PROCEEDINGS, 41st Workshop on Geothermal Reservoir Engineering
Stanford University, Stanford, California, February 22-24, 2016
SGP-TR-209

On the Versatility of Rheoreversible, Stimuli-responsive Hydraulic-Fracturing Fluids for Enhanced Geothermal Systems: Effect of Reservoir pH

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Keywords: fracturing fluid, rheoreversible, CO_2, pH, polyallylamine, viscosity

ABSTRACT

The primary challenge for the feasibility of enhanced geothermal systems (EGS) is to cost-effectively create high-permeability reservoirs inside deep crystalline bedrock. Although fracturing fluids are commonly used for oil/gas, standard fracturing methods are not developed or proven for EGS temperatures and pressures. Furthermore, the environmental impacts of currently used fracturing methods are only recently being determined. These authors recently reported an environmentally benign, CO2-activated, rheoreversible fracturing fluid that enhances permeability through fracturing due to in situ volume expansion and gel formation. The potential of this novel fracturing fluid is evaluated in this work towards its application at geothermal sites under different pH conditions. Laboratory-scale fracturing experiments using Coso Geothermal rock cores under different pH environments were performed followed by X-ray microtomography characterization. The results demonstrate that CO2-reactive aqueous solutions of environmentally amenable polyallylamine (PAA) consistently and reproducibly creates/propagates fracture networks through highly impermeable crystalline rock from Coso EGS sites at considerably lower effective stress as compared to conventional fracturing fluids. In addition, permeability was significantly enhanced in a wide range of formation-water pH values. This effective, and environmentally-friendly fracturing fluid technology represents a potential alternative to conventional fracturing fluids.

1. INTRODUCTION

Enhanced geothermal systems (EGS) are a promising alternative for energy production for the United States and worldwide. [Blackwell (2006), Duffield (2003)] With its negligible CO_2 emissions, EGS can help to mitigate climate change. However, one of the primarily challenges for the application of enhanced geothermal systems is to cost-effective and safely create high-permeability reservoirs inside deep crystalline settings. It is well-recognized the lack of suitable technologies to cost-effectively create high-permeability reservoirs within the 900–4000 m depth in a temperature range of 150 - 400 ºC. [Duffield (2003), Pruess (2006)] Moreover, to our knowledge, no prior EGS project has sustained production at rates greater than 50% of what is needed for economic viability.

A key driver for the development of economic EGS is the continuing advances in hydraulic fracturing techniques and horizontal drilling implemented on unconventional oil and gas exploitation. [Kargbo (2010), Kerr (2010), Tester (2006)] Hydraulic fracturing processes utilize brute force, hydraulic pressure, from millions of gallons of water per well that are pumped down the wellbore at high pressures to create a network of cracks in the source rock. Among the industry standard practices, surfactants and macropolymers (e.g., sodium dodecyl sulfate [SDS] or xanthan gum) are employed to reach fracture pressures. However, some of the fluid components used in hydraulic fracturing for recovery of tight oil and gas cannot be employed for EGS due to the high subsurface temperatures. The main limitation is the fact that macropolymers developed to modify fluid rheology in oil/gas standard hydraulic fracturing degrade at geothermal temperatures above 150°C. Second, these macropolymers are difficult to remove from the formation after fracture creation/propagation. The so called "fracture skin", which is the residue of injected polymers and drilling mud not removed during fracture cleanup, limits the efficiency of hydraulic fracturing because it decreases flow rates and heat transfer in EGS. In addition, the most challenging issue is the large volume of wastewater generated, which may include radioactive and toxic elements extracted at EGS temperatures (150 °C – 380 °C), and that needs to be treated and disposed of. Finally but not least, the large volumes and chemical content of hydraulic fracturing wastewater have stocked public fears of contamination of soil and drinking water resources generating both environmental and economic concerns. [Tester (2006)] Thus, to enhance the economic viability of EGS and decrease/minimize the environmental impact of fracturing processes, the development of more effective and environment-friendly fracturing fluids is imperative.

Our group has developed an environmentally benign, CO_2-activated, rheoreversible fracturing fluid that undergoes large volume expansions at EGS-relevant pressures and temperatures (Fig. 1). [Fernandez (2015)] This volume expansion in confined environments significantly enhances rock permeability at measured effective stress up to an order of magnitude lower than current technology. In this work we study the effect that reservoir pH will have on the rheology/viscosity of the fluid at different pressures, temperatures and shear rates and the feasibility of this stimulation technology to create fracture networks in rock samples obtained from the Coso EGS site. This novel hydraulic fracturing fluid could dramatically reduce the environmental impact and water usage of fracturing practices, potentially making geothermal energy production cost-effective and cleaner.

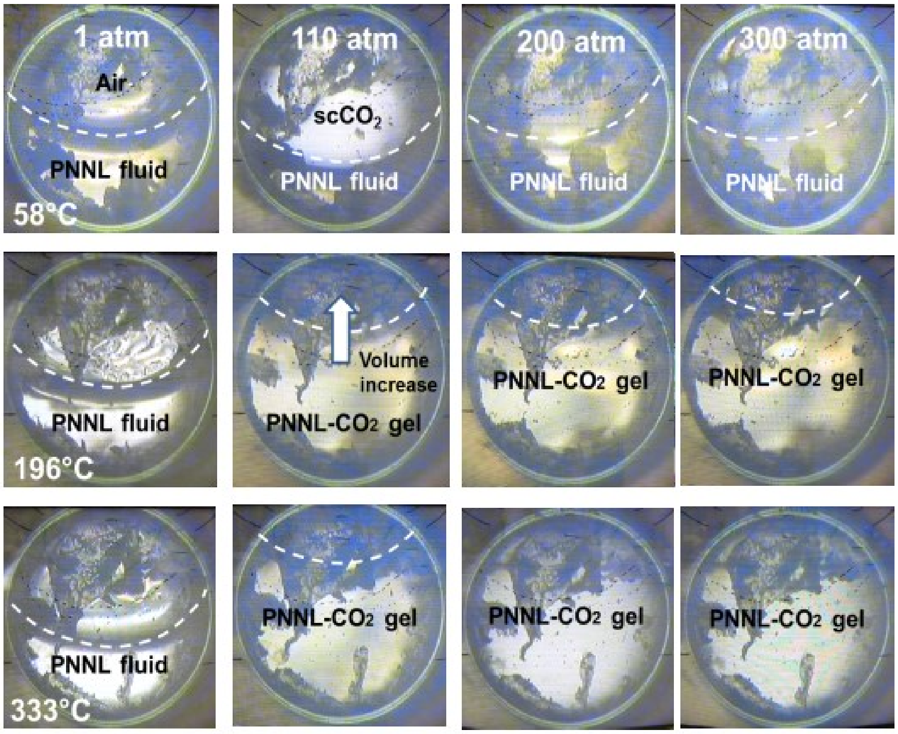


Fig. 1. Volume changes of PAA solution reacted with CO_2 as a function of temperature (58–333°C) and CO_2 pressure (0–300 atm). CO_2 pressure was increased from 0 to 300 atm. The dashed lines indicate the boundary between PAA fluid and supercritical CO_2 (after CO_2 injection) or air/water vapor (before CO_2 injection). The window is partially covered by some residual PAA [Fernandez (2015)].

2. MATERIALS AND METHODS

2.1 Chemicals and rock samples

Polyallylamine MW= 17,000 Da (PAA) solution was obtained from Sigma Aldrich. This polymer solution was diluted 20 times with either deionized water (DIW) or pH= 7 and pH= 10 buffer solution (Sigma-Aldrich). The diluted solutions were used for hydraulic fracturing experiments as described below. Rock cores obtained from Coso geothermal field in California at depth of ~490 m were utilized for fracturing experiments. Coso Rock samples consisted of Mesozoic diorite metamorphosed to greenschist facies. These raw sample materials were cut into small cylindrical rock cores (1.59 cm diameter and 5.08 cm length) and a centred hole (0.32 cm diameter and 2.54 cm deep) was drilled from the top of the cylinder. The top and bottom of the cores were polished to ensure the sealing with an O-ring (see next section).

2.2 Viscosity measurements

The rheology properties of the 1 wt% PAA solution and the 1 wt% sodium dodecyl sulfate (SDS) solution (control experiment), and DIW (second control experiment) at constant temperature (300°C) and incremental CO_2 pressure (up to 500 atm) were determined using an Anton Paar Physica MCR101 rheometer equipped with a temperature-controlled pressure cell (C-ETD 300/PR1000, temperature and pressure limits 300°C and 1000 atm). In addition, viscosity measurement s were performed in 1 wt% solutions of PAA in buffers pH=7 and pH=10 and in their corresponding control experiments (both buffer solutions without PAA).

Before rheology measurement, the measuring system and the pressure cell were coupled and the rheometer motor was adjusted. A bearing/air check was conducted to evaluate the conditions and performances of the bearings in the head of the pressure cell. For the measurement, the pressure cell (vol. = 24 mL) was first preheated to 90°C. Water or a chemical solution (18 mL) was then injected into the cell through a port and the temperature increased to 300°C. The cell was sealed and CO_2 was injected into the cell using a syringe pump in steps of about 20 atm to a final pressure of 500 atm with minimum variation in temperature. The rheology measurement at a fixed shear rate (100 s-1) was started immediately after CO_2 was introduced. Finally, a measurement of viscosity as a function of shear rate from 3 to 120 s-1 was conducted at 300°C and 500 atm on all fluid mixtures.

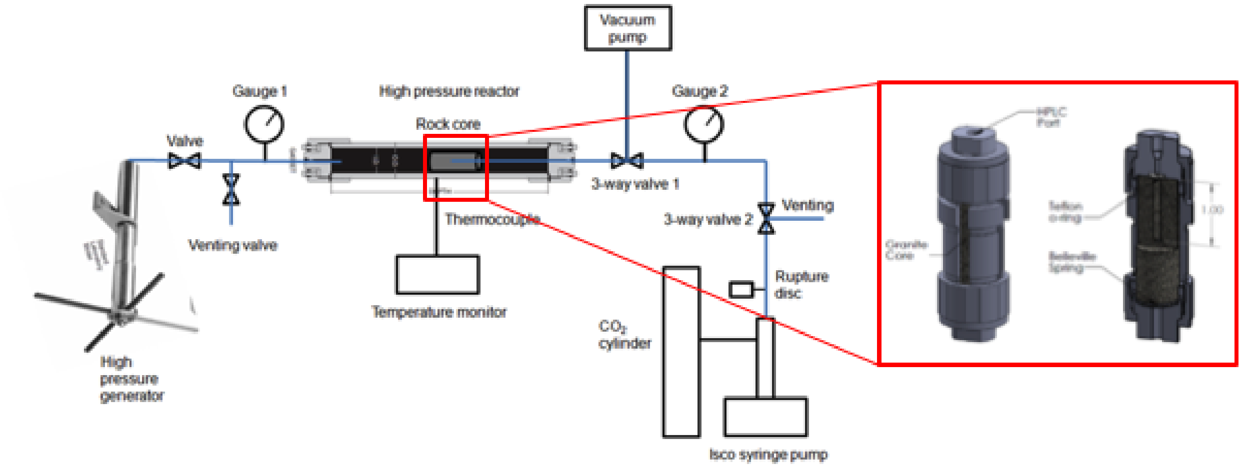


Fig. 2. Schematic diagram for the hydraulic fracturing experimental setup. Inset: cradle designed to seal the rock samples from the applied confining pressure and introduce the fracturing fluid.

2.3 Hydraulic fracturing experiments

Laboratory-scale hydraulic fracturing experiments were performed using the rock cores prepared according to the previous section. A schematic diagram for the hydraulic fracturing experimental setup is shown in Fig. 2. Each rock core was placed in a state-of-the-art cradle designed to expose the entire sample to the desired confining pressure while isolating the central hole drilled to introduce the fracturing fluid (Fig. 2, inset).

Stainless steel tubing (0.16 cm OD) was introduced 0.5 cm into the hole leaving an internal dead volume of ~200 µL in the rock core. Teflon O-ring was used on the top of the cradle to ensure sealing of the connections and avoid any communication between the external fluid (water) used to apply the desired confining pressure and the internal dead volume in the rock core during the course of the fracturing experiment. The rock core in the cradle was then connected to the head of a high pressure vessel (High Pressure Equipment Company, Erie, PA), and the cradle was introduced to the pressure vessel. A 3-way valve was used to deliver polymer solution and CO2. The temperature of the reactor was controlled with heating tape, a thermocouple, and a temperature controller. After the target temperature was reached, water was injected into the pressure vessel using a syringe pump to increase confining pressure. The confining pressure was first increased to either 68 atm, immediately followed by introduction of 200 µL of 1 wt% PAA solution and 68 atm of CO2 inside the core sample via the three-way valve. Then, the confining pressure and rock core pressure were increased in 3.4 atm (50 psi) intervals until the target pressure was achieved. Subsequently, the rock core internal pressure was gradually increased in intervals of 1 atm with the fracturing fluid while maintaining the confining pressure constant, until an increase in the confining pressure was observed. This pressure equilibration indicates communication between the internal and external fluids as a result of fracture formation/propagation. The pressure difference between the applied fracturing fluid pressure required for rock core fracturing and the confining pressure is defined as “effective pressure” or “effective stress.” After pressure equalization was observed, the system was cooled to room temperature and the pressure was released before the pressure vessel was opened and the rock core retrieved. For comparison, control fracturing experiments using DIW/CO2, pH= 7 buffer/CO2 and pH= 10 buffer/CO2 solution were also performed at identical experimental conditions for comparison purposes. The rock cores retrieved from the high pressure vessel after fracturing experiments were subjected to a series of tests to examine the formation and distribution of fractures as described below. Hydraulic fracturing experiments were performed with 1 wt% PAA solution to evaluate the fracturing fluid performance, in terms of permeability enhancement) in different pH environments at a typical EGS temperature (300 ºC) and confining pressure (330 atm).

2.4 Post experiment fracture connectivity measurements

Gas leaking test (gas bubbling experiment) was performed for each core after fracturing experiment to verify the presence of fractures connected to the external surface of the sample. This experiment was conducted by injecting N2 gas at 6-10 atm into the center hole of the rock core. The core was placed inside water, so the fracture opening to the external surface of the rock cores can be identified by the appearance of gas bubbles. The pressure was maintained at low levels (6-10 atm) to minimize the potential creation of new fractures during the gas leaking tests.

2.5 Post experiment permeability measurement

Pre-experiment rock intrinsic permeability values were below the lower limit of measurement. However, the intrinsic permeability of the fractures formed after fracturing experiments were calculated from measurements of tracer travel time through fractured core within the high pressure vessel using 1 M copper acetate solution. The blue coloured solution was injected into the fractured rock cores using a water-filled syringe pump to allow the application of a constant pressure gradient of 6-8 atm. The time when the blue coloured solution was observed at the region where gas leakage occurred was used to estimate the seepage velocity, which in turn was used to calculate the permeability using Darcy’s law. Since we do not know explicitly the exact fracture pathway in the rock core, we assumed that the pathways of all the fractures were straight lines perpendicular to the central hole and extend directly from the central hole to the external surface (i.e., 0.635 cm). It is noteworthy that more than likely this assumption underestimates the actual fracture pathway. Therefore, the measured permeability represents the lower limit of the intrinsic permeability.

2.6 X-ray microtomography (XMT) analysis

XMT scanning was performed at 110 keV and 190 µA for optimum image quality and contrast. The samples were rotated continuously during the scans with momentary stops to collect each projection (shuttling mode) and minimize ring artifacts. A total of 3142 projections were collected over 360 degrees with 0.5 second exposure time and 4 frames per projection. Image voxel size varied from 25–30 µm depending on specimen dimensions. The images were reconstructed to obtain three-dimensional datasets using CT Pro 3D (Metris XT 2.2, Nikon Metrology, UK). For rock cores after fracturing experiment, pressurized scans were conducted using a cell containing the sample connected to a nitrogen tank regulated to 8 atm (similar to pressure gradient of permeability measurements). The cell was made of polyether ether ketone (PEEK) to minimize the adsorption of x-ray.

3. RESULTS AND DISCUSSIONS

The formation water chemistry and pH of brine solutions varies from one geothermal site to another one with the majority of geothermal sites in U.S. reporting pH values in the range of 6-10 (a few sites report pHs as low as 4). [Clark (2010)]

3.1 Viscosity behavior of PAA-CO2 mixtures

Viscosity analysis as a function of CO2 pressure at constant temperature (300 ºC) was performed on 1wt% PAA solutions prepared in DIW, in a pH=7 buffer and in a pH=10 buffer and compared to the corresponding control experiments as described in the experimental section (data not shown). Compared to the control fluid systems, aqueous PAA-CO2 fluids at different pH values appeared to have the highest viscosity at any given pressures under the tested conditions. Viscosity of the DIW-CO2, pH=7 buffer-CO2, pH=10 buffer-CO2 and 1 wt% SDS-CO2 mixtures were in the order of 1-4 cP. Viscosity was measured at a shear rate of 100 s-1. The viscosity values were relatively constant independently of CO2 pressure for all control experiments. The viscosity of the PAA-CO2 system was in the range of 12-18 cP in DIW, pH=7 buffer and pH=10 buffer after CO2 pressure reached 500 atm. These viscosity values are significantly higher than the obtained with the corresponding control experiments indicating that, in the presence of PAA, the formation of a viscous fluid/gel triggered by CO2 crosslinking reaction takes place. It is important to mention that all the starting solutions (with and without PAA) had similar viscosity values at 300 °C previous to injecting CO2. These results are of great significance and in agreement with the observed volume expansions of PAA-CO2.

3.2 Laboratory-scale stimulation experiments

As mentioned previously, the formation water chemistry and pH of brine solutions at geothermal sites in U.S. have been reported to range between 6 and 10. Therefore, to evaluate if this newly developed fracturing fluids can enhance permeability of crystalline highly impermeable rock materials at different pH conditions, six experiments were conducted with rock cores from Coso geothermal field at 300 °C and 333 atm in DIW, buffer solution pH= 7 and buffer solution pH=10 with and without PAA 1 wt% (see Table I).

Rock core Coso 1-1 (Experiment #1) was fractured applying an effective pressure of only 17 atm with PAA-CO2 fracturing fluid system, whereas Coso 1-2 (control experiment with DIW-CO2) was not fractured even when the internal pressure was 170 atm higher than the confining pressure (note that at this effective pressure, the pressure of CO2 was actually near the upper limit of the ISCO pump). Gas leaking test for Coso 1-1 showed that N2 bubbles appeared immediately and profusely and occurred within the top 2.54 cm of the rock core, consistent with the location of the center hole (2.54 cm depth) where the hydraulic fracturing fluid was injected. XMT analysis showed no presence of preexisting fractures on Coso 1-1 before the fracturing experiment, while after fracturing experiment fractures can be clearly seen and extend approximately 2 cm longitudinally (Fig. 3). The experimental permeability of Coso-1-1 after fracturing was determined to be approximately 8.8 mD, which is comparable to the typical permeability of sandstone reservoir rocks.[Pape (1999)] It is noteworthy that, as we discussed in Section 2.4, the actual intrinsic permeability could be higher than the experimental permeability reported in Table 1 due to the assumptions employed for its calculation. When the experimental permeability for Coso 1-2 was determined, no liquid was observed on the external surface of Coso 1-2 after applying 10 atm pressure on the center hole for 5 min. Based on this result, we estimate that the permeability of Coso-1-2 after fracturing experiment is less than 0.001 mD. Thus, the permeability of Coso rock core fractured with a conventional fracturing fluid (Coso 1-2) is at least three orders of magnitude lower than the permeability of a Coso rock core fractured with PAA-CO2 fracturing fluid (Coso 1-1). Previous studies have shown that Coso geothermal field rock consist mainly of diorite which is highly impermeable rock with compressive strengths in the range of 1500-2000 atm. [Berkman (2001), Davatzes (2006)] Coso 1-1 was fractured and the measured applied effective stress was 17 atm when employing PAA-CO2 fracturing fluid as compared to more than 170 atm for an identical rock core (Coso 1-2, control experiment), suggesting that the novel PAA-CO2 fracturing fluid can create/propagate fractures in highly impermeable crystalline rock.

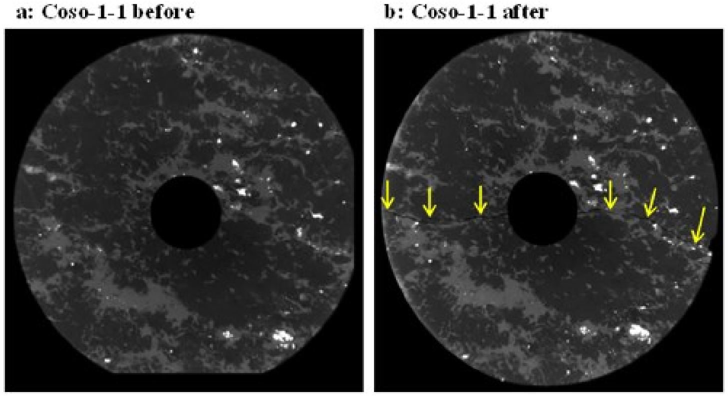


Fig. 3. XMT images of Coso 1-1 before (left) and after (right) hydraulic fracturing experiment. Control experiment (#2, coso-1-2) did not show the presence of fractures even at differential pressures as high as 170 atm (XMT data not shown). The fractures in Coso 1-1 extended ~0.5 mm above and ~17 mm below this transversal section.

Fracturing experiments performed in pH= 7 buffer solutions are also shown in Table I (experiments #3 and #4). The results shows that Coso 1-3, which was subjected to a 1 wt% solution of PAA in buffer pH=7 followed by CO2 injection, was fractured at an effective stress of 27 atm, while the effective stress required to fracture an identical rock core using the conventional fracturing fluid (pH= 7 buffer solution in the absence of PAA, Coso-1-4) was nearly 70% higher. Similarly, the experimental permeability values of Coso-1-3 fractured with PAA-CO2 fluid system was 2.0 mD, which is nearly four times higher than the experimental permeability obtained when fracturing with a conventional fluid (control experiment, Coso 1-4). XMT analyses also provide evidence that the fractures on Coso 1-3 were larger than Coso 1-4 demonstrating once again the potential of PAA-CO2 fracturing fluids for creation/propagation of fracture networks in highly impermeable crystalline rock (Fig. 4). The fractures in rock sample Coso 1-3 were located about 0.5 mm from the top and extended longitudinally~10 mm further down.

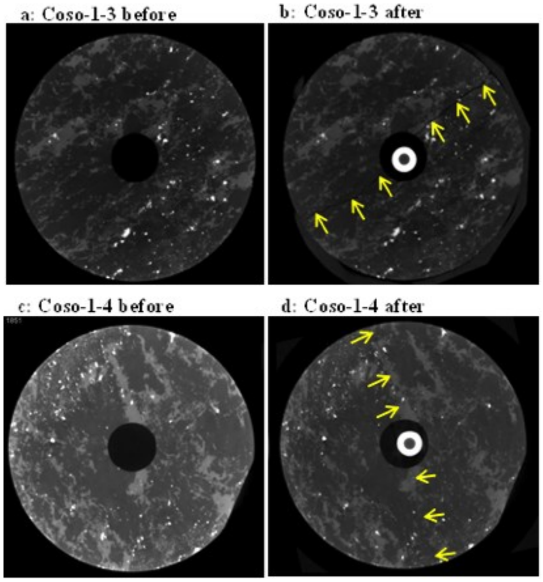
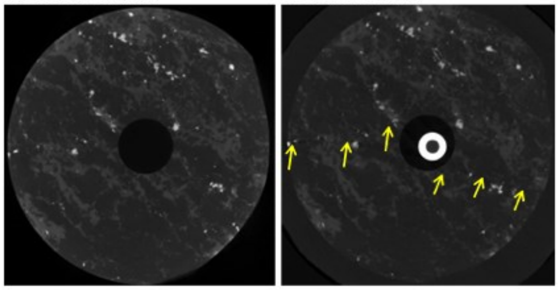


Fig. 4. Representative XMT images of Coso 1-3 (a, b) and Coso 1-4 (c, d) rock cores before and after hydraulic fracturing experiment. Coso 1-3 and Coso 1-4 rock cores were subjected to pH= 7 buffer solutions and CO_2 with and without 1wt% solution of PAA, respectively. The arrows point to the fractures. Note the much larger aperture of the fractures created on Coso 1-3.

Similar experiments were performed in pH=10 buffer solutions with and withouth a 1 wt% solution of PAA and similar results were obtained (see Table I, experiments #5 and #6). Fig. 5 shows the fractures created/propagated from the center hole to the rock external surface in both samples. Effective stress as low as 24 atm was required to create/propagate fracture networks in Coso 1-5, as compared to a 70 % higher effective stress required to create fracture networks in Coso 1-6 using pH= 10 buffer solution-CO2 in the absence of PAA as the fracturing fluid system. Furthermore, the experimental permeability resulting from fracturing Coso 1-15 with PAA (in pH= 10 buffer)-CO2 fluid was an order of magnitude higher than the experimental permeability of the fracture network created when employing a similar fluid system in the absence of PAA. The results demonstrate that the novel fracturing fluid PAA-CO2 can reproducibly generate fracture networks and significantly enhance permeability in highly impermeable rock cores at different reservoir pHs validating the versatility of PAA-CO2 fluid systems for potential EGS reservoir stimulation.


a: Coso 1-5 before
b: Coso 1-5 after

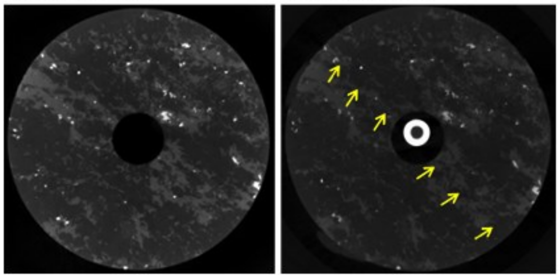

c: Coso 1-6 before
d: Coso 1-6 after

Fig. 5. XMT images of rock cores Coso-1-5 (fractured with PAA in pH= 10 buffer solution and CO_2, top) and Coso-1-6 (fractured with pH= 10 buffer solution and CO_2, bottom) before (left) and after (right) hydraulic fracturing experiment. Note the significant larger aperture on Coso-1-5. The fractures on Coso-1-5 extended ~3 mm above and ~ 6 mm below this transversal section shown in the figure.

Table 1. Experimental conditions and results for the hydraulic fracturing experiments

Exp. No.	Rock Sample	Temp. (°C)	Pressure (atm)	Fracking fluid	Effective pressure of rock fracking. (atm)	Permeability after fracturing exp. (mD)	Gas leaking test
1	Coso-1-1	300	333	PAA-CO_2	17	8.8	Yes
2	Coso-1-2	300	333	DIW/CO_2	>170 [a]	< 0.001 [b]	No [c]
3	Coso-1-3	300	333	PAA (pH7)-CO_2	27	2.0	Yes
4	Coso-1-4	300	333	pH 7 buffer-CO_2	45	0.60	Yes
5	Coso-1-5	300	333	PAA (pH10)-CO_2	24	0.35	Yes
6	Coso-1-6	300	333	pH10 buffer-CO_2	41	0.032	Yes

a: No communication observed even at maximum pressure allowed by the pump. Also, gas leaking test and XMT analysis did not indicate any fractures on this rock core; *b*: no liquid was observed on the external surface even after applying 10 atm pressure to the center hole for 5 min, suggesting the permeability is lower than 0.001 mD. c: no gas flow observed.

4. CONCLUSIONS

The urgent need for alternative renewable energy sources is well recognized. Geothermal heat recovery is among the cleanest emission-free energy sources and EGS the future of geothermal energy. There are two primary limitations that makes commercial EGS infeasible; 1) the current inability to cost-effectively create high-permeability reservoirs from impermeable, igneous rock within the 1-4 km depth range and 2) the widespread concern about the synthetic and highly-proprietary chemicals employed in conventional fracturing with the resulting potential contamination of shallow drinking water aquifers. This work evaluates at the laboratory scale the versatility, with respect to reservoir pH, of an environmentally-friendly and potentially recyclable hydraulic fracturing fluid. This single-component fracturing fluid undergoes a controlled and large-volume expansion with a simultaneous increase in viscosity triggered by CO_2 at temperatures relevant for reservoir stimulation in EGS. The results demonstrate that this novel fracturing fluid could, in principle, be applied for permeability enhancement in EGS reservoirs across the US due to the consistent increase in permeability, respect to control fluids, independently of the pH. The phase-switchable fluid fractures impermeable igneous rocks at lower effective stress, and increases permeability to a significantly (several times) larger level, compared to conventional hydraulic fracturing methods. This novel fracturing method promises to be cost-effective and environmentally cleaner than currently employed approaches for reservoir stimulation in EGS. This is due to both, the efficiency and effectiveness of the permeability enhancement in a range of pHs and the potential extractability of the proposed fracturing fluid due to its previously reported rheoreversible properties. [Fernandez (2015)]

5. ACKNOWLEDGEMENTS

We are grateful to Joseph Moore at Energy & Geoscience Institute for providing rock cores from the Coso geothermal field, Dr. Maura Zimmerschied and Dr. Steven Wiley for very useful edits and suggestions. XMT, and NMR analysis was performed in EMSL (Environmental Molecular Sciences Laboratory; EMSL proposal # 47743), a DOE national scientific user facility at Pacific Northwest National Laboratory (PNNL). Funding for this research was provided by the Geothermal Technology Office of the U.S. Department of Energy. Pacific Northwest National Laboratory is operated by Battelle for the U.S. Department of Energy under contract DE-AC06-76RLO 1830.

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