Evaluating sealing efficiency of caprocks for CO2 storage: an overview of the Geocarbone Integrity program and results

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1. **Résumé —** Un aperçu du programme et des résultats du projet multipartenaire Geocarbone-Intégrité est donné. Il concerne le développement de méthodes expérimentales et numériques pour évaluer l'intégrité d'un stockage de CO_2. Les critères essentiels d'une couverture sont l'épaisseur de la formation et sa perméabilité. Une migration locale et limitée du CO_2 dans la couverture due à une pression capillaire d'entrée insuffisante est étudiée dans ce travail. A grande échelle, des profils sismiques sont nécessaires pour caractériser la continuité d'une couverture. Quand on dispose de données de puits, des critères simples pour estimer l'argilosité peuvent être utilisés. On montre également que les techniques de lithosismique peuvent être appliquées aux couvertures. Pour les formations considérées, nous n'avons pas observé au laboratoire de réactivité géochimique importante, ni d'effet marquant sur les propriétés mécaniques. Des simulations hydromécaniques à grande échelle montrent que les critères de rupture ou de réactivation de fractures préexistantes ne sont pas satisfaits. Des simulations de transport réactif par diffusion et écoulement diphasique dans la couverture montrent une migration du CO2 sur une dizaine de mètres au plus et une baisse de la porosité par précipitation, et localement une augmentation de la porosité par dissolution.
2. ***Abstract* —**An overview of the three year program and results of the Geocarbone-Integrity French project is given. It focused on the development of experimental and numerical methodologies to assess the integrity of an underground CO_2 storage at various scales. The primary criteria in the selection of a caprock formation for CO_2 storage purpose is the thickness and permeability of the formation. Local and limited migration of CO_2 into the caprock due to insufficient capillary entry pressure has been studied as a probable scenario. At large scale, caprock characterization requires at least seismic profiles to identify lateral continuity. When well logging data are available, simple rules based on clay content can be used to estimate thicknesses. For the formation considered, the geochemical reactivity to CO_2 was small, making reaction path difficult to identify. Similarly, artificial alterations of samples representing extreme situations had little impact on geomechanical properties. Finally, with realistic over pressure due to injection, shear fracture reactivation criteria are not reached and migration of CO_2 either by diffusion or by two phase flow within the first meters of the caprock produce mostly a decrease of porosity by precipitation, and very locally an increase of porosity by dissolution.

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INTRODUCTION

Carbone Capture and Sequestration (CCS) is one of the many solutions to limit the current global warming [1]. In 2005, the newly formed French funding agency ANR decided to launch mixed academia-industry research program focusing on CCS. One of these programs is dedicated to the study of the sealing efficiency of formations above the storage (caprocks), called Geocarbone Integrity (ANR-05-CO2-006), bringing together 10 French partners[1] with very various technical knowledge. The main objective of the program (1/2006 to 12/2008) is the development of experimental and numerical methodologies to assess the integrity of an underground CO_2 storage at various scales. These methodologies are illustrated using samples and data from geological formations of the Paris basin, in conjunction and coordination with other programs such as GeoCarbone-Picoref and GeoCarbone Injectivity also presented in this issue.

Caprocks are essentially defined as low (μD, 10^{-18} m^2) or very low (nD, 10^{-21} m^2) permeability formations, and sometimes but not necessarily with low porosity (<15%). Caprocks are generally viewed as hermetic layers above the storage in which no CO_2 should migrate. However, there is some evidence from natural gas fields [2, 3] containing CO_2 that a migration occur over geological time scales without significant impact. The approach taken here is to study such a possibility and hence the problem is rather to estimate how slow and how far will be the migration of CO_2 into the caprock formations, and to study various scenarii in which different transport property values are taken. Hence, the usual term "leak" should be clarified when considering a limited migration in a caprock. Also, if a caprock formation is partially invaded by CO_2, it may contribute to a faster decrease of the overpressure in the short term, and to the storage capacity in the long term in a non negligible way [4].

Different mechanisms for CO_2 migration are possible, from small to large scales:
- molecular diffusion of dissolved CO_2 in the pore water from the reservoir zone to the caprock formation,
- CO_2 diphasic flow after capillary breakthrough,
- CO_2 flow through existing open fractures.

The following mechanisms can accelerate or slow down the migration:
- Chemical alteration of the mineralogical assemblage of the caprock formation under the influence of acid water,
- Re-opening of pre-existing fractures or micro cracks induced by overpressure of the reservoir below,
- A combination of the above (chemical alteration of the mineral filling the fractures).

It can be immediately seen that a broad range of discipline and scales are interacting each other: large and small scale geological characterization, petrophysical and geomechanical characterization at the plug scale, geochemical processes at the grain scale. In terms of numerical simulations, one need to couple multiphase transport, geochemical and geomechanical effects in porous media, a challenging task.

From the petroleum perspective, a broad range of technique is available for characterizing oil and gas reservoir, from seismic to logging and laboratory techniques. However, caprocks are usually (and obviously) studied neither by oil and gas companies nor by hydrologist. In addition, coring is not performed, giving little access to petrophysical or petrographical description. Low permeability formation acting as barriers have been studied in details for nuclear waste storage purpose, and in this case, the scientific issues are very similar [5, 6] although the time scales are larger for nuclear waste problem and requires a perfect confinement. However, linking these two problems may be confusing in terms of communication and public acceptance. Finally, caprocks have also been studied in details for gas storage purpose. Unfortunately, data are not published and kept confidential within the operating companies. Note that a major difference between CH_4 and CO_2 storage is a decrease by a factor of two of the capillary entry pressure; indeed the brine/CO_2 interfacial tension is much smaller than brine/CH_4 at high pressure because of the larger affinity of CO_2 to water (about 30 mN/m compared to 58 mN/m at moderate temperature).

Most of the techniques and methodologies used for studying hydrocarbon reservoirs or aquifers can be used for caprocks. At large scales, seismic data, logging data and well to well correlations techniques can give an indication of the lateral continuity and thickness of the caprock formation. Major faults can be detected using seismic data, while small scale fractures can be identified using well bore imaging techniques. For petrographical and petrophysical properties, advanced techniques are necessary. For example, low permeability measurements are difficult and time consuming to perform using standard steady techniques and others techniques must be used [7]. Similarly, standard thin sections for petrographical observations give limited information and Scanning Electron Microscopy (SEM) must be used. For the chemical reactivity and geomechanical properties, standard techniques can be used but over extended period of time.

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The project was organized in 6 work packages comprising:
- a geological description at a regional scale focusing on the St Martin de Bossenay depleted oil field, and a detailed petrographical analysis of caprock samples [8],
- a petrophysical characterization caprock samples including diffusion measurements [7, 9], and a study of the contact angle to evidence wettability changes [10, 11],
- the measurements of geomechanical properties before and after alteration by CO_2 [12], and a methodology to predict the onset of a fracture network preexisting in the caprocks formation and their evolution during CO_2 storage: the variation of mechanical properties are measured before and after CO_2 percolation through the sample.
- the study of the reactivity of three caprock formations [13, 14, 15]
- large scale hydromechanical [16] and reactive transport simulations [17].

Finally, the last work package focused on the possibility of cement well failure. It will not be described in this synthesis.

1 LARGE SCALE DESCRIPTION

At large scales, we are interested by the variations of thickness of the caprock formation, and the lateral changes of facies. This information can be provided by logging data and seismic and lithoseismic information. In the present project, we focus on the Saint Martin de Bossenay (SMB) field as an example of how such information can be gathered from existing data. In particular, we give the example of the caprock above the Dogger formation (see reference 18 for a description of the geological setting). This formation has a storage capacity estimated to 4 Gt.

For clay-rich formations, a key logging data is the gamma ray (GR), a well know indicator of clays. Since this data has been and is still acquired systematically, it is largely available even in very old wells. From the GR data, a useful clay indicator is defined as:

$$GR_{rel} = \frac{GR - GR_{\min}}{GR_{\max} - GR_{\min}} \tag{1}$$

This allows having comparable data from wells where different tools or tool generations have been used. After the identification of the formation on the well logs, a mean of the clay indicator of the formation can be calculated at each well. Then, a 3D interpolation is performed. The results are indicated in Figure 1 in the case of the SMB field. In the case presented here, the trapping efficiency of the caprock formation has not to be proven because a hydrocarbon reservoir is present. For the zone considered, raw Gamma-ray data show good clay qualities for the Callovo-Oxfordian formation just above the Dogger aquifer. When analyzing the relative indicator, best clays are found in the North-West. However, it is obvious that such an interpolation between distant wells may not detect sudden changes or fractures. For this purpose, seismic profiles originally designed to reveal hydrocarbon reservoir were reanalyzed to focus on the caprock. In addition, for a few wells on the NS02 line (Figure 1), P-wave sonic and density logs were available, making possible a lithoseismic interpretation, i.e. a translation of seismic impedance into facies. It was found that the classical lithoseismic interpretation usually performed for reservoir was also applicable to caprock in this case. In general, the results show that lateral changes observed continuously with seismic information at a reasonably high resolution were very smooth and that sparse local characterizations available at each well could reasonably be interpolated, giving a reliable map of thickness as shown in Figure 1.

Finally, hydraulic and geochemical arguments can also be used to evidence the lack of exchange across a caprock. For example, in the framework of nuclear waste problems, water movements have been studied in the Bure Area [5]. The Callovo-Oxfordian caprock formation is sandwiched between the Oxfordian and Dogger formation. In these two formations, hydraulic charges, isotopic water composition and flow direction are very different and provide convincing arguments in favor of the efficiency of the Callovo-Oxfordian caprock to limit vertical exchanges over geological time scales [8].

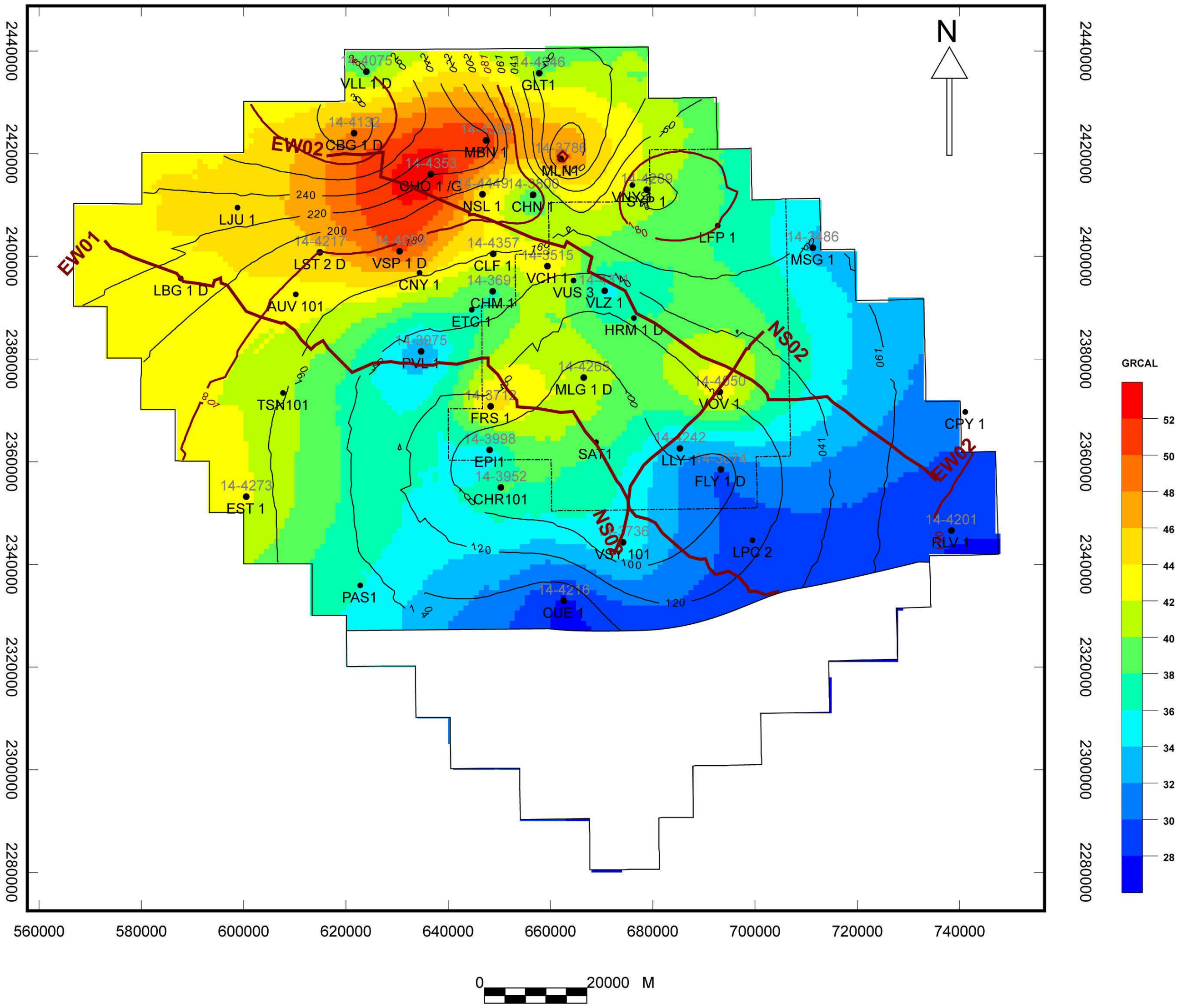

Figure 1 : Result of the interpolation between wells to determine the quality of the Callovo-Oxfordian clayey formation above the Dogger formation (from reference 8). The right scale is calibrated between 0 = GR_{min} and 100 = GR_{max}. The thick lines indicate the available seismic profiles, while thin lines indicate the formation thickness. The SMB field is located approximately at the crossing of NS02 and EW02 seismic lines. The inset indicates the Picoref area. The lack of available wells doesn't allow a reliable interpolation in the south part of the picture.

2 SMALL SCALE DESCRIPTION: PETROGRAPHY AND PETROPHYSICAL PROPERTIES

Petrography and mineralogy:

The knowledge on mineralogy is an important aspect. For quantitative analysis and visualizations, various methods can be used such as X-ray diffraction (XRD), Scanning Electron Microscopy (SEM), Transmission Electron Microscopy (TEM), etc. Standard thin section visualizations are not very useful in general because typical length scales in caprocks are below 0.1 µm. An example of such analysis is given in Figure 2 (left). The caprock of the SMB area contains a significant amount of carbonates (about 50%) beside clays, and therefore, the microstructure is quite complex. Interestingly, the mineralogical composition does not vary very much neither vertically nor horizontally (the sample from the Bure area is from the same Callovo-Oxfordian formation but distant by about 100 km from the SMB area). Hence, reactivity tests performed on samples from the Bure area are also representative of the SMB area [14].

Another study has been performed on tight carbonates present below the Callovo-Oxfordian formation (Comblanchien, Figure 2, right) with permeabilities around 1 µD. Strictly speaking it cannot be considered as a caprock (aquiclude) but rather as an aquitard where the vertical transfer are very slow. In this case, a dolomitization process is clearly observed, therefore increasing the porosity and potentially the permeability. Dolomitization being linked to the circulation of magnesium rich water during the geological history of the formation, it produces non uniform spatial characteristics and is an important aspect to study in such formations.

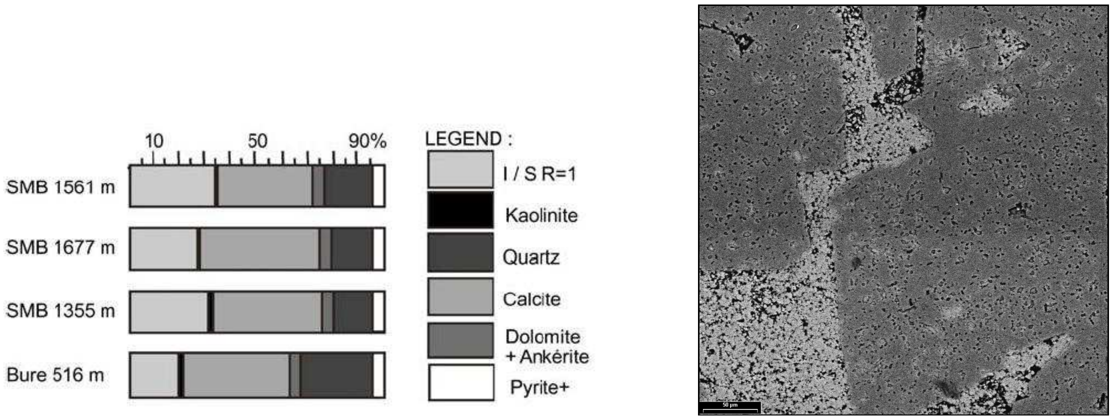

Figure 2: Example of analysis of two caprock faciès. Left: main mineral content of Callovo-Oxfordian argilites from the SMB and Bure area [14]. Right: SEM vizualization of a dolomitic tight carbonate caprock, Comblanchien faciès [8]. The thick black line indicates a scale of 50 µm.

Petrophysical properties: permeability, capillary entry pressure

Permeability is the most important parameter for characterizing a caprock. The measurement of permeability in the range of µD down to nD is not only time consuming using standard techniques (steady state techniques) but can also depend on the type of fluid used and is strongly dependent on the degree of preservation of samples, especially when the clay content is high. A study of different methods for measuring permeability on the same samples [7, in this issue] indicates that the permeability deduced from the measurements can vary by one order of magnitude. These experiments were performed on low permeability carbonate samples, well adapted to multiple tests with water and gas. An order of magnitude in permeability measurements is not critical because the natural variability of caprock formation may be much larger.

The capillary entry pressure or threshold is the minimum pressure difference between gas and water necessary for the gas to enter the porous media. In the laboratory, several methods are available: mercury injection capillary pressure (MICP), a standard characterization technique needing a complete drying of the sample, and direct flow tests using either nitrogen or CO_2. Previous studies [19, 20] indicate some issues when comparing these methods: sensitivity to anisotropy, connectivity of the largest pores in the sample. In addition a change of wettability between N_2, CO_2 and CH_4 vs brine was also suspected. This last issue is discussed later.

An example of the variability of permeability and entry pressure is given in Table 1 (adapted from reference 7). At different depths of the Comblanchien formation above the reservoir in the SMB field, we can find low porosity and small to medium permeability values (0.01 up 10 µD). However, from mercury injection experiments and after interfacial tension corrections, the capillary entry pressure can be quite low suggesting that a migration is likely to occur in some part of this formation which should be viewed as a transition between the storage itself and the clay-rich formation above. The entry pressure values reported here are also comparable to values measured in the case of clay-rich marlstones samples with permeability in the range of 30 nD [21]. Hence, in the simulation presented later, we took a permeability of 1 µD and low capillary entry pressure to study the migration and reactivity of CO_2. In general in a field study, it may be difficult to obtain high enough entry pressure on all tested samples and an experimental proof of the sealing efficiency will not be available. Therefore, the migration of CO_2 into the caprock must be considered as a probable scenario.

Table 1: Example of the variability of permeability and brine/CO_2 capillary entry pressure of a low permeability zone above the Dogger formation (adapted from reference 7).

Sample	Depth m	Permeability microD (10^{-18} m^2)	Porosity %	Entry pressure (bar)
106-5-1 eH	1911.39	0.1 - 6	2.8 /4.0	-
106-5-2 cV	1910.95		2.0 / 2.6	***22***
106-5-2 dV	1910.5		2.5 / 2.9	**16**
107-1-2 cV	1958.87	0.06 - 2	3.8	***12***
107-2-1 aV	1959.12		3.8 / 5.8	**5**
109-1-2 aH	2000.0	0.03 - 12	3.3 / 7.6	***9***
109-2-1 aV	2006.73		7.6 / 8.8	**-**
109-2-1 bV	2006.56		5.9	***0.8***
109-2-1 eV	2006.46		7.6	**0.4**
109-2-2 aH	2006.28		3.3	***10***

Diffusion properties:

As mentioned above, a possible transport mechanism is molecular diffusion of dissolved CO_2 in the pore water. Molecular diffusion is a slow transport process at the time scale of a storage site while injecting. However, due to the rapid migration of CO_2 at the base of the caprock acting as a permeability barrier, long term diffusion transport should be evaluated carefully. Diffusion and permeability are distinct properties that do not depend upon the same petrophysical properties. As an illustration, if we take a simple system of grain packs of uniform grain size, permeability depends typically on the square of the grain size whereas porosity and diffusion will be constant. Therefore, despite the very low permeability of caprocks, diffusion coefficients can be of the same order of magnitude as in the reservoir zone and only reduced by one or two orders of magnitude compared to bulk values (i.e. unconfined fluid). In contrast, caprocks have permeabilities reduced by six orders of magnitude or more compared to the permeability in the reservoir zone. This is due to the fact that caprocks are made of fine grains such as clay particles, and/or they have been subjected to specific diagenetic processes such as dissolution/ recristallization and cementation.

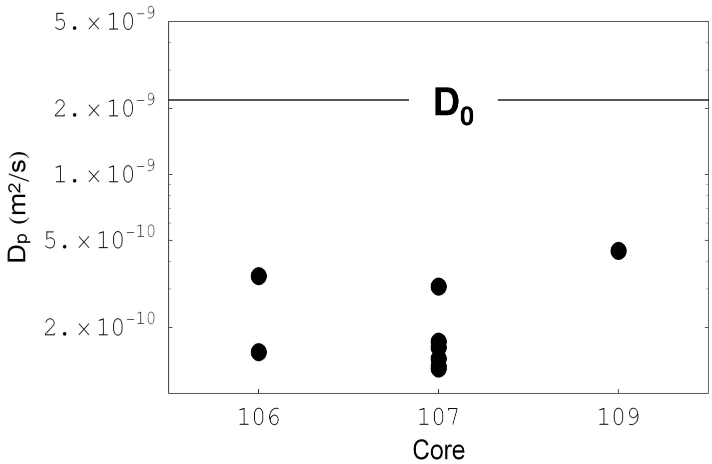

Figure 3: Pore water diffusivity measurements on three samples from the SMB field, transition zone, representing an upper limit for the diffusion of dissolved CO_2, from reference 9. Porosity 6 %, permeability around 1 µD.

A summary of the results from reference 9 is given in Figure 3. The diffusion of dissolved CO_2 is close to but not larger than the self-diffusion of water in the porous medium and we take these measurements as an upper limit. Based on the analogy between electrical and diffusion properties, and using the 1st Archie law, a relationship between the pore diffusion D_p and porosity is suggested, as well as the temperature dependence:

$$\frac{D_p}{D} = \varepsilon^{m-1} \text{ with m≈2 and } D = D_0\left[\frac{T}{T_S}-1\right]^{\gamma} \text{ with } D_0 = 1.635\ 10^{-8} m^2/s \quad T_S = 215.05\ °K\ ,\ \gamma = 2.063 \qquad (2)$$

where D is the self diffusion of bulk water. At a porosity ε of 5 and 15 %, diffusivity is typically reduced by a factor of 20 and 7 respectively.

Concerning the diffusion of charged species, the above relationships do not apply. When measuring the diffusion of bicarbonate ions (HCO_3^-) in the same porous media as shown in Figure 3, an additional reduction of one order of magnitude is observed [9 and references therein]. Hence, two diffusion fronts are present: the diffusion of dissolved CO_2 and the diffusion of ions. This additional complexity has not been taken into account in the simulations.

3 EFFECT OF CO_2:

We described here different potential effects induced by the presence of CO_2 in the caprock.

Geochemical alteration:

The geochemical reactivity of caprock formations should be evaluated gradually essentially in two steps: classical batch experiments on crushed or small pieces of rock samples to evaluate reaction paths and possibly the reaction kinetics, and flow tests on plugs to evaluate, among other important quantities, the porosity variations. In fact, the second type of experiments was never performed because of the very slow reactivity observed in batch experiments. Two types of formation were investigated: the Callovo-Oxfordian formation containing the largest amount of clays using samples from the SMB field and the Bure area, and the Comblanchien formation consisting essentially of tight carbonates with a very small amount of clays, above the Dogger formation.

The initial condition of the experiment is very important and water must be equilibrated prior to starting the experiments. There are also several choices of experimental parameters. Two temperatures (80 and 150°C) were chosen in order (i) to represent a deep storage case and (ii) to accelerate the kinetics in order to maximize reactivity even if this high temperature may generate components that may not exist at a lower temperature. The pressure was set at 150 bar and the duration varied up to 6 months. Experiments were performed with super critical CO_2 present as a separate phase, and dissolved in water. An example of the results obtained on the Callovo-Oxfordian formation after 6 months of exposure is shown in Figure 4 [14]. In a triangular diagram comprising the most appropriate poles (M+ vs R^{2+} vs 4Si), reaction trends are very difficult to identify at any temperature. This is not to say that no reaction occurred but the complex natural mineralogical composition envelop all reactions. No new component is formed and no component originally present in the rock has disappeared. In other experiments on the same formation and using an image analysis technique [13], an illitization of the illite-smectite components was identified, as well as the formation of gypsum. But similarly, these reactions paths are very difficult to identify using standard bulk measurements such as X-ray diffraction (XRD). With such difficulties for identifying reaction paths, the reaction kinetics is obviously out of reach.

Modification of mechanical properties

Despite the slow reactivity of minerals, the alteration of the pore structure by CO_2 cannot be ruled out. For the study of mechanical properties after potential alteration, another approach was taken to modify the pore structure. Homogeneous (and efficient) alteration can be performed using a retarded acid solution [12]. It consists of injecting the acid solution at ambient temperature, and then activating this acid at 60°C. This procedure can be repeated several times in order to obtain different degrees of alteration. It represents an extreme alteration because of the low pH (between 3 and 4) and the number of alteration cycles (from 3 to 6). As a result, the porosity was modified uniformly by a few units, i.e. the final porosity after alteration increased from an initial 5 to about 7 % for example. When considering only the samples from the Comblanchien formation in reference 12, a small decrease of both the shear modulus and drained bulk modulus were observed. Similarly, the failure points were not significantly modified by the alteration. However, because the alteration effects are of the same order of magnitude as the natural variability between samples, it is quite difficult to obtain any consistent trends.

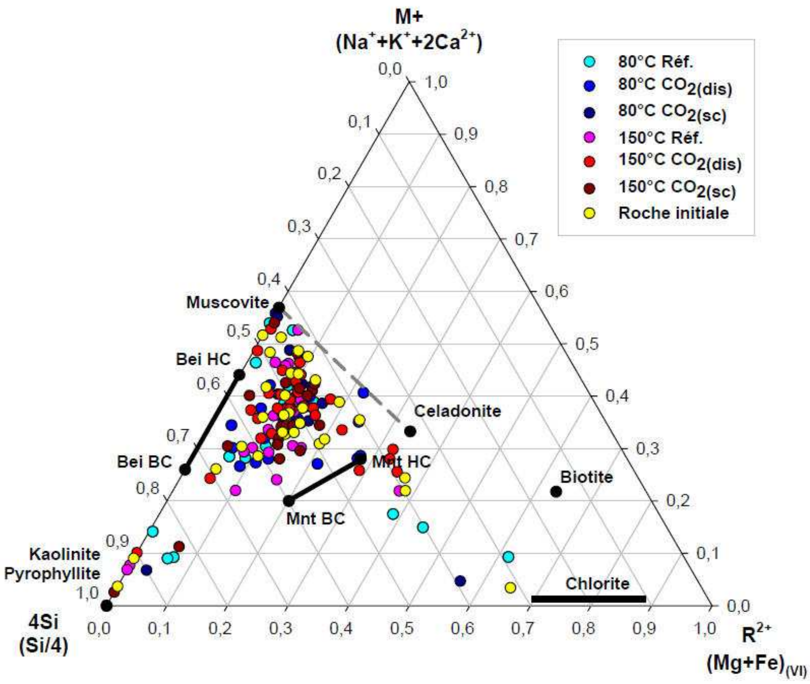

Figure 4: Triangular diagram summarizing the TEM mineralogical analysis of samples exposed to CO_2 at 150 bar during 6 months at 80 and 150°C. The dispersion of the points representing the rock before alteration is similar after CO_2 exposure (from reference 14).

Modification of capillary entry pressure (wetting properties)

The capillary entry pressure is defined as the CO_2 entry (or displacement) pressure minus the brine pressure in the caprock. The assumption is often made that the capillary entry pressure into the brine-saturated caprock is proportional to the capillary entry pressure of an inert gas (such as N_2) into the brine-saturated caprock, or to the mercury entry pressure into the dry rock. The proportionality coefficient is equal to the ratio of interfacial tensions between the non-wetting (CO_2, N_2 or mercury) and wetting (water, mercury vapor) fluids. Hence, possible wettability alteration effects are overlooked.

A series of contact angle measurements has been undertaken to assess whether dense CO_2 is able to alter the strongly water-wet behavior of typical caprock minerals, such as quartz, mica, and calcite. The experimental setup and procedure used in a preliminary set of measurements [10] were improved and the final results show that the contact angles corresponding to the drainage process of interest (i.e., CO_2 displacing water) were barely affected by an increase in CO_2 pressure up to 14 MPa [11, 22]. Hence, even though all types of rock substrates have not been tested, it can be reasonably assumed that dense CO_2 does not significantly alter the water-wettability of typical rock minerals, at least in the drainage process of CO_2 displacing water. This is a rather important simplification because various data and simple measurements can be used with different fluid systems (air-brine, mercury, etc) providing they are corrected for interfacial tension.

Modification of transport properties (permeability, diffusion)

Modifications of permeability and diffusivity before and after alteration by CO_2 were not systematically studied in this program. Increase of permeability (a factor of two) and diffusivity (50 %) are reported in literature [21, 23] after several flow through experiments. Some experiments performed at CEA (P. Berne, personal communication) indicate also a slight increase of diffusion coefficients after CO_2 exposure. However, even if these increases are significant, they are well contained within the natural variability of permeability and diffusivity of caprock formation. These variations also strongly depend on the protocol used for altering the samples. It is worth noting that not variation of pore sizes (as measured by MICP) nor specific surface (as measured by BET) were observed in reference 21, strongly suggesting that the permeability variations are due to the slight modification of preferential pathways in the sample. This is a major difficulty when dealing with plug measurements.

4 LARGE SCALE GEOMECHANICAL IMPACT OF CO_2 INJECTION AND CO_2 MIGRATION IN THE CAPROCK

At the storage scale, two aspects were studied: the possibility of caprock mechanical failure investigated by 2D hydromechanical simulations, and the migration of CO_2 due to diffusion and two phase flow investigated by 1D reactive transport simulations.

Impact of overpressure on geomechanical properties:
The main objectives are to predict the possibility of tensile fracturing and the shear slip reactivation of pre-existing fractures. The difficulty of hydromechanical simulations is that all layers above and below the caprock and reservoir formations must be considered at a fairly large scale (typically 50 km). Therefore, there is an unavoidable simplification because most of the fine details are not known. In addition the overpressure propagation is much larger than the CO_2 front and therefore, two mechanisms with very different length scales (and hence mesh sizes) must be coupled. Since geomechanical and transport simulators are usually designed and run separately, the coupling is performed externally either by introducing the pore pressure history into the geomechanical simulator [e.g. 24], or by updating both simulations in terms of pressure at given time steps [16].

In the frame work of this project, an injection into the Dogger aquifer was studied using different scenarios [16]. The rate of injection of CO_2 is 10 Mt/y at a single location during the first ten years of simulations (such a rate is certainly an upper limit in practice). The reservoir permeability is about 90 mD with a high permeability layer (40 m) at 700 mD present at mid height (total thickness 150 m). In these conditions, the maximum overpressure, defined as the difference between the initial and final average pore pressure, is a combined effect of gravity and injection pressure. It reaches a maximum of 34 bar vertically above the injection point at the reservoir caprock limit (Figure 5). Interestingly, it can be noted that most of the pressure build-up is occurring within the first year of injection, when the lateral extension of the plume is limited. In these conditions, it can be shown that the caprock stays in compression, therefore preventing the tensile fracturing possibility. More important, the shear fracture reactivation criterion is never reached, even when taking lower permeabilities.

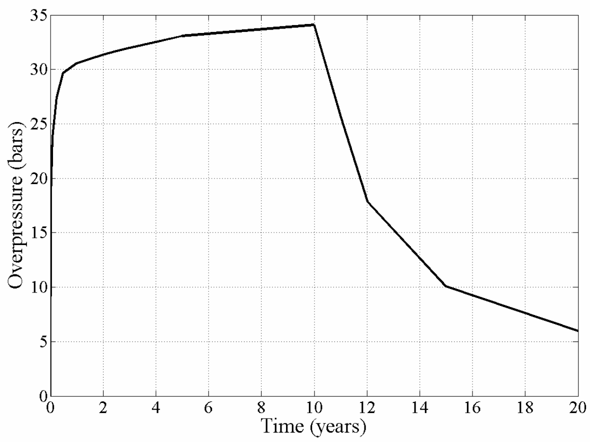

Figure 5: Hydromechanical simulations of the injection of CO_2 into the Dogger formation (from reference 16). The maximum overpressure is observed above the injection point at the reservoir-caprock limit. The injection is stopped after 10 years.

Scenarii of 1D modeling of CO_2 migration in caprocks:
The purpose of these simulations is to quantify the space and time variation of the caprock properties when CO_2 is migrating either by diffusion or by multiphase flow in the case of a breakthrough. For the sake of simplicity, only a 1D geometry was considered, representing a column of caprock in contact with the reservoir. Due to buoyancy effects, the plume is rapidly migrating upward around the injection point and reaches the caprock-reservoir interface quite rapidly (as seen in the previous paragraph). Hence, the simulated situation is a vertical column of caprock those lower end is in contact with CO_2. In a similar way as studied by Gherardi et al. [25] reactive transport was simulated over long periods of time [17]. Several scenarii were considered; essentially: reactive diffusion of dissolved CO_2 from the reservoir into the caprock, reactive two phase flow and

diffusion through the caprock. A clay rich carbonate caprock is considered (as in Figure 2). A key result is shown in Figure 6, indicating the vertical changes of porosity at different times. A dissolution-precipitation front pattern is seen in both diffusion and two phase flow cases. Dissolution is occurring in the first few decimetres of the caprock with an increase of porosity from an initial 15 % up to about 16.5 %. Above, calcite precipitation is occurring with a much larger vertical extension in the case of two phase flow. In the latter case, a constant overpressure is assumed during all the simulation and therefore represents an extremely pessimistic situation. Despite a relatively high absolute permeability of 1 µD, the two phase front has a vertical extension that does not exceed 10 m. In fact, the CO_2 effective permeability is governed by relative permeability curves, and is in practice about two orders of magnitude smaller than the absolute permeability. This explains the relatively small vertical extension in the case of two phase flow. From the geochemical point of view, the buffering capacity of the caprock is a key factor for limiting the effect of CO_2 to small changes of porosity. We conclude that the migration of CO_2 into the caprock is limited if the permeability is low enough and that no major geochemical effect is occurring.

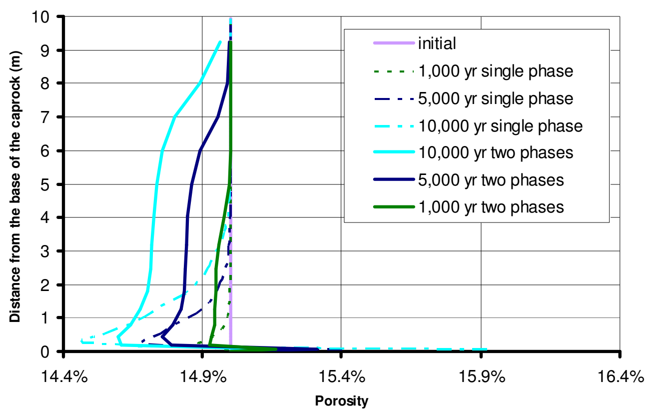

Figure 6: Results of the reactive transport simulations. The single phase simulation indicates pure diffusive transport, and the two phase case is a two phase flow into the caprock after breakthrough. Permeability: 1 µD, pore diffusivity: $D_p=10^{-11}$ m^2/s.

5 CONCLUSION

The primary criteria for the selection of a caprock formation for CO_2 storage purpose are the thickness and permeability of the formation. An insufficient capillary entry pressure is not a criterion for rejecting a formation for storage purpose because the CO_2 migration may be very limited in time and space. The various scenarii for estimating the extend of the migration can be studied using numerical simulations, even though some petrophysical and geochemical parameters may remain uncertain. At large scale, caprock characterization requires at least seismic profiles to identify lateral continuity. When well logging data are available, simple rules based on clay content can be used to estimate thicknesses and lithoseismic interpretation usually performed in reservoir zones can be applied to caprocks. For the formation considered, the geochemical reactivity to CO_2 was small for clay minerals, making reaction path difficult to identify. Similarly, artificial alterations of samples representing extreme situations had little impact of geomechanical properties. Finally, with realistic overpressure due to injection, shear fracture reactivation criteria are not reached and migration of CO_2 either by diffusion or by two phase flow within the first meters of the caprock produce mostly a decrease of porosity by precipitation, and very locally an increase of porosity by dissolution.

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