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Experimental Investigations into CO_2 Injection Associated Fracture Behaviour in Shale Caprocks

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Abstract

Leakage of CO_2 through fractures is a risk for the secure storage of CO_2. Fracture closure, stress state and fracture geometry will control CO_2 leakage rates in storage reservoirs where the seal has been compromised by fractures. This study investigates an acid induced fracture closure remediation technique using a series of laboratory acid injection experiments in shale caprocks. The test aims to reduce the permeability of fractures through induced fracture closure in order to demonstrate the technique as a remedial measure to reduce leakage. In the tests viscous acid is injected through a range of fractured shale caprock samples under confining stress. Preliminary tests on a single sample of shale caprock show that a significant reduction in fracture permeability is achieved using acid injection across a range of confining stresses. CT scans of the fracture show the fracture closure, and appear to suggest that dissolution of asperities on the fracture face may promote fracture closure. Ongoing work will involve testing the technique in a wide range of caprock samples with different mineralogy, artificial fracture surfaces and with gaseous CO_2 and CO_2 rich brines included in the injected fluids to determine any reduction in the effectiveness of the remediation technique, more extensive analysis of the CT imagery will also be carried out.

Peer-review under responsibility of the organizing committee of GHGT-13.

Keywords: Leakage; caprock; fractures; acid injection; remediation; shale

1. Introduction

The risk of CO_2 leakage through fractures is one of the primary risks for the secure containment of CO_2 in geological

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storage formations. The potential for post-leakage fracture closure with pore pressure reduction and sub-surface stress changes, and fracture geometry will control CO_2 leakage rates in storage reservoirs where the seal has been compromised by induced fractures or reactivated pre-existing fractures. Modelling of fracturing formation and reactivation with injection induced stress changes will be important in preventing leakage from occurring, however if leakage does occur, understanding the rate and extent of potential fracture closure with stress changes and whether fracture closure can be increased or accelerated will be important for remediation efforts.

In this study we present a preliminary investigation into an acid induced fracture closure remediation technique using a laboratory acid injection experiments in a shale caprock. Viscous acid is injected through fractured shale caprock core sample under confining stress. A previous study has suggested that fracture apertures in claystone caprocks are likely to increase, and fracture permeability is likely to increase in caprocks exposed to acid CO2 rich brines and water vapour saturated CO2 gas [1]. In this study the permeability of the fractured samples under confining stress is measured before and after acid treatments using brine flow rates and pressure differentials, to determine how the acid treatment affects fracture apertures, fracture aperture and brine flow. Fracture aperture geometry is characterized from sets of CT sections taken across the core during the acid injection and brine permeability measurement cycles. The study focusses on the extent to which fracture closure can be enhanced, and the effectiveness of the technique in sealing a pre-existing fracture.

2. Method

2.1. Sample type

Kimmeridge Clay collected from Kimmeridge Bay in Dorset was used as the fractured caprock sample in this study. Kimmeridge Clay is an important caprock in several fields in the UK North Sea, e.g. Mary field [2], and would potentially form a seal for several CO_2 storage reservoirs if CO_2 storage in the North Sea was widespread. Table 1 shows the results of QXRD analysis for the Kimmeridge Clay used in the study, the sample has particularly high calcite and dolomite fractions compared to other Kimmeridge Clay samples in comparable studies [3].

Table 1.

Composition	%
Quartz	14.5
Albite	1.7
Calcite	10.8
Dolomite	35.5
Mica	5.0
Illite-Smectite	23.9
Kaolinite	3.7
Pyrite	3.7

2.2. Sample preparation

A 37.8 mm (diam.) x 71.5 mm (length) core was taken from the sample and was cored perpendicular to the bedding. Due to the difficulty in producing a core containing a natural axial fracture, an artificial fracture was induced along the length of the core using Brazil disk testing equipment. To produce an open fracture that would have significant permeability, the two halves of the fractured core were offset using PVC spacers at opposite ends of the core, so that the fracture was held open by a series of asperities. The core was then saturated in brine produced from equilibrating distilled water with crushed Kimmeridge Clay prior to testing.

2.3. Testing procedure

The testing programme consisted of an initial measurement of fracture permeability using brine flow, followed by an injection of viscous acid (pH 1) into the fracture. The acid was then flushed from the fracture using brine injection and subsequent measurements of brine permeability were taken until a stable value was reached. Acid injection and subsequent permeability measurements were then repeated, at the initial and higher confining stresses.

The experimental setup used is illustrated in Fig. 1. The core was placed in the core holder, and initially confined at 1500 psi, brine was then injected into the sample at a constant flow rate to measure the permeability using the differential pressure, the downstream outlet from the sample was at atmospheric pressure, and the outflow was monitored with a pH meter.

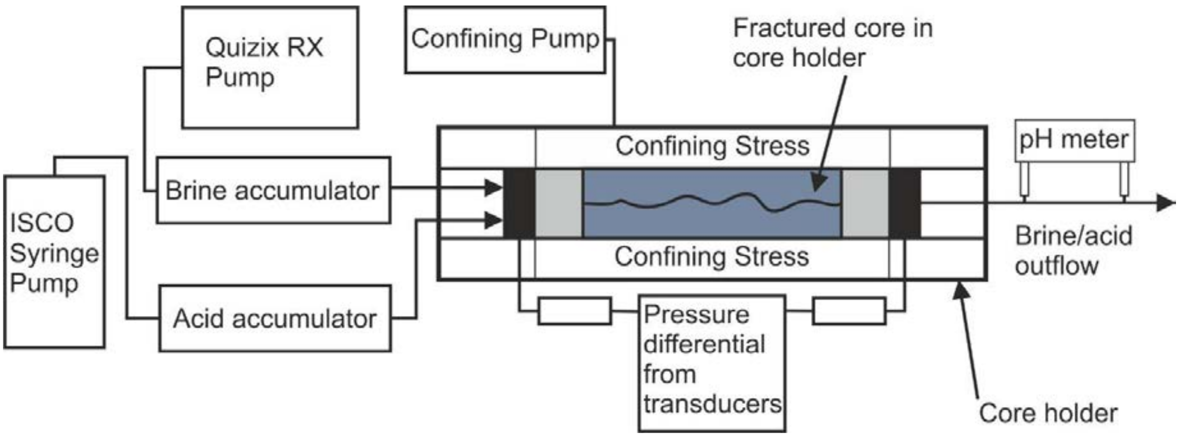

Fig. 1. Experimental set-up

The permeability was calculated for the whole sample (ks) using Darcy's law:

$$k_s = \frac{Q\mu L}{A_s \Delta P} \tag{1}$$

where Q is the flow rate, μ is viscosity, L is the length of the core, A_s is the cross sectional area of the sample, and ΔP is the differential pressure measured across the core. The fracture permeability was calculated using the cubic law assumption and the method of [1] and [4], where the fracture hydraulic aperture (α_h) is given by:

$$\alpha_h = \left(12 \frac{Q\mu L}{w_f \Delta P}\right)^{\frac{1}{3}} \tag{2}$$

and the equivalent fracture permeability (k_f) is given by:

$$k_f = \frac{{\alpha_h}^2}{12} \tag{3}$$

where w_f is the width of the fracture, which is roughly equivalent to the core diameter.

A gradual decrease in the initial permeability measurements suggested that creep in the sample was reducing the initial fracture aperture during the initial confining stage, and the sample was left for 1 week to stabilize to ensure the final initial permeability measurement was unaffected.

After initial permeability characterization a viscous acid was injected into the sample at a constant rate of 0.5 ml/min until acid was observed in the outflow container, the sample was then left for 24 hours before brine was flushed through the sample and permeability measurements were attempted. Initial permeability measurements after acid injection were not successful as a steady state was not reached and the sample appeared to be exhibiting creep. The sample was left for approximately 1 week before stable permeability measurements were achieved. The acid injection and brine flushing and permeability measurement was then repeated at a 1500 psi confining stress, and then confining stress was increase to 3000 psi and then 4000 psi. Therefore the initial 'pre-acid' permeability measurements at the later confining stresses are affected by the initial acid injections at 1500 psi.

The experiment was set up on a custom rig in a CT scanner, and so could be scanned periodically throughout the testing programme, scans were obtained before acid treatment, immediately after acid injection and a various intervals subsequently.

3. Results

3.1. Permeability Measurements

The permeability measurements obtained before and after two acid treatments under 1500 psi confining stress are shown in Fig. 2. The figure shows that there is a significant decrease in permeability in the sample after 1 acid treatment, k_s decreases from 128 mD to 19 mD, and k_f from 106 D to 30 D, the decrease in k_s is more significant than that of k_f. The figure also shows that the second acid treatment is less effective, with only a small decrease in permeability observed, roughly 4 md/D in each case.

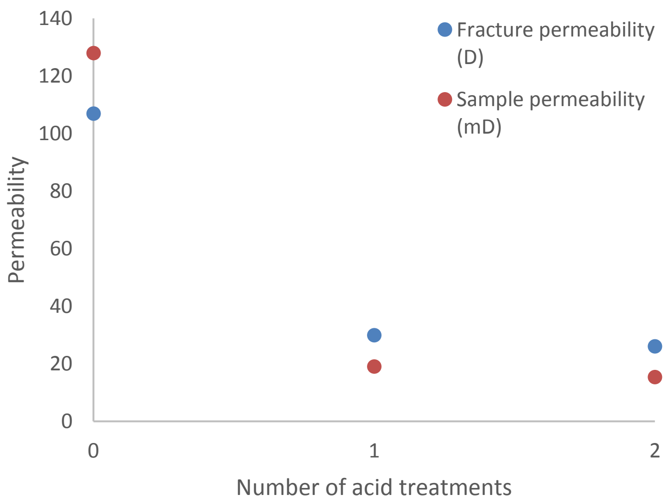

Fig. 2. Fracture and sample permeability before and after acid treatment at 1500 psi confining pressure.

Fig. 3 and 4 show the fracture and sample permeability at different confining stresses, the figures show that the most significant decrease in permeability is observed in the 1500 psi confining stage, but that a reduction in permeability continues to occur with repeated acid treatments at higher confining stresses. The percentage decrease in fracture permeability due to acid treatment is 72% in the 1500 psi stage, 29% at 3000 psi and 57% at 4000 psi. The percentage decrease in sample permeability is generally higher than for the fracture permeability, 85% at 1500 psi, 40% at 3000 psi and 72% at 4000 psi.

The figures also show the decrease in permeability due to the increase in confining stress in the sample. This is represented by the change in permeability from the post-acid permeability to the pre-acid permeability for the next confining step. For an increase in confining stress from 1500 psi to 3000 psi, fracture permeability decreases by 65% and the sample permeability by 79%, for the 3000 psi to 4000 psi, the decrease is 44% (k_f) and 58% (k_s).

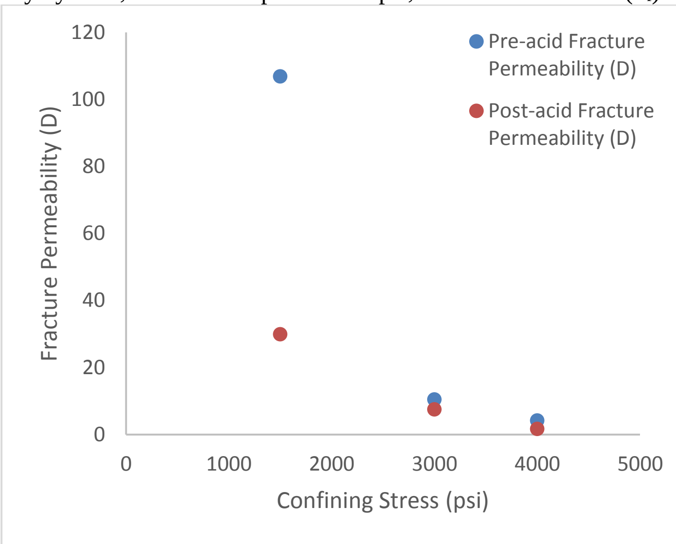

Fig. 3. Fracture permeability before and after acid treatment at a range of confining pressures.

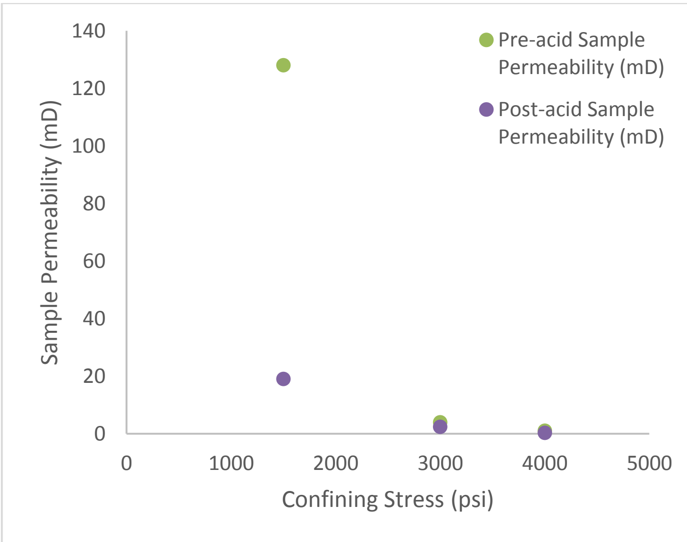

Fig. 4. Sample permeability before and after acid treatment at a range of confining pressures.

3.2. CT Investigation

CT scans were taken throughout the acid injection experiments, Fig. 5 is a plot of the profile of the CT number (Hounsfield units) along the core during the before and during the first acid injection (1500 psi confining) and for several points in the following 60 hours (up to T60 = 60 hours). A moving average is applied to the profile. The plot represents density for a specific region of interest immediately surrounding the fracture aperture along the core. The profile shows that the fracture region was densest at the initial state prior to acid injection, and after the 60 hour time period had elapsed. In between these times the fracture region density is consistently lower, with scans taken at T0, T6.5 and T24 showing very similar density along the profile. T3 is also similarly low, but there is some variation and higher density close to the acid injection inlet. After 60 hours the T60 profile is almost the same as the original pre-acid profile, although is markedly lower at 0-20 mm and 35-40 mm along the core. The smallest reduction in density with acid injection occurs at the end of the sample, (60-70 mm) but is fairly constant throughout the rest of the sample.

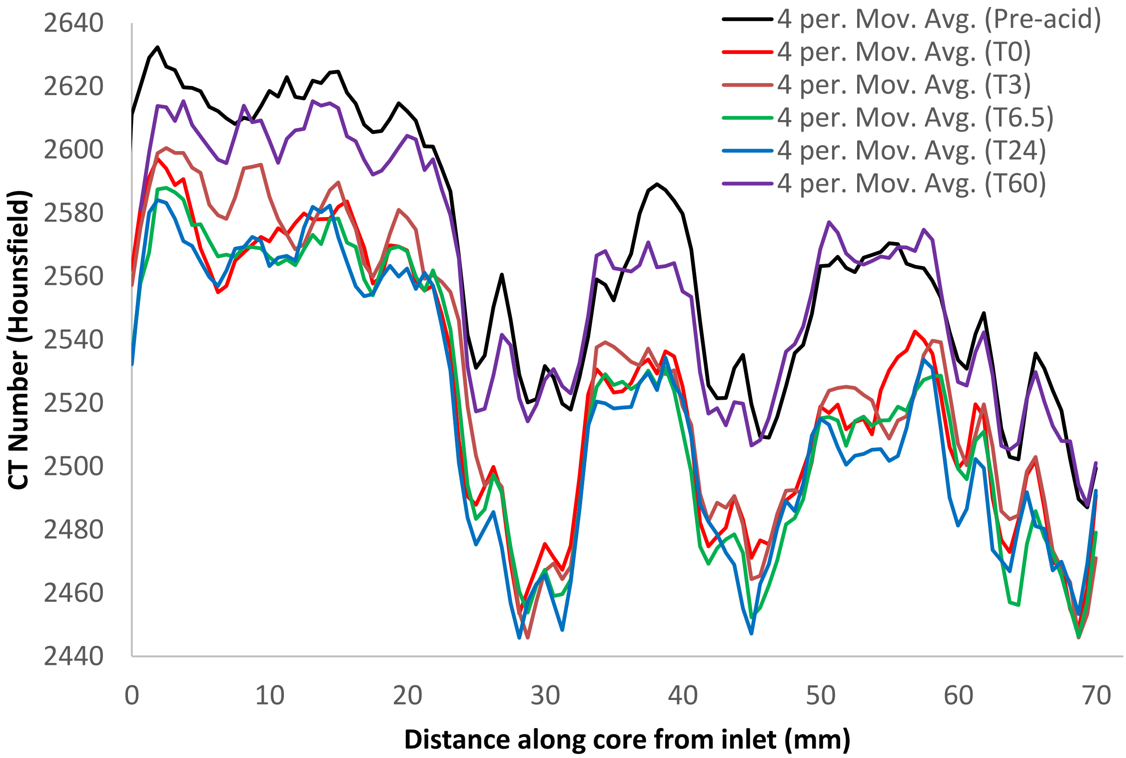

Fig. 5. Profile of CT number along fracture region in sample prior to first acid treatment, and up to 60 hours (T60) after the acid treatment.

Fig. 6 is a comparison of CT image sections along the axis of the core for scans taken during the first acid treatment, lighter colour represents higher density and the fracture is manifested as a low density band running across the sample. The figure shows scans before and after acid treatment at several locations along the core. Regions are highlighted that show significant difference in the fracture between the pre- and post-acid scans. In the image at 66.9 mm from the fluid inlet, the fracture aperture in the post-acid scan appears to have reduced, and the fracture region has a higher density. In the image at 67.5 mm the fracture aperture appears to have widened, and density in the fracture appears to have reduced after acid treatment. In the final image at 70.0 mm, there again appears to be fracture aperture closure in the post acid sample, and the fracture aperture is no longer visible in the region highlighted.

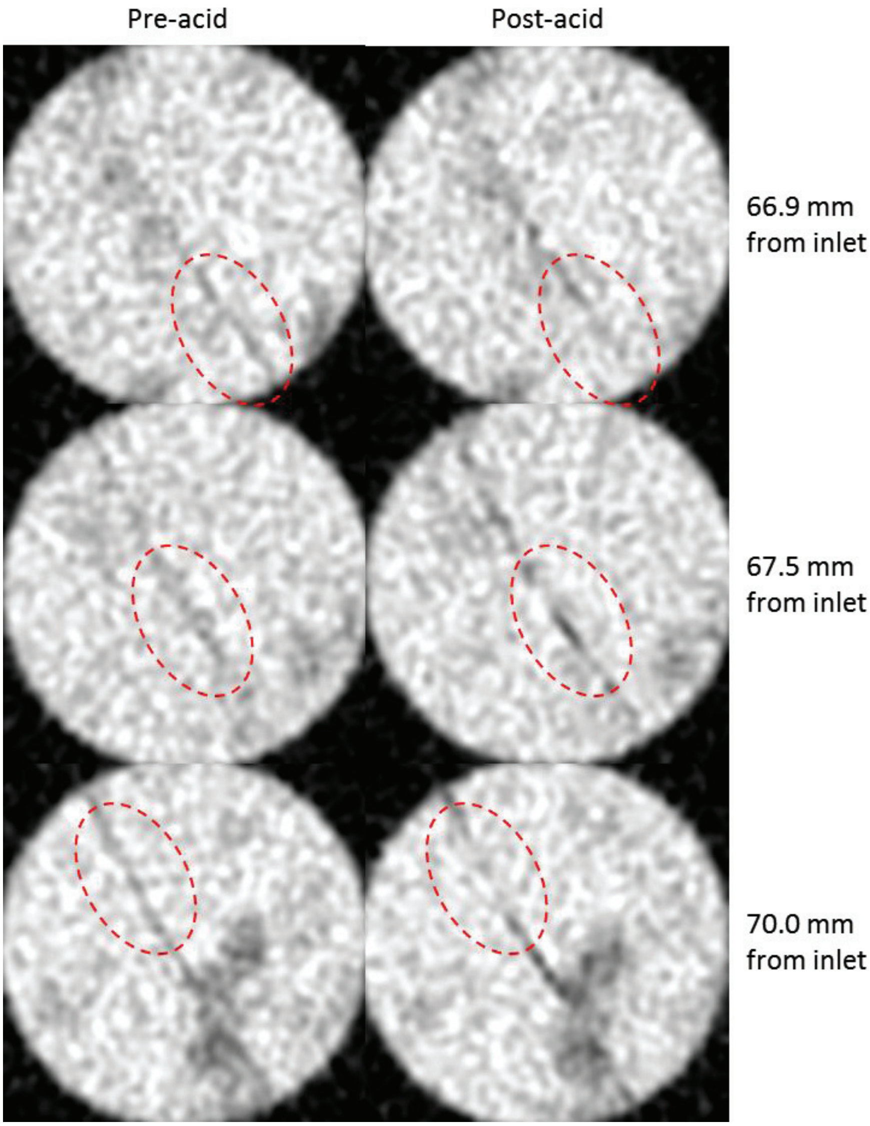

Fig. 6. Axial CT sections of sample showing fracture geometry before and after acid treament at several locations along the sample.

4. Discussion

The permeability measurements and CT scan data show that the acid injection into the fractured caprock sample does induce a reduction in fracture permeability and this appears to be linked to fracture closure in the sample under confining stress. The CT density profiles along the core show that the acid injection reduces the density response in a region of interest around the fracture immediately after acid injection and this appears to be linked to dissolution of material along the fracture face. After 60 hours the density profile returns almost to the pre-acidified state, however associated permeabilities are then significantly lower. The density decrease is interpreted as indicating preferential dissolution of carbonate material in asperities on the fracture surface which hold the fracture open against the confining stress. Once these asperities are degraded the confining stress closes the fracture aperture, closure of the aperture leads to an increase in denser material within the region of interest, as the fracture walls move inwards, and the density response is higher. The scan results suggest this process was complete within 3 days of acidification of the fracture.

Analysis of CT image data shows that the change in the fracture aperture is non-uniform, some regions of the fracture appear more open after acid treatment, and others appear to show fracture closure. The variability is likely to be due to the variable nature of flow paths in the sample, allowing different exposure to acid flow, and the inhomogeneity of the sample composition, with different areas preferentially dissolved. Artificial samples with regular sets of asperities could reduce this variability in the analysis.

The trends in the sample and fracture permeability were similar in the test, but the sample permeability showed greater variation. This discrepancy is potentially linked to fact that the cross-sectional area used in the calculation of k_s is not updated during the test, while the hydraulic aperture in the calculation of k_f implicitly derives from the measured variables.

The acid injection appeared to continue to be effective throughout the test, with reductions in permeability in each test, the smallest reduction in permeability observed was in the 2nd acid injection in the 1500 psi confining stage, only 10-20% reduction was observed, in this test the acid was flushed from the sample prematurely, and may not have reacted fully with the sample. The results in this preliminary test are contradictory to those presented in [1], where it is shown that fracture apertures and fracture permeability increases with acid flow in a mudstone fracture. Acidic CO_2 rich brine and CO_2 gas are flowed through a claystone in [1] and the test is carried out under zero confining stress, the mechanism for aperture increase is decohesion of clay particles during the gas flow phase. The two tests are therefore probably not directly comparable, as there is no mechanism for fracture closure in [1] which may alter flow paths, particularly in areas that have reacted significantly with the acid, and no gas related decohesion would occur in this study. However, leakage through a caprock may involve CO_2 in a gaseous state under the appropriate pressure and temperature conditions, and the combined effect of confining stress, and gaseous CO_2 would be an interesting extension to this study, particularly relating to the effectiveness of attempted sealing of fractures that have experienced gas flow.

5. Conclusion

Overall this preliminary study has demonstrated the potential for fracture permeability reduction using injection of a viscous acid into a caprock fracture, particularly where leakage is predicted to occur via CO_2 saturated brines. This indicates that it may be possible to mitigate CO_2 leakage through a caprock fracture to some extent using an acid injection treatment. However, many uncertainties remain and several extensions to the study can be suggested. The caprock used in the study has a high proportion of carbonate material, other caprocks should be investigated to determine the effect of acid injection on samples with very low carbonate fractions. Different fluid such as CO_2 rich brines and gas should be used in the study, to simulate conditions that may be encountered in a real CO_2 storage leak. The impact of water saturated gas flow CO_2, shown to increase permeability in other studies [1], in conjunction with acid treatments and brine flushing would show the feasibility of the acid injection technique where CO_2 leaked in a gaseous state. More sophisticated analysis of CT data e.g. [4] and the analysis of the geochemistry [1] of the samples and artificial samples with regular asperities could reveal more information about how the permeability reduction is occurring and the change in fracture geometry. Finally a more extensive test of acid treatments at a single confining stress stage should be carried out to determine the maximum level of fracture sealing that can be achieved with continued acid treatment, and to determine if there is a point at which acid injection becomes counter-productive and leads to an increase in fracture apertures.

References

[1] Andreani M, Gouze P, Luquot L, Jouanna P. Changes in seal capacity of fractured claystone caprocks induced by dissolved and gaseous CO_2 seepage. Geophys. Res. Lett. 2008;35:L14404.

[2] Chandler PM, Dickinson B. The Maureen Field, Block 16/29a, UK Central North Sea. In: Gluyas JG, Hichens HM, editors. United Kingdom Oil and Gas Fields, Commemorative Millenium Volume. London: Geological Society; 2003. 20:587-601.
[3] Gutierrez M, Øino LE, Nygård R. Stress-dependent permeability of a de-mineralised fracture in shale. Mar. and Pet. Geol. 2000;17:895-907.
[4] Deng H, Ellis BR, Peters CA, Fitts JP, Crandall D, Bromhal GS. Modifications of Carbonate Fracture Hydrodynamic Properties by CO_2 Acidified Brine Flow. Energy & Fuels. 2013;27.8:4221-4231.