CO2-DISSOLVED: a Novel Concept Coupling Geological Storage of Dissolved CO2 and Geothermal Heat Recovery – Part 1: Assessment of the Integration of an Innovative Low-cost, Water- based CO2 Capture Technology

Christophe Kervévan, M.-H. Beddelem, K. O'neil

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CO_2-DISSOLVED: a Novel Concept Coupling Geological Storage of Dissolved CO_2 and Geothermal Heat Recovery – Part 1: Assessment of the Integration of an Innovative Low-cost, Water-based CO_2 Capture Technology

C. Kervévan[a,]*, M.-H. Beddelem[b], K. O'Neil[c]

[a] BRGM, 3 av. Claude Guillemin, BP 36009, 45060 Orléans Cedex 2, France
[b] CFG Services, 3 av. Claude Guillemin, BP 6429, 45064 Orléans Cedex 2, France
[c] Partnering in Innovation, Inc., 14122 Chicora Crossing Blvd, Suite 100, Orlando FL 32828, USA

Abstract

The CO_2-DISSOLVED project evaluates the feasibility of coupling capture and storage in saline aquifer of dissolved CO_2, and geothermal heat recovery. The proposed system basically relies on the integration of a patented water-based in-well CO_2 capture facility (Pi-CO_2) in a classical low-enthalpy geothermal doublet. The results of this preliminary engineering design study demonstrated that (1) the use of composite materials for the wells casings would be compatible with the injection of the corrosive CO_2-rich brine; (2) the housing of the Pi-CO_2 system in a separate large diameter well associated with a dedicated CO_2 injection line integrated in the doublet injection well is the best option for efficient CO_2 capture and dissolution in brine.

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Keywords: CO_2-DISSOLVED project; CO_2 capture and storage; geothermal doublet; composite materials; Pi-CO_2 capture system; drift flux modeling

* Corresponding author. Tel.: +33-2-38-64-35-09; fax: +33-2-38-64-47-97.
E-mail address: c.kervevan@brgm.fr

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1. Introduction

Projects for the geological storage of CO_2 generally plan for its injection under supercritical conditions, thus maximizing the stored quantities that can involve several million tons per year. When no site is available for the secure and durable storage of CO_2 near a major industrial producer, it must be transported to its injection site, entailing very high induced costs.

The CO_2-DISSOLVED project [1], funded by the ANR (French National Research Agency), studies a different option that consists in injecting locally the emitted CO_2 as being previously dissolved in brine. Since it is intended to be a local solution, the costs related to CO_2 transport would then be dramatically reduced, provided that the local underground geology is favorable. The infrastructure for this system is based on a set consisting of a production well and an injection well (just like in classical geothermal doublet facilities), which allows pumping the brine from the reservoir and then re-injecting it after having been saturated in dissolved CO_2. With this process, it will be possible to overcome the inherent problems of the "classic" CCS approach (injection of supercritical CO_2), such as pressure buildup and migration of the initially present brine, or the risk that CO_2 migrate into overlying geological formations as no light gaseous or supercritical phase is involved. In addition, it is planned to recover the heat from the warm brine pumped at the production well for local use by the industrial CO_2 emitter for its own process or heating needs, and possibly for feeding a heating network close to the storage facility.

One of the technological issues to address for this concept to be applicable, assuming that CO_2 is previously captured in the emitted flue gas, relies on our capacity to propose an efficient CO_2 dissolution system. On this key aspect, CO_2-DISSOLVED brings another novel approach by offering to assess the feasibility of integrating a patented capture and dissolution technology (Pi-CO_2) as part of the integrated CO_2 capture, storage, and geothermal heating system. After a brief general presentation of the project, this paper focuses on (1) the main features of the Partnering In Innovation, Inc. (Pi-Innovation) Pi-CO_2 system [2] that is proposed to be used as the CO_2 capture and dissolution facility, and (2) the assessment of this system performance under the specific constraints of the CO_2-DISSOLVED context.

2. The CO_2-DISSOLVED project

The objective of the CO2-DISSOLVED project (CO_2 Dependable Injection and Storage System Optimized for Local Valorization of the geothermal Energy Delivered) is to assess the technical-economic feasibility of a novel Carbon Capture and Storage (CCS) concept integrating aqueous dissolution of CO_2 and injection via a doublet system (*i.e.* one injection well and one production well targeting the same deep saline aquifer), an innovative post-combustion CO_2 capture technology, and geothermal energy recovery (Fig. 1). Compared to the use of a supercritical CO_2 phase, this approach offers substantial benefits in terms of storage safety, due to lower brine displacement and no pressure build-up risk, lower or no CO_2 escape risk, and the potential for more rapid mineralization.

However, the solubility of CO_2 in brine will be a physical limiting factor to the amount of CO_2 that can be injected. Consequently, and as another contributing novel factor, this project targets specifically low-to-medium range industrial CO_2-emitters (ca. 10-150 kt/yr), that could be compatible with a single doublet installation. Since it is intended to be a local solution, the costs related to CO_2 transport would then be dramatically reduced, provided that the local underground geology is favorable.

Even though this project basically is a feasibility study using engineering methods, such as dimensioning calculations and numerical simulations, ambitious research work is carried out as well:

- The viability of the concept first requires the availability of a potential market, *i.e.* at least the presence of both an industrial CO_2 emitter, compatible in terms of the CO_2 tonnage emitted, and a favorable local geothermal potential. An inventory of the potentially compatible sites available in France, Germany, and the U.S.A., realized in the framework of this project, demonstrated the existence of a real potential of application of the CO_2-DISSOLVED concept to many low-to-medium industrial emitters in these countries [3].

- Preliminary hydrodynamic modeling work of the doublet functioning evidenced that dissolved CO_2 will inevitably reach the production well after a while (*ca*. 2 to 15 years, depending on the doublet operating parameters), at low-medium concentrations. However, this CO_2 production will not generate any gaseous emission to the atmosphere, since CO_2 remains dissolved in the brine circulating in a closed loop between the injection and the production wells. In addition to this, the mass balance calculations show that a significant fraction of the injected CO_2 actually remains stored in the aquifer, corroborating the viability of the concept from a hydrodynamic point of view [4].

- The brine acidified by dissolved CO_2 will be chemically reactive with the mineral phases of the aquifer as soon as it exits the injection well, contrary to the supercritical approach where the reactive acid front follows the extension of the CO_2 "bubble". Specific work will focus on the near-well area, based on new experimental and modeling approaches. An experimental laboratory installation (MIRAGES-2) was specifically designed in the framework of this project and has recently been used for a first series of experiments [5].

- The usual methods for site monitoring and risk assessment must be reviewed in the light of new constraints inherent in this original approach proposed. Innovating solutions for geochemical and geophysical monitoring are currently evaluated and tested, both in the field and in the laboratory. A new methodology for risk analysis is being specifically designed and will be applied on selected test-cases, in accordance with the modeled and observed properties of the system as a whole, *i.e.* capture, injection, CO_2 storage, and heat recovery.

- Earlier economic models of CCS will be rendered obsolete by the local application of an associated capture-storage and geothermal-heat-production technology to small CO_2 emitters. This will require development and validation of new economic models that will then be applied to two test cases in France and Germany.

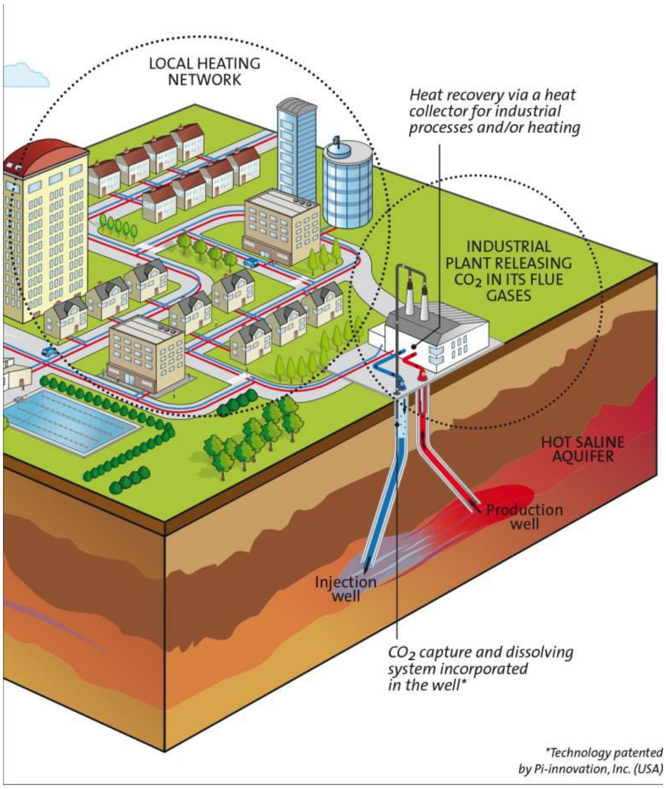

Fig. 1. Schematic view of the CO_2-DISSOLVED concept

Finally, the approach proposed in the CO_2-DISSOLVED project, dedicated to low-medium range CO_2 emitters, is complementary to the supercritical approach for which it is generally planned to inject several million tons of CO_2 per year. Moreover, the potential for energy and/or revenue generation through geothermal heat recovery constitutes an interesting way of valorization of the CCS operations, demonstrating that an actual synergy between CO_2 storage and geothermal activities may exist.

3. Key features of the geothermal doublet

As mentioned previously, the CO_2-DISSOLVED concept basically relies on the use of a low-enthalpy geothermal doublet in which we plan to dissolve CO_2 in the extracted brine at the outlet of the surface heat exchanger system, prior to its reinjection into the aquifer.

In order to provide relevant data for the pre-dimensioning calculations required for the engineering design of the whole system and for assessing both the operational and economic parameters, it was necessary to work with data from actual geothermal operations. CFG Services, partner of the CO_2-DISSOLVED project, carries out engineering, control-builder, exploitation, advice, and expertise services in the field of heat and power geothermal applications. In particular, they are mainly involved in several of the currently exploited low-enthalpy geothermal sites of the Dogger aquifer of the Paris basin (France). That is why we decided to use these available data for this project. Moreover, in a next step, it is planned to study much more in details a specific test-case which will be located near a low-tonnage industrial CO_2 emitter of the Paris basin.

The Dogger aquifer of the Paris Basin is the most exploited geothermal reservoir for heating purposes. In the Paris area, its oolitic limestone found at a depth between 1600 to 1800 meters holds hot saline brines at a temperature ranging between 60 to 80°C. The transmissivity of the reservoir (10 to 110 D.m) allows extracting the geothermal water at a flow rate up to 300-350 m^3/h.

Because of the salinity of the brine (20 g/l in average) and in order to limit drawdown and hydraulic impact on the geothermal facility (moreover in areas with a high density of Dogger geothermal facility aiming at the Dogger), all the pumped geothermal brine is re-injected in the same reservoir after calories extraction. It has to be noticed here that this closed-loop functioning with re-injection in the same aquifer is a pre-requisite for the CO_2-DISSOLVED concept to be viable, since it guarantees (1) no water feeding and/or disposal problems, and no pressure buildup effect in the aquifer. The major drawback of this strategy is the expected induced hydrodynamic migration of part of the injected CO_2 toward the production well that we also have to account for in the design of the doublet system [4].

Table 1. Key operational parameters of a typical geothermal doublet of the Paris basin

Typical operating parameter	Dimension
Casing diameter	9’’5/8
Open hole diameter	8’’1/2
Well deviation angle	30-40°
Distance between wells (bottom hole)	1500 m
Distance between well heads (ground level)	10 m
Maximum injection/production flow rate	300-350 m^3/h

The geothermal device that we defined here is a doublet of drillings that spread apart from a single platform (Fig. 2). The spreading apart of the impact points at the reservoir is worked out, through numerical modeling, in order to guarantee proper operation of the new device over a minimum lifetime of 30 years without any thermal penetration at the production well (production temperature drop lower than 0.5°C), taking into account the relative uncertainty of the reservoir productivity. The main exploitation features related to this doublet are given in Table 1.

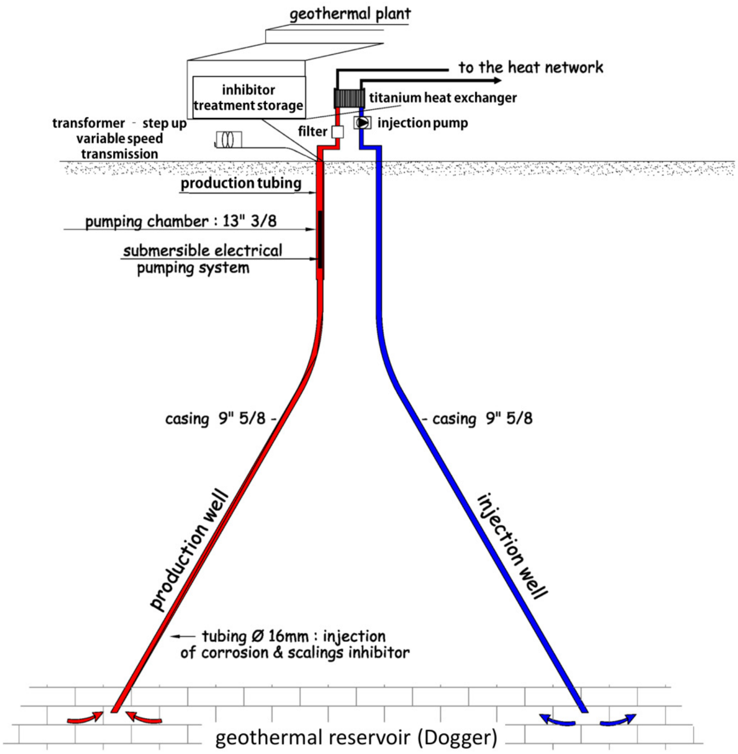

Fig. 2. Main features of a typical low-enthalpy geothermal doublet of the Dogger aquifer (Paris basin, France)

4. The Pi-CO_2 water-based capture technology

Pi-Innovation's patented aqueous post-combustion capture system (Pi-CO_2) was initially designed for applications to Enhanced Oil Recovery operations and Enhanced Geothermal Systems (where CO_2 can be used as a working fluid). It uses well-known principles and standard components in a novel configuration. The design includes an innovative integration of compression and energy recovery steps that greatly reduce parasitic energy costs. Preliminary coupled thermodynamic and thermal-hydraulic mass transfer modeling results fully support proof of concept [2].

In a brief overview (see Fig. 3), the Pi-CO2 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 (or suspended in deep water) 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.

The flue gas is injected in the MTS at depth in the deep water column. The gases (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). The non-CO_2 ex-solved gases are sequentially removed in the return line to produce nearly pure 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).

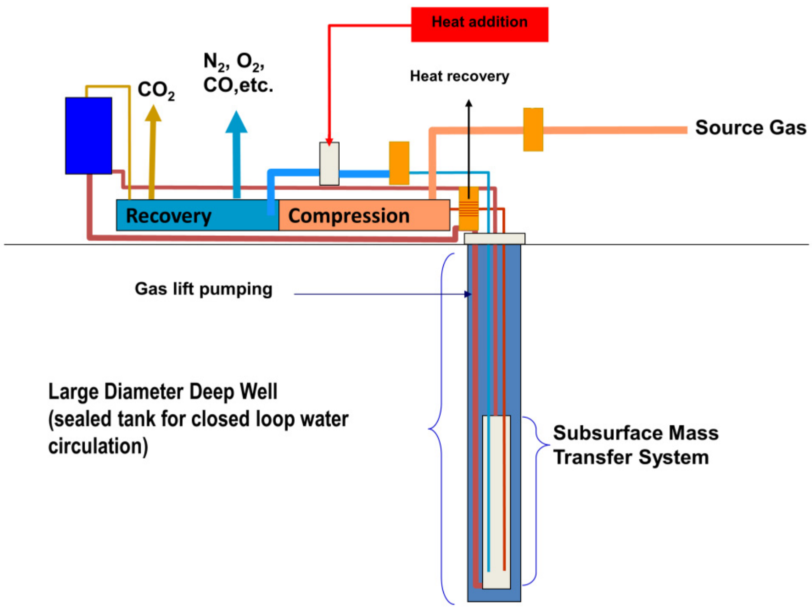

Fig. 3. Simplified system sketch of the Pi-CO2 water-based in-well capture technology

This aqueous process is easily scalable using commercially available equipment for the aboveground components. Analysis of the gas-lift pumping capacity indicates that the volumetric flow of off-gassing CO_2 is many times greater than that required to circulate the large volumes of water needed to achieve this production scale, thus confirming gas lift pump efficiencies and the assumption that additional energy and mechanical pumping are not needed for circulation.

The non-degradable nature of the physical solvent provides an opportunity to capture CO_2 from flue gases associated with coal combustion, refining, smelting, and cement manufacture. Desulfurization common to existing flue gas sources is required to limit the contact of sulfur with the water. Compression of the flue gas prior to entry into the mass transfer system offers the opportunity to condense reduced mercury that would otherwise be released to the atmosphere. This offers a unique differentiator and additional positive cost-benefit for coal-fired power plant operations.

5. Integration of the Pi-CO_2 technology in the CO_2-DISSOLVED system

As presented previously, the CO_2-DISSOLVED concept basically relies on the coupling of a geothermal doublet and an innovative CO_2 capture/dissolution facility. In this paragraph, we present the results acquired so far in the project about the integration of the Pi-CO2 system in the geothermal doublet. The main points that are addressed in the next paragraphs are (1) the choice of the appropriate material for the wells casings because of the injection of corrosive CO_2-rich brine, (2) the selection of the best option for integrating the Pi-CO2 system, and (3) the assessment of the actual CO_2 dissolution rate capacity of the system for the selected configuration with typical doublet operational parameters.

5.1. Corrosion issues and operational constraints

Low enthalpy geothermal energy facilities (cf. Fig. 2) traditionally rely on the use of mild steel wells to transfer the geothermal brine from the reservoir to the surface installations and back into the reservoir. Mild steel is generally the material of choice because it provides the necessary mechanical properties at the lowest cost. In the anoxic environment of geothermal brines, the corrosion rate of mild steel would be relatively acceptable. However, as corrosion proceeds, iron sulphide is created when dissolved iron encounters the natural hydrogen sulphide present in the exploited brine of the Dogger aquifer. Iron sulphide (FeS, FeS_2) gradually covers the tubing and creates an electrochemical cell that accelerates further the corrosion phenomena. The FeS / FeS_2 deposit subsequently houses Sulphate-Reducing Bacteria (SRB), which convert sulphate into hydrogen sulphide, lowering the pH and accelerating further the corrosion phenomena. In order to reduce the rate of progression of this cycle, expensive corrosion inhibitors must be used, to reduce the adhesion of deposits on the surface of the metal, and cover it with a passive film that shields it from the aggressive environment of the brine.

In the envisaged CO_2-DISSOLVED system, massive CO_2 dissolution is expected to decrease pH even further and then significantly increase the corrosion rate of any mild steel well. Corrosion-Resistant Alloys (CRAs) would be valid candidates in the replacement of mild steel as the material of choice. In particular, their low rate of general corrosion strongly reduces the cycle: Corrosion => FeS / FeS_2 deposit => bacterial colonization => increase in H_2S content => accelerated rate of 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_2-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 geothermal doublet in the Dogger aquifer, 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).

At this stage of the project, considering (1) their expected resistance to corrosion, (2) their appropriate mechanical properties, and (3) the reasonable initial overcharge with respect to mild steel, composite materials remain our preferred option for the design of the CO_2-DISSOLVED doublet facility.

5.2. Best option for integrating the Pi-CO_2 system

As suggested on Fig. 1, we initially thought about integrating the whole Pi-CO_2 mass transfer system (MTS) within the injection well of the doublet. This option was beforehand very seducing since the CO_2-DISSOLVED concept only requires the CO_2 capture and dissolution steps, with no need for further degasing at the surface. As a consequence, we first planned to mainly rely on the MTS to both capture the CO_2 from the feed gas by dissolving it in the circulating brine, and then inject the 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 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, as indicated on Fig. 2 and Table 1, the well diameters used are relatively small (about 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.

The option of using a large-diameter well housing the Pi-CO_2 system and dedicated to the CO_2 capture operations was then considered. With this solution, this third well would be designed according to the actual needs in terms of

CO_2 separation and injection, depending on the targeted flow rate and on the flue gas composition. The captured CO_2 flux would then have to be piped to the injection well as a gas phase. Consequently, a supplementary step is required for achieving CO_2 dissolution in brine. For that purpose, Pi-Innovation proposed to use a very simple technical solution based on a bubbler being positioned at the appropriate depth in the injection well according to the CO_2 mass rate demand. This would simply necessitate integrating a small diameter tube ended by a bubbler system designed for a prescribed bubble diameter in order to optimize the in-well CO_2 dissolution process. CFG Services confirmed that this system could be easily fitted in a standard geothermal injection well after a slight modification of the well head.

However, in a test-case involving a bioethanol production plant we just started to work on, the emitted flue gas is almost pure CO_2. In this context, the gas separation step is not necessary, so that we can avoid using the third well including the Pi-CO_2 system. This specific context could easily be generalized to other industries emitting smokes with high CO_2 content (ca. > 80%),

Consequently and as a general preliminary conclusion on the design of the capture, separation, and dissolution units in the CO2-DISSOLVED system, two cases have to be envisaged:

- (1) When the flue gas has a high CO_2 grade (ca. > 80%), no CO_2 separation unit is necessary. Gaseous CO_2 could then be directly co-injected with water into the injection well, using a specific small diameter in-well CO_2 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_2 concentration in the emitted smokes is lower than 80%, which corresponds to the most general cases, a gas separation unit is necessary. A specific large diameter well housing the Pi-CO_2 system would then be built, in order to recover almost pure gaseous CO_2 which will be co-injected in the injection well in a similar manner to what is done in case 1).

5.3. First CO_2 mass transfer modeling results

In order to assess the achievable dissolved CO_2 mass transfer rates relying on the previously selected co-injection option, Pi-Innovation performed complex fluid mechanics calculations using a thermal-hydraulic drift flux model.

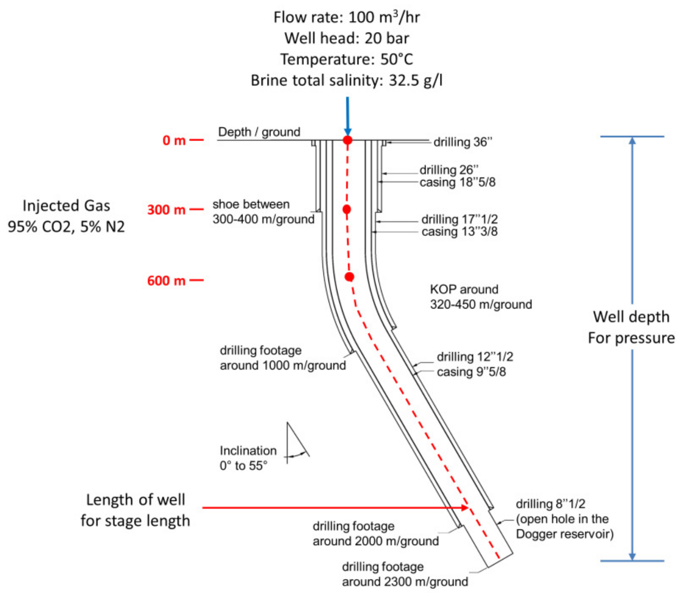

Fig. 4. Injection operational parameters and well geometry considered in the modeled test-case

The drift flux model code is based upon empirical data, with inputs including: mass of the water and gas, temperature and salinity of the water, Henry's Gas Law of solubility at various pressures, water velocity, bubble fraction and diameter, and diameter and length of the absorber. It has to be noticed that the bubbler location and the length of the remaining portion of the well determine the absorber length used for the calculations. The preliminary tests of validation of the numerical code used showed a good agreement with the empirical data available.

Mass transfer modeling runs were made to mimic CO_2 solubilization into brine injected down a geothermal doublet with three possible locations of the bubbler in the well as illustrated on Fig. 4: 1) at the well head; 2) at 1000 feet (~305m) of depth; or 3) at 2000 feet (~610m) of depth. The well depth and length of the remaining portion of the well were considered for hydrostatic pressure and absorber length in each modeling run, respectively. In order to evaluate minimum achievable CO_2 dissolution rates, we voluntarily used very conservative values for flow rate (much lower than typical flow rates of up to 350 m^3/hr observed in current geothermal doublets of the Paris basin), salinity (higher than the average salinity around 20 g/l in the Dogger aquifer), and temperature (higher than the observed injection temperature generally closer to 40°C). Model runs used a well head pressure at 20 bar; flow rates at approximately 100 m^3/hr, brine temperatures at approximately 50°C; and a brine salinity of 32.5 g/l.

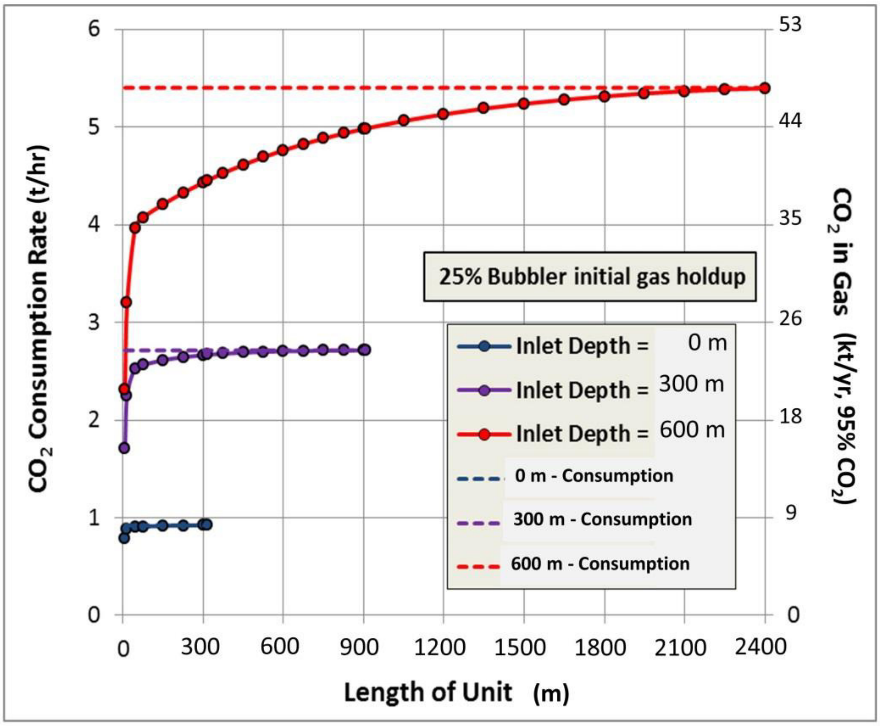

Fig. 5. Calculated soluble CO_2 in gas rate (solid lines) as a function of the length of absorption unit below the bubbler location in the well, assuming a 25% bubble fraction. Comparison with the achievable CO_2 consumption rate (dashed lines) at the same bubbler locations.

Fig. 5 illustrates the absorber (*i.e*. the well length below the bubbler in this case) effects in metric tons of CO_2 consumption per hour (or kt/yr on the right side scale) for the three bubbler locations studied. The bubble fraction injected was set at 25% with a feed gas consisting of 95% CO_2 and 5% N_2. The consumption rates are calculated for maximum CO_2 without bubbles being present at the well bottom, to guarantee a complete dissolution before the injected CO_2-brine mixing reaches the target aquifer, which is a key condition to fulfill in order for the CO_2-DISSOLVED concept to be viable. This graph clearly shows that a longer well would result in higher tonnages consumed. It must also be noticed that the greater the depth of CO_2 injection, the higher the consumption, due to increased solubility. It is also possible to increase CO_2 rates by increasing the bubble fraction and/or the injection flow rate. However, injection of CO_2 at high hydrostatic pressures results in higher compression energy demand.

It can then be concluded, based on the results of this preliminary modeling work, that CO_2 dissolution rates of a few tens of kt/yr, consistent with the low-medium industrial CO_2 emitters targeted in this project, can be easily achieved. Considering that very conservative well characterization data were used in these simulations, it can be anticipated that higher CO_2 dissolution rates (up to 100 kt/yr) might be achieved for higher injection flow rates (200-300 m^3/h), lower salinity (20 g/l or even less), lower injection temperature (40°C or even less), higher bubble fraction. This will have to be confirmed by a new series of simulations relying on more favorable datasets.

6. Conclusion

This preliminary engineering design study aimed at assessing the technical solutions available for optimizing the CO_2 capture and injection processes to be associated with a classical geothermal two-well facility (doublet). The proposed technology for capture relies on the Pi-CO_2 system, a low-cost, water-based patented technology provided by Pi-Innovation. Our first conclusions on the design of the capture, separation, and dissolution units in the CO_2-DISSOLVED system are that:

- Considering (1) their expected resistance to corrosion, (2) their appropriate mechanical properties, and (3) the reasonable initial overcharge with respect to mild steel, composite materials are our preferred option for the design of the CO_2-DISSOLVED doublet facility.

- When the flue gas has a high CO_2 grade (ca. > 80%), no CO_2 separation unit is necessary. Gaseous CO_2 could then be directly co-injected with water into the doublet injection well, using a specific small diameter in-well CO_2 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.

- When the CO_2 concentration in the emitted smokes is lower than 80%, a gas separation unit is necessary. A specific large diameter well housing the Pi-CO_2 system should then be built, in order to recover almost pure gaseous CO_2 which will be co-injected in the injection well using the abovementioned injection line/bubbler technology.

In addition, CFD modeling results carried out, on purpose, using very conservative operational parameters, demonstrated the viability of the injection line/bubbler system for achieving expected CO_2 injection mass rates of a few tens of kilotons per year, with a complete dissolution of the bubbles before the open-hole zone. Using parameter values closer to the actual data of current geothermal operations at the Dogger aquifer (Paris basin), higher dissolved CO_2 injection flow rates can reasonably be expected. However, further modeling work will have to confirm this assumption.

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