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CO₂ Injectivity in Geological Storages: an Overview of Program and Results of the GeoCarbone-Injectivity Project

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Résumé — Injectivité du CO₂ dans les stockages géologiques : programme et principaux résultats du projet ANR GéoCarbone-Injectivité — L'objectif du projet GéoCarbone-Injectivité était de définir une méthodologie pour étudier les phénomènes complexes intervenant aux abords des puits lors de l'injection de CO₂. La méthodologie proposée s'appuie sur des expérimentations interprétées numériquement à l'échelle de la carotte afin de comprendre (modélisation physique et lois de comportement) et de quantifier (paramétrisation des outils de simulation) les différents mécanismes susceptibles de modifier l'injectivité : les interactions roche/fluide, les mécanismes de transport aux abords du puits d'injection et les effets géomécaniques. Ces mécanismes et les paramètres associés devront ensuite être intégrés dans une modélisation à l'échelle métrique à décimétrique des abords du puits d'injection. Cette approche a été appliquée pour l'étude d'une injection potentielle de CO₂ dans la formation géologique du Dogger du Bassin Parisien, en relation avec les projets ANR GéoCarbone.

Abstract — CO₂ Injectivity in Geological Storages: an Overview of Program and Results of the GeoCarbone-Injectivity Project — The objective of the GeoCarbone-Injectivity project was to develop a methodology to study the complex phenomena involved in the near wellbore region during CO₂ injection. This paper presents an overview of the program and results of the project, and some further necessary developments. The proposed methodology is based on experiments and simulations at the core scale, in order to understand (physical modelling and definition of constitutive laws) and quantify (calibration of simulation tools) the mechanisms involved in injectivity variations: fluid/rock interactions, transport mechanisms, geomechanical effects. These mechanisms and the associated parameters have then to be integrated in the models at the wellbore scale. The methodology has been applied for the study of a potential injection of CO₂ in the Dogger geological formation of the Paris Basin, in collaboration with the other ANR GeoCarbone projects.

INTRODUCTION

During the lifetime of a CO₂ geological storage operation, large volumes of CO₂ will have to be injected through a minimum number of wells, for economical reasons but also to minimise leakage risks. The design of a storage will thus require the control of the injectivity: which are the phenomena and the parameters governing the injectivity, how do they vary during the injection, and how to modify them in case of an injectivity loss?

The main goal of the “GeoCarbone-Injectivity” project was to develop a methodology to study these complex phenomena involved in the near wellbore region, and to apply it to the context of the Paris Basin which is studied for site screening and characterization within the framework of the companion project “GeoCarbone-PICOREF”.

The proposed methodology is based on experiments and simulations at the core scale, in order to understand through physical modelling and determination of constitutive laws, and to quantify (calibration of simulation tools) the mechanisms involved in injectivity variations. Figure 1 presents the main mechanisms which can potentially impact the CO₂ injectivity within a CO₂ geological storage operation. Three main classes of phenomena can be distinguished and justify the project organization:

- fluid/rock interactions (Phase 1 of the project),
- transport mechanisms (Phase 2),
- geomechanical effects (Phase 3).

These mechanisms and the associated parameters have then to be integrated in the models at the wellbore scale (Phase 4).

The first three steps of the methodology were applied on samples representative of the Dogger formation of Paris Basin site: Lavoux limestone and/or Charmottes carbonate plugs, and in the different laboratories of the project partners. In-house and commercial softwares were used for the interpretation of these experiments and for simulations of CO₂ injection at the wellbore scale.

1 MOST OUTSTANDING RESULTS

1.1 Rock/Fluid Interactions

In the first phase of the project, the influence of geochemical reactions on CO₂ injectivity was studied through batch and percolation experiments. The interpretations were performed through numerical simulations using models taking into account both transport and geochemical phenomena (Coores™ and ToughReact simulators).

Experiments in batch reactors have been conducted on plugs of Lavoux oolitic limestones in presence of CO₂ + brine, N₂ + brine and supercritical CO₂, without thermal gradient at 80°C and at 150 bar bulk pressure. Weak dissolution

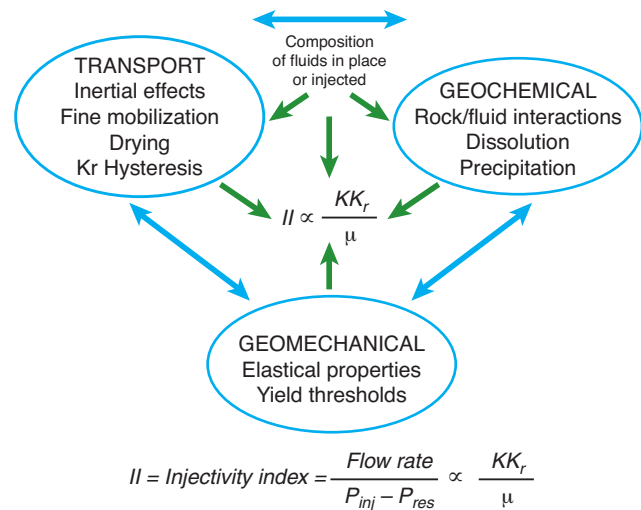


Figure 1

Mechanisms involved in injectivity evolution. Injectivity Index II depends on flow rate and pressures. K : absolute permeability, K_r : relative permeability, μ : viscosity of the injected fluid.

and/or precipitation phenomena have been detected. Analysis of water chemistry after experiments shows that less than 1% of limestone is dissolved. On the contrary, important mass transfers have been observed in the case of experiments in grain columns (COTAGES device) with thermal gradient (35 to 100°C on 70 cm). Dissolution occurred in the coldest part of the reactor whereas clear sealing calcite precipitated in the hottest part (Fig. 2). This mechanism is accompanied by strong pressure variations with time marking dissolution of calcite during the first 10 days and precipitation after. This behaviour has been qualitatively reproduced by numerical simulations (Sterpenich *et al.*, 2009; André *et al.*, 2009).

Whereas batch experiments with supercritical CO₂ showed only a slight modification of the pore structure due to calcite dissolution, the dissolution/precipitation phenomenon was more strongly highlighted during brine/CO₂ displacement experiments. The results of the percolation experiments performed in Lavoux and Charmottes samples clearly show the influence of the flow rate (effect of the Peclet and Damköhler parameters) on the dissolution patterns observed by NMR and CT-scanner: dissolution with wormhole formation for high flow rates and compact dissolution for low flow rates. These observations gathered in Table 1 are in good agreement with the Peclet/Damköhler diagram of the literature (Fig. 3) established for acid flows (Bazin, 2001; Egermann *et al.*, 2005; Lombard *et al.*, 2007). In most of the cases, the evolution of pressure and effluent composition during the percolation experiments as well as final porosity and permeability, were reproduced qualitatively by simulation (Coores™).

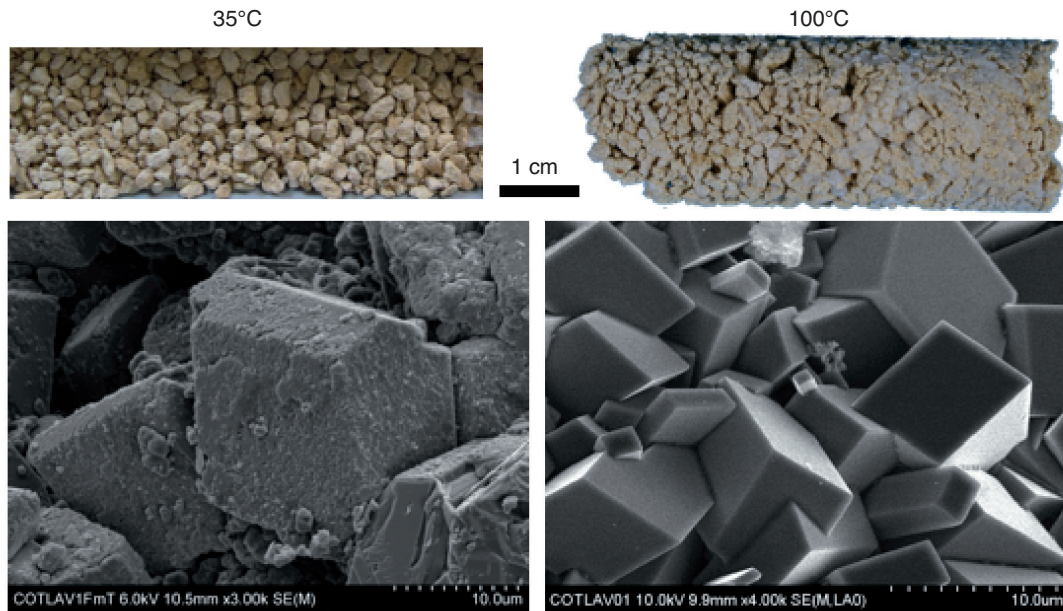


Figure 2

Effect of the thermal gradient on the mass transfer (CaCO₃) for the COTAGES experiment. Left: calcite dissolution at the cold side, right: calcite precipitation at the hot side.

Nevertheless, it was observed that particle displacements may occur and modify the pressure profiles which were not reproducible by simulation. At the present time, this impact is not taken into account by the simulators (Bazin *et al.*, 1994; Bazin, 2001) and this still represents a research challenge.

1.2 Transport Properties

Without considering the pore structure alteration due to the acidified brine, transport parameters such as capillary pressure and relative permeability are directly linked to interfacial properties such as interfacial tension (IFT) and wettability.

TABLE 1

Percolation experiments in Lavoux and Charmottes plugs – $T = 80^{\circ}\text{C}$, $P = 180$ bar

Field	Sample	Percolation exp. $Q_w + Q_{\text{CO}_2}$ (cc/hr)	Initial porosity	Final porosity	Initial K_w (mD)	Final K_w (mD)	Ca ⁺⁺	SO ₄ ⁻	Comments
Lavoux	LJ001	10	18	17.1	155	154.5 –	No variation	↓ and ↑	Compact dissolution ϕ ↑ at the inlet and ↓ slightly at the outlet (as K)
Lavoux	LJ004	40	20.6	20.5	174.8	157.6 ↓	↑	↓ and ↑	Dissolution (inlet + preferential paths) + precipitation (outlet)
Lavoux	LJ003	200	20.8	19.4	89.7	120.9 ↑	↑ and ↓	Small variations	Wormholes (cf. NMR T2 profiles): dissolution > precipitation due to HFR
Charmotte	4-1-aH	4	12.3	12.8	0.47	0.02 ↓	↑ slightly	↓ and ↑ and stable	Compact dissolution at the inlet + uniform porosity
Charmotte	4-2-cH	30	14.4	15.2	2,3	0.95 ↓	↑ and stable	↓ and ↑ and stable	Small wormholes
Charmotte	4-1-bV	140	15.3	16	20.7	21.1 ↑	stable ↑ and ↓	stable	Wormholes and dissolution (uniform porosity increase)

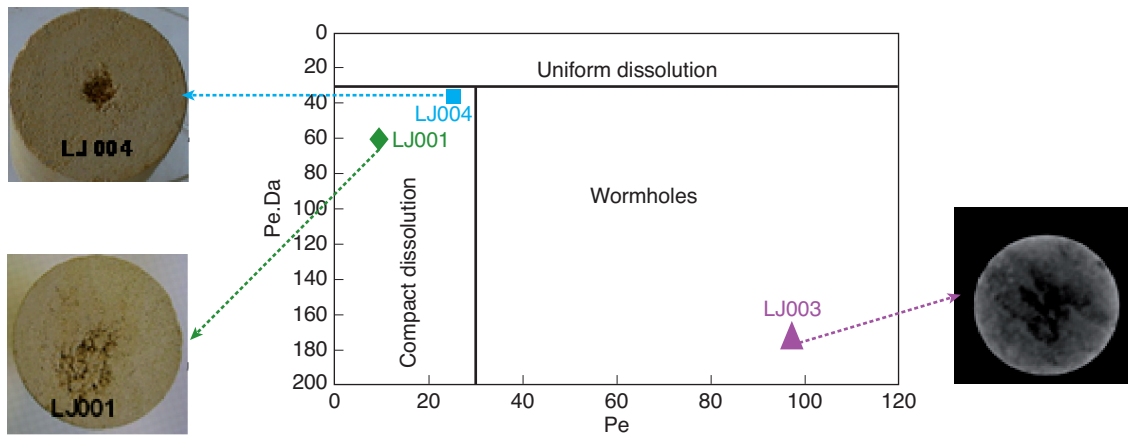


Figure 3

Reactive percolations on Lavoux limestone samples – PeDa diagram.

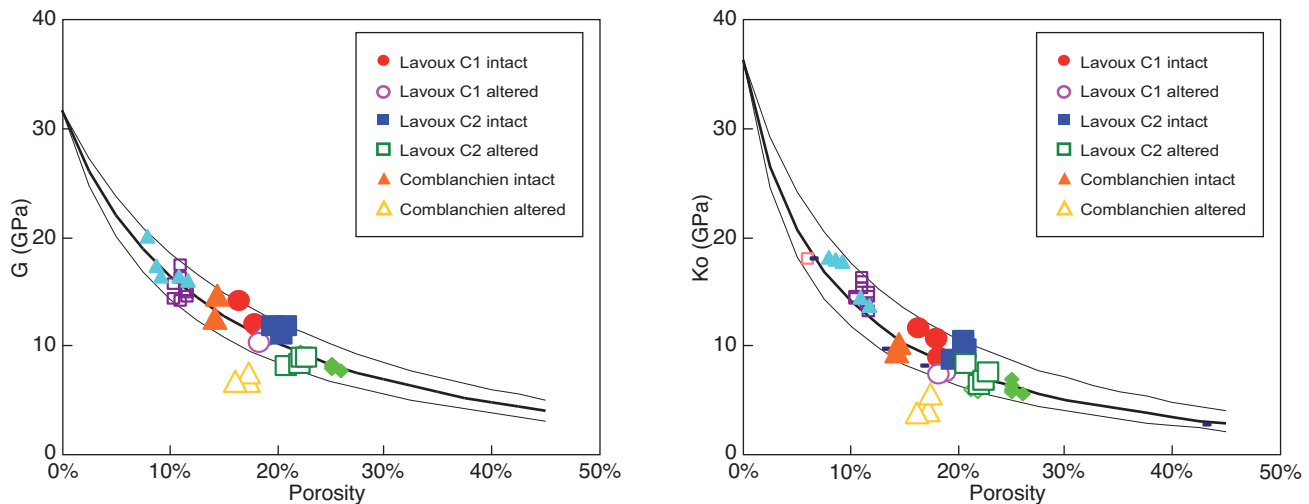


Figure 4

Natural trends for carbonates drained elastic moduli (Bemer *et al.*, 2009).

Chalbaud *et al.* (2009) (in this volume), present an experimental study, performed in parallel to the Injectivity project, which allowed new brine/CO₂ IFT data necessary for CO₂ storage issues to be acquired, and propose a correlation with thermodynamical conditions. Observations at the pore and core scales have also shown the possible wetting behaviour of CO₂ and the authors propose a discussion on its influence on the storage capacity of the investigated formations.

Within the Injectivity project, the evolution of transport properties (single phase and two phase flows) generated by the pore structure alteration of the rock near the well was studied through the characterization of both native samples

representative of the Dogger formation (Lavoux limestone) and samples altered uniformly by an acid solution (Egermann *et al.*, 2006). The acidity has implied a slight increase of permeability and porosity and a decrease of inertial coefficients. Both relative permeabilities and Klinkenberg effects remained unchanged. When no particle displacement takes place, the injectivity of the studied rock seems not to be altered by the acid solution (Radilla *et al.*, 2010, in this volume).

In the first phase of the project it has been shown that the structural modification patterns and the porosity/permeability variations with the reactive and flow regimes can be interpreted with Peclet and Damkhöler numbers. In parallel to this

work, Algive *et al.* (2009) used these dimensionless numbers and a Pore Network Modelling approach to study the impact of such a reactive flow on transport parameters. For different flow regimes (Peclet and Damköhler numbers) they calculated the porosity/permeability evolution due to dissolution/precipitation phenomena, and the modified two phase flow parameters: capillary pressure and relative permeability.

1.3 Geomechanical Parameters

The consequences of the injection of an acid solution on geomechanical parameters of Lavoux samples were investigated. During percolation experiments of a CO₂ saturated brine, the same dissolution patterns were observed as in the first phase, with an axial deformation of the samples. Water+CO₂ percolation experiments have shown an increase of limestone dissolution with the increase of the fluid flow. Dissolution is accompanied by an increase of the size of the “wormholes”. CO₂-saturated water, chemically equilibrated with the limestones in room conditions, dissolves calcite until the solution becomes equilibrated at *P*, *T* conditions of the experiment. Mechanical behaviour is not affected by the percolation of dry CO₂ (gaseous or supercritical). Replacement of liquid water by CO₂ at the same pressure does not induce any deformation of the samples (Grgic *et al.*, 2010).

The poroelastic properties as well as the rupture criteria (induced fracturing) have been measured on native and altered Lavoux and Comblanchien Charmottes carbonate samples. The homogeneous alteration procedure with an acid solution was the same as in Phase 2 (Egermann *et al.*, 2006). Clear trends of chemically induced mechanical weakening have been outlined (Fig. 4). The experimental data have been compared to the data obtained by phenomenological and empirical models (Bemer *et al.*, 2009, in the previous OGST volume). Nevertheless additional experimental data are needed to further investigate the effects of chemical alteration on carbonates geomechanical properties and to integrate them in reactive transport simulators.

1.4 Coupling Phenomena

The information on the coupling of Thermo-Hydro-Mechanical-Chemical (THMC) phenomena was partially integrated in the simulators used by the partners (ToughReact, Coores™) within Phase 4 of the project. The effects of several types of coupling (HC, TH, THC, THM) were also tested (ToughReact) in the context of various scenarios of CO₂ injection in a generic aquifer representative of Paris Basin. 1D and 2D radial models were used. The petrophysical properties of the Lavoux limestone were considered. The initial

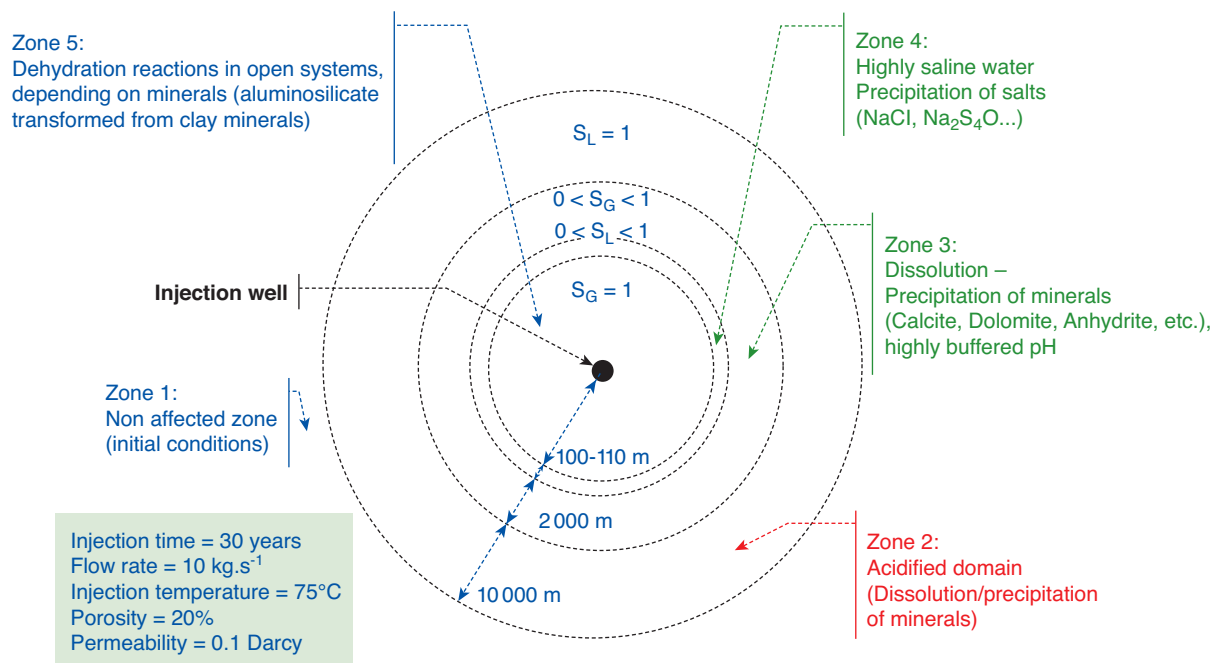


Figure 5

Structured zones and physical (evaporation, overpressure, cooling and relative saturation) and reactive chemical (pH, mineral dissolution/precipitation/transformation) phenomena within different zones around the CO₂ injection well (schematic concept based on the integrated results on experiments and numerical simulations) (Azaroual *et al.*, 2007).

pressure and temperature of the formation were 180 bar and 75°C.

In terms of Pressure-Rate-Time relationship, the modifications of CO₂ injection flow rate (temperature $T = 75^{\circ}\text{C}$) led to only slight variations of the (stable) final pressure (Tab. 2). For a CO₂ injection at a constant flow rate of $Q = 5 \text{ kg/s}$ ($T = 50^{\circ}\text{C}$), the Thermo-Hydro-Mechanical coupling (THM) also does not show significant overpressures (André *et al.*, 2007).

TABLE 2

Final pressure vs flow rate during the injection of CO₂ in the generic Dogger aquifer. $P_{\text{initial}} = 180 \text{ bar}$, $T_{\text{initial}} = 75^{\circ}\text{C}$.
Final pressures do not vary significantly with time

Flow rate (kg/s)	Final pressure (bar)
10	213
20	242
30	268

Finally Figure 5 summarizes the main phenomena occurring in the different zones around the injection well and partially highlighted within this study. The phenomena of drying and dehydration of the near wellbore region were not studied within the Injectivity project. The observed displacement of fines is also not yet taken into account in the simulators and require further research work.

CONCLUSIONS AND PERSPECTIVES

This Injectivity project objective was to develop a methodology to study the complex phenomena involved in the near wellbore region during CO₂ injection. It allowed new experimental tools to be developed and first sets of geochemical, petrophysical and mechanical experimental data to be acquired before and after CO₂ injection in the Dogger Paris Basin formation:

- the dissolution/precipitation phenomena linked to the acidity of the brine/CO₂ solution (not equilibrated with the rock) have been outlined and can be studied through dimensionless numbers. In presence of water, equilibrated with the rock and saturated with CO₂, dissolution rates and pore rearrangements are weak, at the detection limit of the analytical techniques;
- the measured interfacial and transport parameters are presented in Chalbaud *et al.* (2009) and in Radilla *et al.* (2010). When no particle displacement takes place, the injectivity of the studied formation is not altered by the acid solution. A Pore Network Modelling Approach should allow a better prediction of the evolution of pore structure and transport parameters during a CO₂ injection;
- clear trends of chemically induced mechanical weakening have been outlined on altered Lavoux and Charmottes samples (Bemer *et al.*, 2009);

- within the simulation of various scenarios of CO₂ injection in a generic aquifer representative of Paris Basin, several types of coupling (HC, TH, THC, THM) were tested. For this aquifer, large overpressures were never obtained throughout the simulations.

In order to improve the interpretation models and for a better implementation in the reactive transport simulators, the experimental data would need to be completed.

The influence of particle displacements which has been observed is not yet well taken into account and should be studied within further research works. Furthermore, drying and dehydration are two other important phenomena occurring near the injection well which were not considered in this project. They are studied within the “Near Well” ANR project (2008-2010). The influence of impurities in CO₂ on geochemical and dissolution/precipitation mechanisms also has to be better understood. This is one of the goals of the “Annex Gas” ANR project (2007-2010).

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