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## Main Results of the CO<sub>2</sub>-DISSOLVED Project: First Step toward a Future Industrial Pilot Combining Geological Storage of Dissolved CO<sub>2</sub> and Geothermal Heat Recovery

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### Abstract

The CO<sub>2</sub>-DISSOLVED project (2013-1016) aimed at evaluating the techno-economic feasibility of capture and storage in a saline aquifer of dissolved CO<sub>2</sub>, coupled with geothermal heat recovery through a standard doublet system. This project mainly investigated: (1) the hydro-geochemical behavior of the injected CO<sub>2</sub> plume in the aquifer, relying on reactive transport modeling, lab experiment and on-site measurements; (2) the integration of a breakthrough CO<sub>2</sub> capture technology with major environmental and economic benefits; (3) the assessment of the economic viability on a bioethanol refinery test-case. All the results obtained confirmed the viability of the concept, which was proved to be potentially applicable to several areas in France, Germany, and the USA. The recently launched second phase of the project now aims at preparing the ground for a future industrial pilot.

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## 1. Introduction

The CO<sub>2</sub>-DISSOLVED project, funded by the ANR (French National Research Agency), mainly is a techno-economic study whose objective is to assess the feasibility of a new CO<sub>2</sub> Capture and Storage (CCS) concept [1]. This concept combines capture, injection, and storage of CO<sub>2</sub> in a deep saline aquifer as being entirely dissolved in brine, rather than as a supercritical state, and geothermal heat recovery (see Fig. 1). All these operations are planned to be performed locally, thus avoiding the problems related to the infrastructure and cost of CO<sub>2</sub> transport.

Basically, a CO<sub>2</sub>-DISSOLVED facility comprises a classical low-enthalpy geothermal doublet from which the warm water (ca. 50-90°C) is extracted, the delivered energy being recovered through a heat exchanger system, and then the cooled brine (ca. 40°C) is injected back to the aquifer after being saturated in dissolved CO<sub>2</sub>. The technology selected for CO<sub>2</sub> capture is provided by Partnering in Innovation, Inc., our American partner in this project [2]. The interest of this innovative capture system is double: (1) environmental, as no other solvent than water is used for capture, and (2) economic, because of an optimized energy consumption and in-process Sox, Nox, Hg, Se removal (thus avoiding an expensive gas pretreatment), leading eventually to a capture price approximately divided by two with respect to competing post-combustion technologies available on the market.

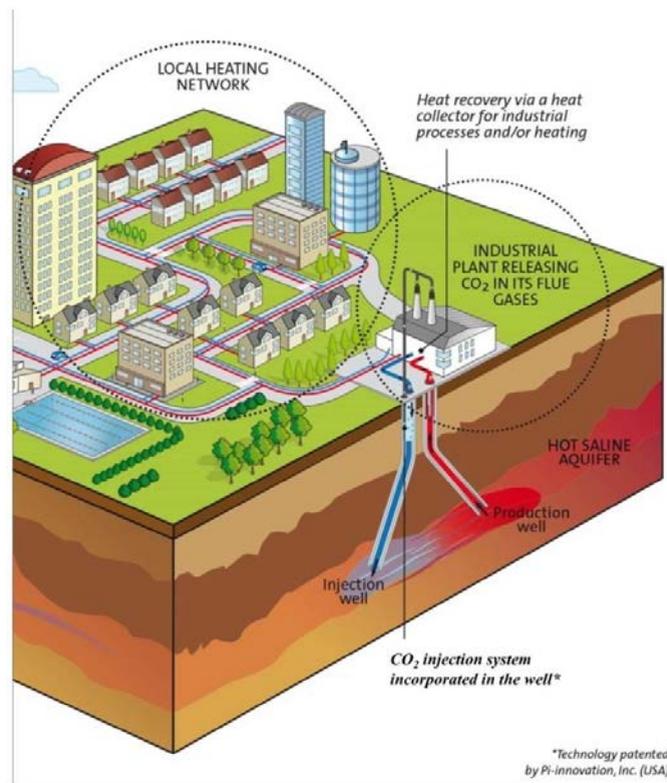


Fig. 1. The CO<sub>2</sub>-DISSOLVED concept combining (1) local storage in a deep saline aquifer of dissolved CO<sub>2</sub>, and (2) geothermal heat recovery.

One of the key features of this innovative CCS concept is the use of dissolved CO<sub>2</sub> in a geothermal doublet. The advantages of using a doublet system for dissolution and injection, with no gas phase being present in the aquifer, are many: no build-up effect in the aquifer because the amount of injected water is exactly balanced by the amount of pumped water, no large distance displacement of the brine initially in place since the impacted area remains centered around the well footage, low to no escape risk of the injected CO<sub>2</sub> to the surface as it remains trapped by dissolution into the brine. However, the CO<sub>2</sub> solubility in brine is a physical limitation on the amount of CO<sub>2</sub> that

could be injected in the aquifer to guarantee that no gas phase is present. Based on the typical water flow-rates obtained in the geothermal doublets of the Paris basin (200-350 m<sup>3</sup>/h), and considering typical downhole pressure and temperature conditions (70°C, 150 bar), we calculated that a single doublet could then typically dissolve up to 80-150 kt/a of CO<sub>2</sub>. Consequently, it appears that the CO<sub>2</sub>-DISSOLVED concept is well suited for small-medium industrial CO<sub>2</sub> emitters with same orders of magnitude of annual CO<sub>2</sub> emission rates and, as such, is complementary to the ‘classical’ supercritical CCS approach which usually addresses high-rate emitters (ca. 1-10 Mt/a).

This paper presents the main results acquired in the framework of this 3-year project (2013-2016), and discusses about the ongoing second phase which just started in June 2016 aiming at preparing the ground for a future CO<sub>2</sub>-DISSOLVED industrial pilot.

## 2. A concept potentially applicable

One of the preliminary tasks of the CO<sub>2</sub>-DISSOLVED project was to map the sites potentially compatible with this technology in all the countries involved in the project: France, Germany, and the USA. It mainly consisted in identifying and prioritizing the industrial emitters that could potentially benefit from the application of this new CO<sub>2</sub> storage strategy, *i.e.* determining regions where geothermal resources match the presence of industrial plants emitting small rates of CO<sub>2</sub> (< 150 kt/a). More detailed information can be found in [3].

The CO<sub>2</sub> emissions data were acquired from publically available national databases. However, it has to be noticed that no data about the total compositions of the emitted smokes of the stationary CO<sub>2</sub> emitters could be found in the public databases available. This information is really important when dimensioning a facility, in particular for the CO<sub>2</sub> separation unit, when needed. Despite this lack of flue gas composition data and some discrepancies between the exact content of the databases for each of the three countries investigated, promising results could however be obtained on the potential for applying this technology to industrial emitters.

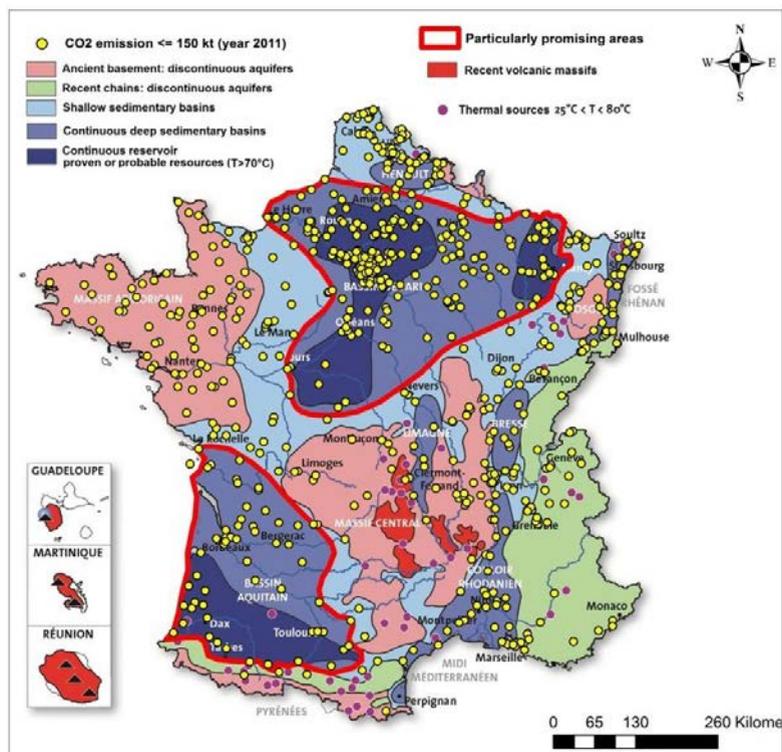


Fig. 2. Small to medium CO<sub>2</sub> emitters (ca. 10-150 kt/yr, yellow spots) vs. geothermal resources in France (modified from [4]).

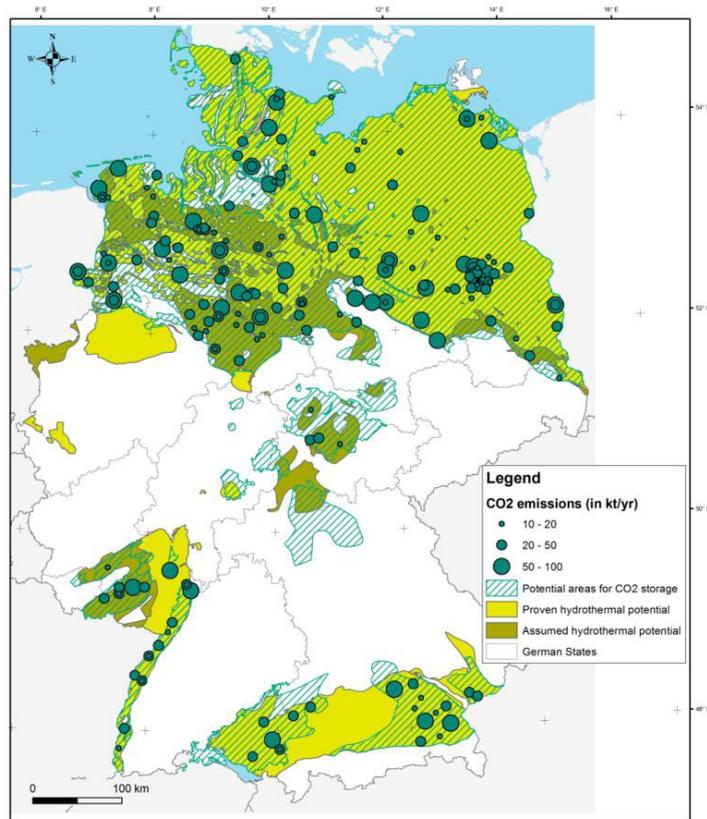


Fig. 3. Selection of 225 small to medium CO<sub>2</sub> emitters (green spots) in Germany located in potential areas both for hydrothermal energy use and CO<sub>2</sub> storage (after [5] and [6]).

In the three countries, there are numerous small to medium emitters listed in the databases. They represent 83, 70, and 46% of all the total CO<sub>2</sub> emitters in France, the USA, and Germany, respectively. However, it has to be noticed that France and Germany have quite similar numbers of small and medium CO<sub>2</sub> sources (881 and 800, respectively). As expected because of the size of the country, in the USA, the number of this category of emitters is much higher (4,645). Moreover, the total CO<sub>2</sub> annually released by these low emitters is also almost the same in France and Germany (33.5 and 32.4 Mt, respectively) while it is five times higher in the USA (200.6 Mt). When comparing the locations of small to medium emitters with areas of geothermal resources, it can be concluded that in all three countries there are relatively large areas matching the presence of many potential candidates for the CO<sub>2</sub>-DISSOLVED concept.

In France, the areas where the geothermal resources could potentially match the compatible industrial CO<sub>2</sub> emitters are composed by all the major sedimentary basins, *i.e.* the Paris Basin, the Aquitaine Basin, the Upper Rhine Graben, the Limagne and Bresse regions, and the Rhone corridor (blue and dark blue areas in Fig. 2). Then, 653 small to medium French emitters can be considered as potentially compatible with the CO<sub>2</sub>-DISSOLVED concept (Fig. 2). These 653 CO<sub>2</sub> sources have emitted a total amount of 25.1 million tons of CO<sub>2</sub> in 2011 (16.9% of the total French CO<sub>2</sub> emissions).

In Germany, only the hydrothermal potential areas (proven or assumed) were considered for determining the potential areas of geothermal energy use. 242 small to medium emitters were then located in favorable areas both for hydrothermal energy use and CO<sub>2</sub> storage (Fig. 3). In total, these 242 CO<sub>2</sub> sources emitted 9.98 Mt of CO<sub>2</sub> in 2012 (7.1% of the total CO<sub>2</sub> emissions).

In the USA, less detailed information was available on the exact location of the emitters (only the total numbers of emitters per state were published). Considering that the eastern part of the U.S. is almost devoid of geothermal resources, only a few states such as New York, Pennsylvania and West Virginia have low-temperature geothermal systems. Thus, the potential areas where the CO<sub>2</sub> storage could be coupled with geothermal activity are mostly concentrated in the western part of the USA, including Alaska and Hawaii.

### 3. Designed to be a turn-key technology

The CO<sub>2</sub>-DISSOLVED system basically relies on well-known elements: a standard low-enthalpy geothermal doublet coupled with a heat exchanger system at the surface. The only specificity that has to be considered for the design of the doublet is the circulation of acidified CO<sub>2</sub>-laden brine, likely to enhance corrosion issues in mild steel materials generally used for wellheads and casing strings. Corrosion-Resistant Alloys (CRAs) would be valid candidates in the replacement of mild steel in this context, in particular because of their low rate of general corrosion. However, their cost is prohibitive and they still have a tendency to suffer from localized corrosion phenomena (including pitting corrosion, extending to stress-corrosion cracking if exposed to tensile stresses). Consequently, this substitution material is not a good candidate for an implementation in a CO<sub>2</sub>-DISSOLVED system. On the other hand, composite materials have improved over the past years and their mechanical properties are now suited to meet the requirements of a geothermal well. Furthermore, the resins and fibers they are made of are nearly unaffected by the chemistry of the geothermal fluid. Rough economic calculations show that the initial cost (wells and surface facilities) of a typical geothermal doublet in the Dogger aquifer of the Paris basin, equipped with fiberglass casings, is expected to be around 5% higher than for the same doublet equipped with steel casings. However the operating costs are anticipated to be lower, particularly because of reduced electrical and inhibitor consumptions as well as lighter maintenance operations. Consequently it is estimated that the initial additional cost would be paid off after a few years of operation (less than 5). Consequently, the choice was made to use glass-reinforced epoxy (GRE) rather than steel for both the injection and the production wells casings [7].

The main novelty in our system comes from the capture technology that we selected (see [2] for detailed description). This innovative technology was brought to the project by our American partner (Partnering in Innovation, Inc.). In a brief overview (see Fig. 4), the Pi-CO<sub>2</sub> capture method uses water as a physical solvent, circulating the water and emission gas through a cascade mass transfer system (MTS) located in a sealed deep large diameter well under ca. 25-60 bar hydrostatic pressure. The hydrostatic pressure significantly increases the solubility of gases in water. The system is closed loop with the high pressure non-dissolved separated gas fraction diverted to the surface and combined with heat to recover compression energy.

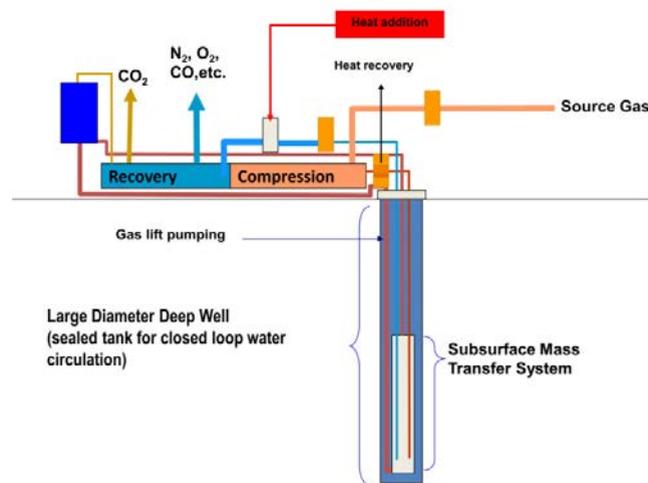


Fig. 4. Simplified view of the Pi-CO<sub>2</sub> water-based in-well capture technology planned to be used in a CO<sub>2</sub>-DISSOLVED system.

The flue gas is injected in the MTS at depth in the deep water column. The gases ( $\text{CO}_2$  and lesser competing gases) are concentrated through a cascading series of absorbers in the MTS (proprietary detail not shown). Water returning to the surface from the MTS becomes less pressured allowing for gas ex-solution; this ex-solution drives the water circulation (gas lift pumping) so that additional energy and mechanical pumping are not needed for circulation. The non- $\text{CO}_2$  ex-solved gases are sequentially removed in the return line to produce nearly pure  $\text{CO}_2$  product. The system integrates compression and energy recovery processes at the surface to reduce parasitic load with heat exchange and turbo-machinery (similar to a Compressed Energy Storage System). Uniquely, the Pi- $\text{CO}_2$  process removes  $\text{SO}_x$ ,  $\text{NO}_x$ , vaporized metals, while capturing  $\text{CO}_2$ , in a single integrated process. The oxides are removed in compression condensate and at inter-cooler and after-cooler steps during flue gas compression [8]. This in-process feature avoids expensive pre-treatment of the flue gas. Another interesting feature of the Pi- $\text{CO}_2$  system is its expected easiness of construction since all of the surface turbomachinery, heat exchange, and shaft installation equipment is currently available “off shelf”. Moreover, as much of the installation is underground, the surface footprint is small.

However, in the  $\text{CO}_2$ -DISSOLVED system, the surface recovery of gaseous  $\text{CO}_2$  is not needed since we intend to inject dissolved  $\text{CO}_2$ . Consequently, we initially thought about integrating the whole MTS within the injection well of the doublet in order to both capture the  $\text{CO}_2$  from the flue gas by dissolving it in the circulating brine, and then inject at the same time the  $\text{CO}_2$ -laden brine back to the aquifer instead of being lifted up to the surface. However, the main drawback of this technical solution is related to the required height of the MTS as part of the injection well: it could then be a potential limiting factor in terms of achievable brine circulation flow rate, and moreover, would certainly render more difficult the maintenance operations in the well. On the other hand, the efficiency of the MTS for producing sufficiently high  $\text{CO}_2$  rates is basically dependent on both the depth and the diameter of the well, the larger (*ca.* > 1 m) being, the better. In geothermal wells, the well diameters used are relatively small (*ca.* 0.2-0.25 m) but correspond to standard values that we cannot reasonably avoid using in our system design. Consequently, considering those aspects, we finally concluded that this solution would not be viable for our project.

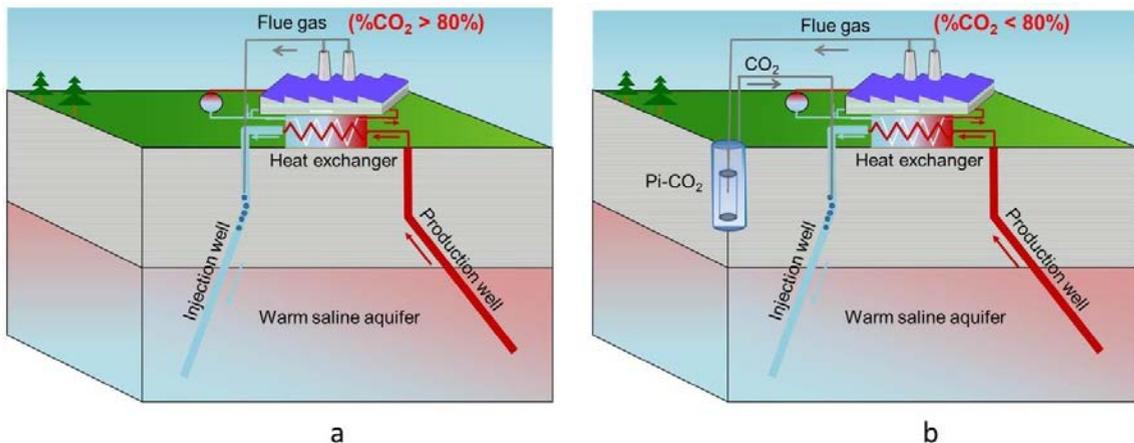


Fig. 5. Typical design of a  $\text{CO}_2$ -DISSOLVED facility: (a) simplified version, close to a standard geothermal doublet, when the flue gas contains a high  $\text{CO}_2$  concentration; (b) standard version including the geothermal doublet and a third large-diameter well housing the Pi- $\text{CO}_2$  mass transfer system, when the  $\text{CO}_2$  rate in the flue gas is lower than 80%.

The option of using a separated large-diameter well housing the Pi- $\text{CO}_2$  system and dedicated to the  $\text{CO}_2$  capture operations was then considered (cf. Fig. 5b). With this solution, this third well would be designed according to the actual needs in terms of  $\text{CO}_2$  separation and injection, depending on the targeted flow-rate and on the flue gas composition. Once recovered at the surface, the separated  $\text{CO}_2$  gas phase would then be injected in the doublet at a controlled mass-rate through a dedicated small-diameter pipe. This pipe would be ended at depth by a bubbler, specifically dimensioned to ensure complete  $\text{CO}_2$  dissolution in brine before it reaches the storage aquifer. Mass

transfer modeling proved the adequacy of such a system for easily dissolving several tens of kilotons of CO<sub>2</sub> per year [9]. CFG Services confirmed that this system could be easily fitted in a standard geothermal injection well after a slight modification of the well head (equivalent to what is done for integrating an inhibitor injection line). An equivalent injection system for injection and dissolution at depth of CO<sub>2</sub> was successfully tested on the CarbFix site in Iceland [10].

However, it has to be noticed that the gas separation step is not necessary in all cases. For instance, on a test-case on a bio-refinery plant studied in this project, the flue gas generated by the distillation process was almost pure CO<sub>2</sub>. In such cases, we can avoid drilling the third well and using the full Pi-CO<sub>2</sub> technology. This could be generalized to other industries emitting smokes with high CO<sub>2</sub> content (ca. > 80%). Consequently and as illustrated on Fig. 5, two cases have to be considered in designing a CO<sub>2</sub>-DISSOLVED facility:

- (1) When the flue gas has a high CO<sub>2</sub> grade (ca. > 80%), no CO<sub>2</sub> separation unit is necessary. Gaseous CO<sub>2</sub> could then be directly co-injected with water into the injection well, using a specific small diameter in-well CO<sub>2</sub> injection pipe ended by a bubbler, at controlled relative flow rates to guarantee the complete dissolution of the gas before the fluid reaches the well shoe.
- (2) When the CO<sub>2</sub> concentration in the emitted smokes is lower than 80%, which is the most general case, a gas separation unit is necessary. A specific large diameter well housing the Pi-CO<sub>2</sub> system would then be drilled, in order to house the MTS and separate CO<sub>2</sub> from the flue gas. Then CO<sub>2</sub> would be co-injected in the injection well in a similar manner to what is done in case 1).

The first case obviously leads to a more simple design, remaining very close to that of a classical geothermal doublet. The second case involving the implementation of the full Pi-CO<sub>2</sub> technology is more complex and expensive. However and as mentioned earlier, the Pi-CO<sub>2</sub> technology mainly relies on an innovative assembling of existing elements and remains far less expensive than competing post-combustion technologies available on the market. Consequently, in both cases, the full technology can be delivered to an industrialist committed in reducing its CO<sub>2</sub> emissions, the step from design and modeling to an actual installation being quite reasonably achievable.

#### 4. Geochemical impact in the aquifer of CO<sub>2</sub> injection

Once out of the injection well, the acidified brine is expected to be chemically reactive with the porous matrix of the aquifer, particularly in the presence of carbonated minerals. In order to assess the viability of the injection process, it was indispensable to quantify these effects. For that purpose, three scales were investigated combining lab experiments, field measurements, and reactive transport modeling: (1) the first centimeters around a miniature injection well as part of the MIRAGES-2 experimental facility [11, 12]; (2) The first meters around the open-hole part of the former injection well of a geothermal doublet in the Dogger aquifer of the Paris basin (for the cooled brine injection effect only, without CO<sub>2</sub>); (3) The first hectometers to kilometers around the doublet wells with purely numerical investigations [13]. The key results acquired are presented in the following paragraphs.

##### 4.1. Lab experiments at the centimeter scale

A dedicated experimental facility (MIRAGES-2), specifically designed for injecting a CO<sub>2</sub>-rich aqueous solution in a well at the 1/20<sup>th</sup> scale [11], supported a series of experiments with both carbonate and sandstone mineral samples. The experimental facility was entirely designed from scratch; it is divided in two parts: the first one is devoted to the CO<sub>2</sub>-solution mixing process, and the second one enables to perform the injection of the prepared CO<sub>2</sub>-rich solution in a core sample through a small steel tube inserted in the core and mimicking an injection well, under controlled pressure and temperature conditions typical of reservoir conditions of interest for our project.

The most visually remarkable results were obtained with the carbonate samples, showing the fast development of a network of wormholes after injection of either “pure water” (equilibrated with atmospheric CO<sub>2</sub>) or a CO<sub>2</sub>-rich solution [12]. The numerical simulation of this experiment with MARTHE-PHREEQC [14, 15, 16] reproduced reasonably well the observed massive dissolution of calcite near the injection tube, and the formation of a simplified

wormhole-like pattern initiated by a prescribed heterogeneity in hydraulic conductivity. For more details, we invite the reader to read [12].

In a second series of experiments with the same carbonate sample (Lavoux limestone), the effect of salinity (15 g/L NaCl) was investigated. The results showed a significant increase in dissolution after 21 days of continuous injection of the acidified brine: 2.58% of the total volume of the sample was dissolved in this case, compared to 0.57% without salt. Furthermore, with this saline solution, all the dissolution was concentrated on a single larger wormhole rather than in a wormhole network like in the first case. Conversely, the cement alteration is less important with salt than without salt. More interpretation and modelling work is still required on this particular example.

As expected, the effects observed for the sandstone experiment (without salt) are very weak: no visible dissolution of the rock but significant cement dissolution can be seen at the interface between sandstone and cement, near the injection point.

Looking at the corrosion processes on both the steel injection tube and the composite material sample inserted near the injection point, nothing was visible, whatever the composition of the solution used. Concerning the composite sample representative of the material selected for the wells casings (rather than steel, see §3), this is not very surprising as it was selected for a very low to no sensitivity to corrosion processes. About the steel tube, the relatively short duration of the experiments (21 days) probably explains this lack of reactivity.

All these results are extensively described in a report currently under final review [17].

#### *4.2. On-site measurements and numerical investigations at the well scale*

Because at this stage, numerical modeling was the only way to assess the expected dissolution effect induced by the acidified brine injection in a carbonated aquifer immediately out of a full-scale well, we performed a series of reactive transport calculations [13] using the MARTHE-REACT [18] and MARTHE-PHREEQC numerical codes. According to the scenarios investigated on the Dogger carbonated aquifer, the results clearly suggested a massive dissolution effect (up to 100% of calcite and dolomite initially present) on a distance of a few tens of meters around the well for a continuous injection of a cooled brine saturated with CO<sub>2</sub> on a period of 30 years (30 years being a typical operational lifetime of a geothermal doublet in the Paris basin). In order to check the potential geomechanical consequences on the reservoir, a risk analysis study was conducted [19]. The results first suggested that the more probable dissolution patterns initiated at the pore scale are uniform dissolution (only dissolution pattern considered in our calculations) and wormhole dissolution (similar to what was observed in the MIRAGES-2 experiments). Regarding the mechanical impact, the models used showed negligible effects of reservoir compaction or cavity collapse, possibly due to dissolution, on surface subsidence and horizontal surface strain, notably because of the great depth of the reservoir (ca. 1,500 m).

However, a similar dissolution effect, in a lesser extent, is also expected out of a standard geothermal injection well targeting a carbonated aquifer such as the Dogger of the Paris basin, because the cooled brine is chemically undersaturated with respect to the minerals composing the porous matrix (mainly calcite in our case). This was also investigated numerically and the results showed 100% calcite dissolution on a distance of a couple of meters around the well after 30 years of continuous cooled brine injection.

These numerical results, based on a simplified approach (2D radial model, homogeneity and isotropy of both the geochemical and hydrodynamic properties of the aquifer all along the single productive layer considered) are important as a first guess of the potential impact of dissolved CO<sub>2</sub> and cooled brine injection. However, in this project and using the well investigation tools made available by FLODIM (<http://www.flodim.fr/>), we had the opportunity to perform a first-of-a-kind on-site experiment aiming at providing an image of the open-hole part of a former geothermal injection well used during the past 27 years. Using sonar to image the hole, we interestingly observed on a 40 m height along the open-hole that the maximum radius of the cavity measured on the productive layers reached about 1 m (vs. 20 cm when the well was drilled). This is very consistent with the calculation results and makes us more confident on our capacity to predict the impact of both cooled brine reinjection and dissolved CO<sub>2</sub> injection. However, as this experiment was conducted at the very end of the project, we did not yet finalized the results interpretation. So far, it looks like our calculations for the cooled brine reinjection slightly overestimate the geochemical impact with respect to what was observed on-site. This will have to be confirmed by further interpretation and modeling work.

### 4.3. Numerical study at the doublet scale

The objective of these 3D simulations at the doublet scale (*i.e.* on a kilometric scale around and between the wells) was mainly to investigate both the thermal and the geochemical changes over time induced by the cooled acidified brine injection. The thermal reactive transport codes MARTHE-REACT and MARTHE-PHREEQC were also used for these simulations.

Based on the well-known aquifers exploited for geothermal heat production in France, generic test-cases were studied, assumed to be representative of (1) a carbonated aquifer (Dogger of the Paris basin - 1,500 to 2,000 m deep, water temperature of 70°C) and (2) a clastic reservoir (Triassic sandstones of the Paris basin - 2,000 to 2,500 m deep, water temperature of 90°C).

In order to maximize the geochemical effects, we assumed that the CO<sub>2</sub> concentration in the injected brine was at a constant value of 1 mol/L all along the injection period, which is close to the solubility limit for the temperature and pressure conditions considered. As a specific paper presenting the simulation results was written for this conference [13], one should refer to it to get more detailed information. However, what can be concluded from these simulations is:

- (1) A massive dissolution of carbonates (calcite and/or dolomite) around the injection wells can be observed on both the carbonated and clastic cases. On the Dogger-type tests-case, this carbonate alteration obviously impacts the porosity, so that a significant increase in porosity can be observed around the injection well. This corroborates the great reactivity of the carbonated minerals under the injection of a CO<sub>2</sub>-rich brine that was observed experimentally [12]. It has to be noticed that a slight increase around the production well was also evidenced by the calculation results, but in a much lesser extent than at the injection well.
- (2) In the clastic reservoir case, a porosity increase can also be observed around the injection and production wells. However, contrary to the carbonated aquifer case, the impact on porosity is here much lower due to the weaker carbonate content (5% against 97.5% for the carbonated aquifer). We also observed a very slight dissolution of quartz and K-feldspar initially present in the clastic reservoir. Illite and kaolinite, considered as secondary phases, precipitated. However, taking into account the corresponding precipitated volumes did not lead to any significant impact on the reservoir porosity [17].

## 5. Best injection scenario and operational lifetime

The question of the expected lifetime of a doublet is central to determining the viability of a geothermal project. Generally, the points that are critical are the capacity of the doublet to deliver an appropriate energy rate and the mechanical degradation of the wells over time. Assuming no significant decrease in the hydraulic conductivity of the exploited aquifer, the first point is directly related to the temperature of the extracted fluid which will inevitably tend to decrease over time, due to the heat transfer processes induced by the continuous reinjection of cold water. When the extracted fluid is no longer warm enough, drilling a new production well is a solution that can be envisaged. The aspects related to the well degradation can be solved with regular work-over periods and appropriate chemical treatment. However, after a while, the well is no longer viable for production and the drilling of a new well has to be considered. Based on the experience acquired in the exploitation of the Paris basin, a typical lifetime of 30 years can be considered as typical.

In the CO<sub>2</sub>-DISSOLVED case, we add another difficulty linked to the injection of CO<sub>2</sub>. Apart from the abovementioned questions about the geothermal doublet itself, we now have to assess what could be the lifetime of the storage part of the CO<sub>2</sub>-DISSOLVED system. Actually, it is expected that a fraction of the injected dissolved CO<sub>2</sub> is going to be produced at the extraction well after a certain period of time, due to the mass transfer processes induced by injection. It is well known however, that mass transfer processes are much faster than heat transfer.

Aiming at assessing the lifetime of such a CO<sub>2</sub> storage system, we performed several series of simulations with the MARTHE code, without considering any reactive processes [20]. This means that dissolved CO<sub>2</sub> was assumed to behave as a non-reactive tracer in a generic 3D model of the Dogger aquifer. All the simulation results confirm that CO<sub>2</sub> concentration is going to increase significantly (above 10% of the injected concentration) at the production well

after a period varying from 2 to more than 20 years after injection started according to the scenario considered (Fig. 6)

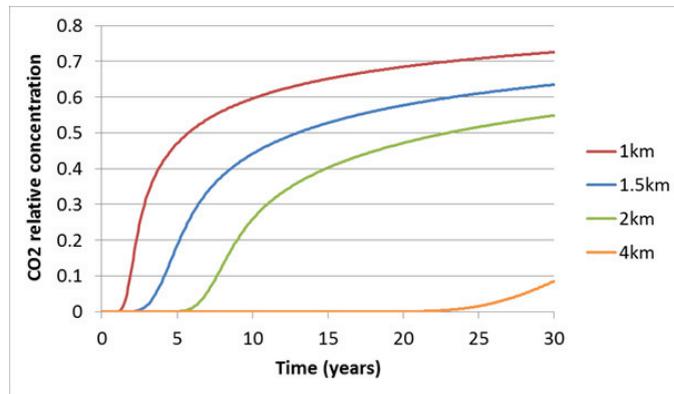


Fig. 6. Dissolved CO<sub>2</sub> breakthrough at the production well as a function of the distance between the injection and the production well shoes at depth, for a constant injection flow rate of 200 m<sup>3</sup>/h.

One important outcome of Fig. 6 is that the key parameter to consider in the design of a CO<sub>2</sub>-DISSOLVED system is the distance between wells at depth. The best strategy is clearly to maximize the distance between the wells shoes. However, this distance is geometrically limited by both the depth of the targeted aquifer, and the maximum drilling deviation angle achievable. Another option to increase this distance would be to pull apart the injection and production well heads which are generally located within a few meter distance.

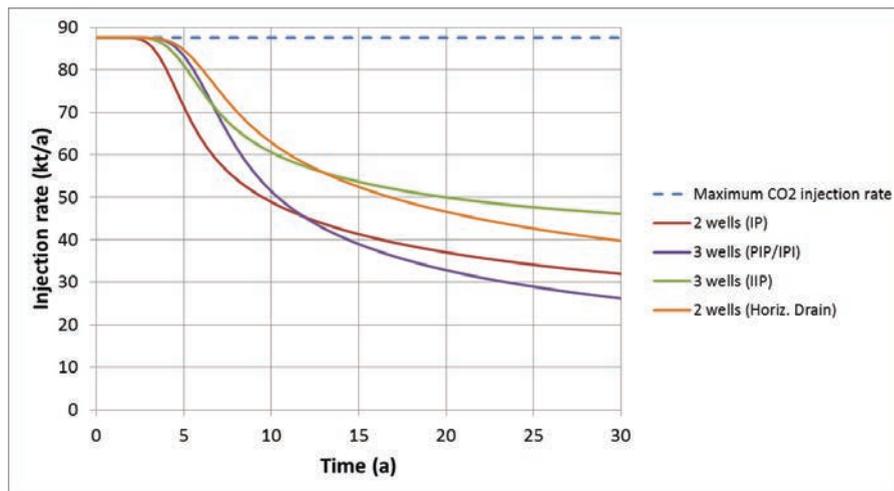


Fig. 7. Evolution with time of the maximum achievable CO<sub>2</sub> injection rate for various injection options (2 or 3 wells). The water flowrate is 200 m<sup>3</sup>/h and the distance between the well shoes is assumed to be 1,500 m. The maximum CO<sub>2</sub> injection rate achievable when the water is fully saturated in CO<sub>2</sub> is represented by the dashed line. IP: 1 injection well, 1 production well (standard doublet); IIP : 2 injection wells, 1 production well; PIP: 2 production well, 1 injection well; IPI: 2 injection wells, 1 production well; Horiz. Drain: Horizontal drain.

These preliminary results finally have to be interpreted as a decreasing CO<sub>2</sub> injection capacity over time rather than as a risk of CO<sub>2</sub> “leakage” at the surface because some of the injected CO<sub>2</sub> would be released by the extracted water. Actually, as the water circulation is closed-loop, none of the produced dissolved CO<sub>2</sub> can be released to the atmosphere. However, because the CO<sub>2</sub> content in the extracted water is going to increase with time (it is initially

close to 0 and several orders of magnitude below the prescribed injection concentration), less CO<sub>2</sub> can be added in the re-injected brine to remain below the solubility limit. The good news is that each drop of dissolved CO<sub>2</sub> injected will remain stored in the aquifer after water circulation is stopped. The drawback is that the amount of potentially injectable CO<sub>2</sub> is going to decrease with time, as illustrated in Fig. 7.

Several injection scenarios involving 2 or 3 wells were tested, considering continuous (constant CO<sub>2</sub> concentration), discontinuous (successive periods of constant CO<sub>2</sub> concentration followed by periods with no CO<sub>2</sub> at all) or alternate CO<sub>2</sub> injection options (when two injection wells are considered, only one of each is used at a time). Discontinuous or alternate options were rapidly discarded as they did not bring any advantage in terms of CO<sub>2</sub> injection capacity and they would have generated operational difficulties on the field. The results presented in Fig. 7 show that the IIP configuration (2 injectors and 1 producer) provides the better injection rate after 30 years of injection (45 kt/a vs. 32 kt/a for a standard doublet). The benefit of those technical solutions using three wells or horizontal wells must however be balanced with their higher initial capital cost and the actual energy needs, in order to define an optimal injection scenario. So far, the standard doublet remains our preferred injection option as being the most experienced and the less expensive.

In all the simulations, we assumed a maximum CO<sub>2</sub> concentration (*i.e.* at saturation) from the very beginning to the end of the injection period. However, this corresponds to a worst case since it is unlikely that the amount of CO<sub>2</sub> to inject be exactly the amount needed to reach saturation. In most cases, the system would be designed considering the actual CO<sub>2</sub> emissions and the actual energy needs. Moreover, these CO<sub>2</sub> emissions rates may have a seasonal variability. Both parameters will enable determining the appropriate water flow-rate and CO<sub>2</sub> concentration to setup. If this concentration is lower than the maximum solubility, we can then expect much favorable injection rate results over the same period of time, allowing to inject a CO<sub>2</sub> rate as close as possible to the actual CO<sub>2</sub> emissions. Ongoing modelling work will enable to confirm this anticipated behavior.

In all cases and although the achievable injection rate tends to decrease with time, we can observe that after a continuous injection period of 30 years, the system still has a capacity to store some CO<sub>2</sub> (even if less than the quantity emitted by the industrial site considered). If we define the storage lifetime as the time during which CO<sub>2</sub> can be injected at a prescribed maximum rate (ideally, equal to the CO<sub>2</sub> rate emitted in the flue gas), the expected storage lifetime might be significantly shorter (2-10 years) than the geothermal doublet lifetime (a few tens of years). However, in a more general strategy of mitigating the CO<sub>2</sub> emissions, it makes sense to conclude that such a storage might work, with a decrease in performance over time, for several tens of years and, at least, as long as the geothermal doublet works.

## 6. First economic assessment on a test-case

The modeling work presented in the previous paragraphs relied on generic datasets assumed to be representative of a carbonated or clastic aquifer of the Paris basin. However, performing economic assessment of the whole CO<sub>2</sub>-DISSOLVED system cannot be achieved on generic cases as real data on energy needs, energy consumption, industrial processes involved, local geological setting, etc., are indispensable to run the models. Consequently, the choice was made to work on the specific test-case of a sugar beet refinery [21]. This choice was justified because data from a previous project which pre-dimensioned a standard CCS facility were available [22].

Transforming sugar beet to ethanol is an interesting process since distillation (for which heat is required) produces almost a pure CO<sub>2</sub> flue gas. Consequently, in this case, the CO<sub>2</sub> separation step is not required, which leads to a significant CAPEX and OPEX offset compared to a case where separation is needed.

The calculation results show that applying the CO<sub>2</sub>-DISSOLVED approach to this biorefinery, with the same operational conditions than in [22], enables significant reductions both on the emissions, between 25 to 60%, and on the energy, between 5 to 30% depending on the assumptions compared to the no CCS case (*i.e.* currently operating conditions). As compared to the standard CCS reference case [23], the CO<sub>2</sub>-DISSOLVED approach can achieve an emission reduction between 15 to 50% while the corresponding non-renewable energy consumption can be reduced by 5 to 30%. CO<sub>2</sub> emission reduction is bigger than the non-renewable energy consumption reduction because the first stages of compression for the CO<sub>2</sub>-DISSOLVED approach require significant compression energy. On the economic standpoint, to reach an emission reduction from 15% to 50% higher, the standard CCS approach would cost between 100% and 350% more than the CO<sub>2</sub>-DISSOLVED approach.

The reader should refer to [21] to get the details of this economic modeling work.

## 7. Preparing the ground for a CO<sub>2</sub>-DISSOLVED demonstration pilot

All the technical and economic results obtained so far confirm the viability of the CO<sub>2</sub>-DISSOLVED concept. However, some complementary work is needed in an objective of preparing a future demonstration pilot of the CO<sub>2</sub>-DISSOLVED concept. Competitively selected and cofounded by the “Geodenergies” consortium (<http://www.geodenergies.com/>), phase II of the CO<sub>2</sub>-DISSOLVED project (“PILOTE CO<sub>2</sub>-DISSOLVED”) was launched in June 2016 for a 18 month duration.

Apart from the technical tasks aiming at deepening our knowledge of the basic geochemical, hydrodynamical and thermal processes related to dissolved CO<sub>2</sub> injection in a doublet, this project scheduled four new tasks:

- (1) Pi-CO<sub>2</sub> bench scale pilot: A first-of-a-kind prototype of the Pi-CO<sub>2</sub> technology is currently being designed and manufactured in the USA by Partnering in innovation, Inc., to be delivered and fully tested at the BRGM’s labs in Orléans (France) by beginning of 2017. This first experimental step is critical in the perspective of getting a fully operational technical system available within the next two or three years.
- (2) Improvement of the global energetic efficiency: with the objective of increasing the temperature of the extracted brine, Enertime (<http://www.enertime.com/en>) who recently joined the consortium, proposed to assess the feasibility and the efficiency of adding heat pump(s) to the surface heat flow facility. The consequences from re-injecting a cooler brine will also be investigated.
- (3) Social acceptability: The CO<sub>2</sub>-DISSOLVED concept is different from the standard CCS approach on many important aspects. However, it is indispensable to assess how a future demonstration pilot relying on this new approach for CO<sub>2</sub> storage could be accepted by the inhabitants of the area pre-selected. For that purpose, the PASSAGES sociology laboratory of the Pau University (France) is now a member of the consortium.
- (4) Legal and regulatory aspects: This is obviously a key question to solve as it is likely, for example, that the legal status of CO<sub>2</sub> as being entirely dissolved in brine is not yet established, at least in France. The IRENEE laboratory from the Nancy University (France) is now involved in the PILOTE CO<sub>2</sub>-DISSOLVED project to work on this important aspect.

However, one of the main preliminary tasks of this new project is to look at all the regional, national, European funding opportunities that could be applied for in order to have better arguments to convince an industrialist, owning a compatible CO<sub>2</sub> emitting plant located on a site with significant geothermal potential, to join our consortium and then prepare together the next phase which is expected to be the pilot construction.

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