasons and since the results were significantly the same with and without surrounding rocks (shale), it was chosen to keep only the coal layer in the modeling. The number of cells in the reference case is 7000 (20 for height, 350 for length).

Sensitivity Analysis

The gas volumetric strains $C_{\varepsilon}^{CO_2}$ and $C_{\varepsilon}^{CH_4}$ are unknown (because of lack of data) and may have a strong impact on results. Therefore, it was decided to perform a sensitivity analysis on a large range of values for these parameters. This wide range of values may have a physical sense since it is admitted that the swelling behavior of a coal depends highly on its composition. The values of the different test cases are presented in Table 5.

Table 5. Sensitivity analysis parameters

Gas volumetric strains	CO ₂	CH ₄
Case A	0	0
Case B	2,13 10 ⁻¹	5,85 10 ⁻¹
Case C (Reference case)	2,13	5,85
Case D	10,65	29,25
Case E	21,3	58,5

All other parameters and modeling choices are kept identical to the reference case.

RESULTS AND DISCUSSION

The output variables of interest include the pressure, the CO₂ flux, the CO₂ fraction, the porosity, the permeability, the adsorbed quantity of CO₂ and CH₄. These variables may be observed on time-dependent or space-dependent scales. In this section, the results obtained for these variables are presented and discussed for the different simulation cases. The computing time is also an interested observable value to assess the efficiency of the simulator. It was about the same for all the simulations, around 6 hours.

Reference Case

The pressure-controlled injection followed by relaxation is visible on Figure 4. It can be observed that the pressure increase exceeds 0.5 MPa only in the first meters.

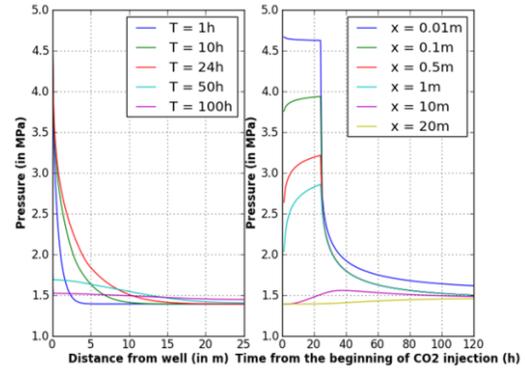


Figure 4. Evolution of pressure, function of space (on left) and function of time (on right)

The porosity varies in a narrow range: it decreases up to 0.085 and increases up to 0.08755 during the injection period, as illustrated on Figure 5. The porosity changes are indeed controlled by two competitive phenomena: the pressure increase tends to increase the porosity while the swelling of coal due to adsorption tends to decrease it.

The analysis of permeability evolution (not shown here) drives to the same conclusions, since permeability is supposed to be connected to porosity by a cubic relation.

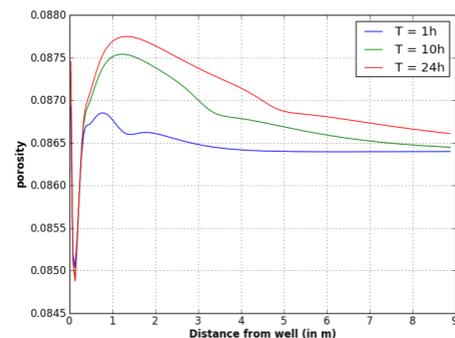


Figure 5. Evolution of porosity depending on space

The flux of CO₂ is maximal at the beginning of injection because the pressure gradient is maximal. The order of magnitude of the flux is 0.008 kg/s, i.e. 691 kg during the 24 hours of injection.

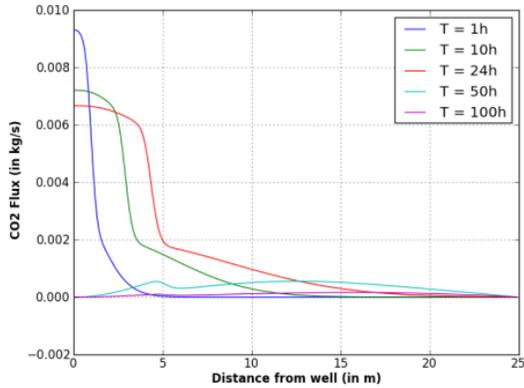


Figure 6. Evolution of CO₂ flux depending on space

The adsorbed quantities of CO₂ and CH₄ are represented on Figures 7 and 8. The orders of magnitude represent the fact that a very large quantity of CO₂ can be stored in coal compared to CH₄. The local quantity of adsorbed CH₄ first increases due to the pressure increase, and then strongly decreases after the arrival of CO₂ due to the competition for the coal sorption sites.

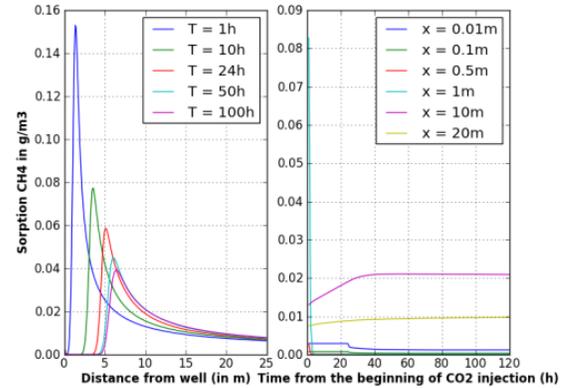


Figure 8. Evolution of CH₄ adsorbed, function of space (on left) and of time (on right)

Sensitivity Analysis

The results of the sensitivity analysis on the gas volumetric strains are presented below (Figure 9 and Figure 10) for the two more influenced outputs: porosity and CO₂ flux. It is noticeable that the gas volumetric strains impact the evolution of porosity. For low-swelling coals, the porosity increases, while for high-swelling coal, the porosity may locally decrease significantly. The CO₂ flux is impacted by the porosity changes: for case C, the flux decreases only slightly (compared to case A) but for case E the flux decreases significantly. This simulation result could be an explanation of the clogging in some specific coals (RECOPOL project in Poland) and not in other projects.

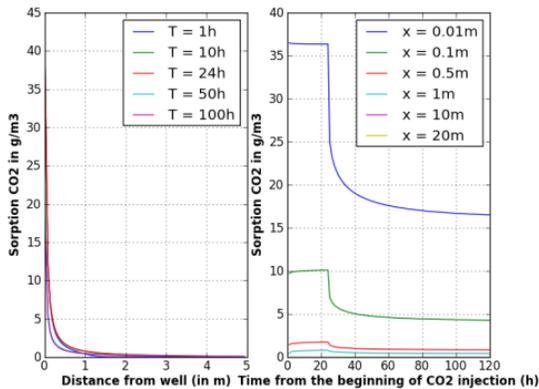


Figure 7. Evolution of CO₂ adsorbed, function of space (on left) and of time (on right)

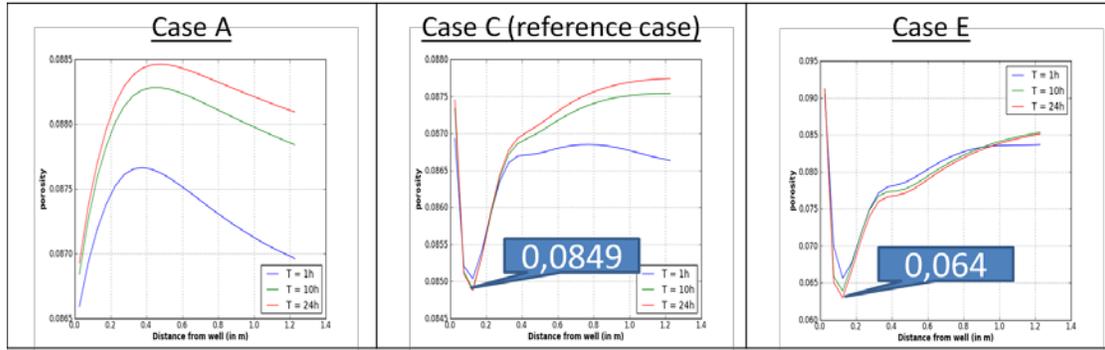


Figure 9. Sensitivity of the porosity to gas volumetric strain (cases A, C and E)

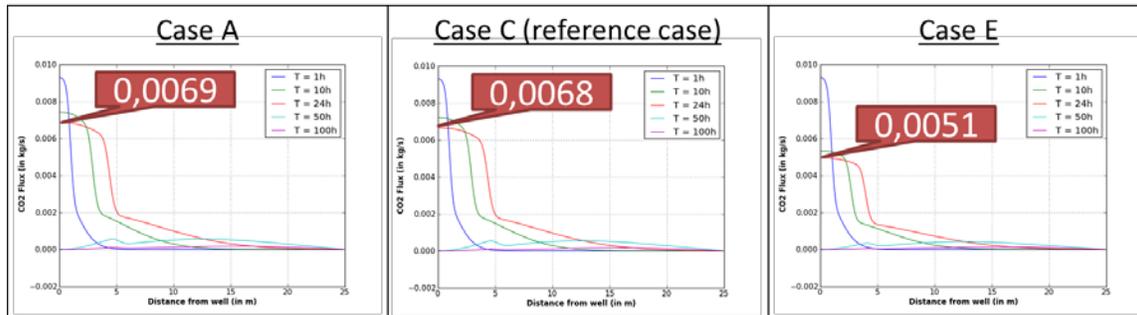


Figure 10. Sensitivity of the CO₂ flux to gas volumetric strain (cases A, C and E)

CONCLUSION

The first stage of numerical developments is achieved and has provided a thermo-hydro-mechanical simulator taking into account the adsorption process, mechanical processes like swelling/shrinkage, and stress/sorption dependent porosity/permeability effects.

Experimental surveys should be launched in next months, and will enable to validate and improve the models by comparison between experimental measures and simulations of experiments. The simulator will also be confronted with the COMSOL-based tool developed by INERIS.

The results of simulations presented in this paper aim at testing the supervisor and should be considered cautiously. Nevertheless, these simulations highlight the utility of such a simulator. It could be used to understand better the very complex behavior of coals. The detailed impact of different parameter perturbations, both individually and simultaneously, in this highly

coupled problem will be the subject of further research. The simulator could also be used for monitoring design, for injection scenario studies and for future project sizing.

ACKNOWLEDGMENT

CARBOLAB is a project supported by the European Fund for Coal and Steel involving HUNOSA, owner of the mine, AITEMIN from Spain, BRGM, INERIS and TOTAL from France, GIG from Poland.

REFERENCES

- Chen, Z., J. Liu, D. Elsworth, L.D. Connell, Z. Pan, Impact of CO₂ injection and differential deformation on CO₂ injectivity under in-situ stress conditions, *International Journal of Coal Geology*, 81 97–108, 2010.
- Defossez, P., *Caractérisations de charbons en vue de leur utilisation pour le stockage géologique de CO₂*, (in French), PhD thesis, May 2011

- EDF, Analysis of Structures and Thermomechanics for Studies & Research, www.code-aster.org.
- Liu, J., Z. Chen, D. Elsworth, X. Miao, X. Mao, Linking gas-sorption induced changes in coal permeability to directional strains through a modulus reduction ratio, *International Journal of Coal Geology*, 83, 21–30, 2010.
- Oldenburg, C.M., G.J. Moridis, N. Spycher, and K. Pruess, *EOS7C Version 1.0: TOUGH2 Module for Carbon Dioxide or Nitrogen in Natural Gas (Methane) Reservoirs*, LBNL Report LBNL-56589, March 2004.
- Settari, A. and F.M. Mourits, A coupled reservoir and geomechanical simulation system, *SPE Journal*, 219–226, 1998.
- Shi, J.Q., S. Durucan, Drawdown induced changes in permeability of coalbeds: a new interpretation of the reservoir response to primary recovery. *Transport in porous media*, 56, 1–16, 2004.
- Shi, J.Q., S. Durucan, A bidisperse pore diffusion model for methane displacement desorption in coal by CO₂ injection, *Fuel*, 82, 1219–1229, 2003.
- Xu, T., and K. Pruess, Modeling multiphase non-isothermal fluid flow and reactive geochemical transport in variably saturated fractured rocks: 1. Methodology, *American Journal of Science*, 301, 16-33, 2001.