An Experimental Approach to Assist in Quantifying Fracture Sealing Mechanisms in Geothermal Systems

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Abstract

Fluid flow in geothermal reservoirs with low primary permeability is largely controlled by faults and fracture networks. Under certain conditions, hydrothermal veins can precipitate within fractures to form barriers to fluid flow, decreasing permeability and the efficiency of a potential resource. The microstructure and geochemistry of veins can be used to determine the stress, strain, pressure and temperature that a rock mass has experienced, as well as fluid pathways and composition. Fracture sealing in rocks containing pore fluid has been shown experimentally to be a function of time, temperature and fracture dimensions. Sealing is also dependent on changes in fluid pressure and composition; however, this is less well understood. Using a combination of published and new experimental datasets, the evolution of quartz and calcite vein precipitation rates, microstructure and permeability during changes in fluid flux can be constrained. To help understand the processes involved in vein evolution, a new high-pressure, high-temperature triaxial deformation apparatus has been designed to simulate a range of upper crustal geothermal gradients whilst under confining pressure. Two types of experiment using fractured core samples have been designed to investigate vein growth mechanisms and resultant effects: 1. Static, where the sample is overpressured with supersaturated fluid and fluid pressure is reduced at varying rates; 2: Evolution, where supersaturated fluids are flowed through the sample at different rates. Data emerging from this new experimental setup, when considered alongside natural microstructures and other theoretical and experimental data, can be used to predict the conditions under which precipitation is most likely to increase scaling in wells or reduce reservoir permeability, thereby improving subsurface models of geothermal reservoir production or stimulation.

1. INTRODUCTION

Natural and enhanced geothermal resources are increasingly found to be hosted in low primary permeability crystalline reservoirs, within which fluid flow is largely controlled by faults and fracture networks (Bertani, 2015; Brace, 1980; Dezayes et al., 2010). Geothermal reservoirs require high permeability and near-hydrostatically pressured fluids (i.e. high flow rates) to efficiently transport hot fluids to the surface (Limberger et al., 2018; Sibson, 2000). In order to understand the evolution of geothermal systems and develop them successfully, it is important to study how fracture networks form (i.e. stress and reservoir mechanics (Davatzes & Hickman, 2010)), how they may seal with minerals precipitated from geothermal fluids (Dobson et al., 2003), and their crack-seal cycle history (McNamara et al., 2016). Transient fluid pressures and chemical disequilibrium between fluids in the host rock and fractures results in pulsed transport through the fracture network, thereby sealing fractures over time by hydrothermal mineral precipitation (Bons et al., 2012), which decreases the permeability and the efficiency of a potential resource (Dobson et al., 2003). The mineralogy, geochemistry and microstructure of mineral veins provides a record of pressure, temperature and strain history, fluid compositions, and interactions between fluids and fractured host rock (Bons et al., 2012).

Study of how hydrothermal minerals may seal fractures and influence palaeo-permeability is critical to establishing the evolution and sustainability of fractured geothermal systems (Gomila et al., 2016; Sibson, 1996). Fracture sealing processes are sensitive to a wide range of interacting factors, such as the degree of fluid supersaturation, mineral growth kinetics, deformation rates and fluid flux (Hilgers et al., 2004), yet these must be investigated in order to fully understand the evolution and sustainability of fractured geothermal systems. In the absence of long-term in-situ monitoring, an experimental approach can be adopted to obtain relevant data. This paper is intended to act as an introduction as to why and how an experimental appraatus has been designed with the intention of answering the following questions: 1) What are the physio-chemical processes involved in fracture healing during quartz/calcite precipitation? 2) At what rates does precipitation occur, and how does this depend on confining pressure and temperature? 3) How does the rate of pore fluid pressure change affect precipitation rates and resulting microstructures?

Section 1 of this paper provides an overview of how mineral veins can be used as a proxy for fluid flow, before going on to review the laboratory studies to date that explore fracture healing, mineral solubility and precipitation. The relevance of each of these factors to geothermal exploration is then briefly summarized. In section 2, the design of the experimental apparatus is outlined, together with a summary of proposed experimental setups and parameters. Section 3 summarizes what has been done to date and gives an outlook on future work to be done.

1.1 Fluid Flow Mechanisms and the Formation of Hydrothermal Veins

Fluid flow, and hence vein formation, is typically focused in the permeable damage zone of a fault, where many deformation mechanisms may play an important role. These include fluid migration and percolation, reseal hardening or weakening, rock mechanical and transport properties, chemical and physical growth of minerals, palaeostress orientations, and crack-seal mechanisms (Bons et al., 2012; Faulkner et al., 2010, 2011; Faulkner & Rutter, 2000; Sibson, 1996; Woodcock et al., 2007). The precise mechanisms by which syntectonic veins (i.e. mineral aggregates precipitated from a fluid in dilatational sites) form is not

well understood - particularly the links between microstructure and geochemistry, whether void formation is necessary prior to vein growth, and whether veins record advective or diffusive mass transfer (Barker et al., 2006; Bons et al., 2012; Gomila et al., 2016; Hilgers & Urai, 2002a). Veins are pervasive within the upper crust and reflect the dominance of diffusive mass transfer processes via solution occurring alongside cataclasis, both of which occur during relatively low-grade deformation (Blenkinsop, 2002). The initiation and growth of veins occurs via processes that supersaturate the pore fluid with respect to a particular mineral (Wiltshko & Morse 2001), hence the internal texture of veins likely represents their formation mechanisms. Both microstructural and geochemical characteristics can be used to understand vein opening kinematics and the interplay between deformation and precipitation (Bons et al., 2012; Herrera et al., 2005; Olivares et al., 2010). More specifically, an understanding of crystal growth competition, crystallographic preferred orientation, the effects of crystal dilation vs. growth rate, force of crystallisation, and the effects of dislocation densities of seed crystals, supersaturation vs. precipitation rate, fluid salinity, pore size, partial pressure of CO₂, and pH conditions is required (Hilgers & Urai, 2002a).

There are four principal mechanisms involved in vein formation (which almost always combine into less simplified mechanisms) each with increasing transport rates and decreasing fluid-rock interaction: (1) diffusion of dissolved matter through stagnant pore fluid; (2) flow of fluid with dissolved matter through pores; (3) flow of fluid with dissolved matter through fractures and (4) movement of fractures together with the contained fluid and dissolved matter (Bons, 2000; Smith & Evans, 1984). Vein formation (fracture sealing) can occur as a single precipitation event (e.g. a rapid fluid pressure decrease), or as multiple crack-seal events caused by fluid pressure or strain oscillations (Ramsay, 1980). Thermally activated healing processes generally occur near the crack tip, whereby 'islands' of precipitate form a non-uniform surface, creating chemical potential gradients that then cause diffusive material transport and a net reduction of interfacial energy (Barker et al., 2006; Smith & Evans, 1984). Diffusion through a fluid in advecting fluid regimes can occur over long distances and is more effective than non-advecting fluids, which diffuse relatively slowly over distances of ~cm. It has been suggested that fibrous, elongate blocky, blocky and crack-seal textures develop in advective flow regimes (Barker et al., 2006). Textures of veins (Figure 1) are a function of nucleation and growth kinetics, the rate of vein opening, opening vector geometry and wall geometry (Blenkinsop, 2002; Bons et al., 2012; Hilgers & Urai, 2002b). When fracture opening rate is greater than mineral growth rate, crystals precipitate as in a free fluid with corresponding microstructures.

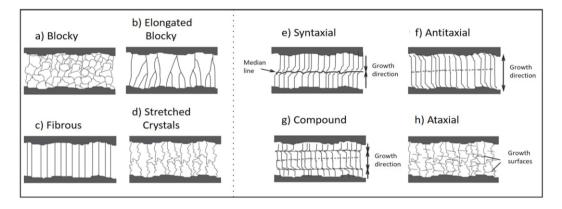


Figure 1: Schematic classification of vein textures, modified from Olivares et al. (2010).

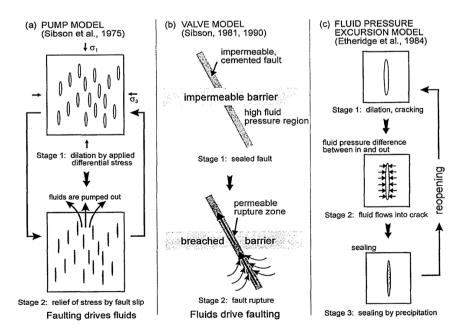


Figure 2: Conceptual models for vein formation (after Lee et al., 1996). a) Pump model (Sibson et al., 1975). b) Valve model (Sibson, 1981; 1990). c) Fluid pressure excursion model (Etheridge et al., 1984).

Several models exist for vein formation by fluid flow processes that are as yet largely unsupported by quantitative data (Etheridge et al., 1984; Lee et al., 1996; Sibson, 1975; 1981; 1990) (Figure 2). In the seismic pumping model (Sibson et al., 1975) fluid migrates down a potential gradient into a dilation site opened by accumulating differential stress or increasing fluid pressure, before dilation site collapse expels the fluids. In the fault valve (suction pumping) model (Sibson, 1981; 1990), dilation rate is greater than fluid flux, developing a temporary underpressure in the void and driving fluid into the dilation site. In both scenarios, mineral precipitation may occur as a result of decreasing fluid pressure. In the fluid overpressure, and resulting potential gradients promote mineral precipitation. Cyclic hydraulic fracturing may also occur, due to sealing during rapid fluid flow along a pressure gradient followed by extensional failure once the minimum effective stress exceeds the tensile strength (Hilgers & Urai 2002a). Veins may also grow from a static fluid via diffusion and solution transfer creep, as material dissolved in sites of high normal stress diffuses through the pore fluid and precipitates at a dilation site (Fisher & Brantley, 1992; Hilgers & Urai 2002a). Planned experimental work discussed in Section 2 aims to further constrain these models, in particular in terms of how they are influenced by differential pressure, temperature, flow rates and crack dimensions.

1.2 Theoretical and Experimental Studies on Fracture Healing

1.2.1 Mineral Solubility

A good knowledge of the conditions at which minerals may precipitate from fluids to form mineral veins is essential to the planned experimental work (Section 2). The concentration of a mineral dissolved within a fluid is a key factor in precipitation mechanisms and rates - a summary of published data on quartz and calcite solubilities and their dependence on various fluid properties is shown in Figure 3. Whilst quartz solubility is mainly dependent on temperature and SiO₂ crystallographic type (Crerar & Anderson, 1971; Fournier, 1989; Rimstidt, 1997), calcite solubility is dependent on a range of factors, including confining pressure (Doubra et al., 2017), partial pressure of CO₂ (Ellis, 1959) and fluid salinity (Ellis, 1963).

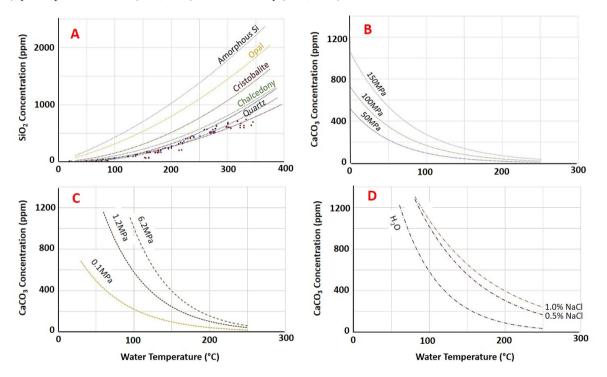


Figure 3: Concentration (ppm) v water temperature (°C) plots for a) SiO₂ crystallinity (Fournier, 1989) with point data from (Rimstidt, 1997) (blue) and (Crerar & Anderson, 1971)); b) calcite, at various confining pressures (Doubra et al., 2017); c) calcite, with various partial pressures of CO₂ (Ellis, 1959); and d) calcite, with various water salinities (Ellis, 1963).

SiO₂ dissolves in water by forming orthosilicic acid, H₄SiO₄. Temperature is a key factor in the rate of precipitation from solution: equilibration of silica may take hours at 250°C, but years at 100°C (Boden, 2016). SiO₂ concentrations in 200–350°C geothermal waters are \sim 300–700mg/kg and are controlled by quartz solubility (Fournier & Rowe, 1966). At room temperature, quartz solubility varies between 6mg/l (microcrystalline quartz) and 120mg/l (amorphous quartz) (Rykart, 1995). At higher temperatures and pressures quartz is easily dissolved by watery fluids percolating the rock. With prograde solubility, these values increase to 700mg/l and >1500mg/l respectively at 300°C and vary slightly due to confining pressures, as well as silica concentration and the specific volume of pure water (Fournier & Potter, 1982).

The behaviour of calcite is controlled primarily by equilibrium in the reaction of solid calcium carbonate $(CaCO_3) + carbonic acid (H_2CO_3) = Ca^+_2 + 2HCO_3$. Any process that increases the amount of CO₂ promotes the production of more carbonic acid, causing the calcite to dissolve. Calcite has a retrograde solubility with respect to temperature, partial pressure of CO₂ and salinity (Figure 3) – i.e. calcite is less soluble in hotter, more CO₂-rich and higher salinity fluids. An increase in confining pressure, however, increases the solubility of gases in liquids causing the production of more carbonic acid and dissolution of calcite.

Beynon et al.

1.2.2 Mineral Precipitation from Pore Fluids

With regards to precipitation in fractures, the amount of healing in rocks containing pore fluids is also a function of temperature, time, and initial mineral concentration in the pore fluid. Any process that decreases the fluid temperature will supersaturate the fluid with respect to silica, making it more likely to precipitate. Once precipitation is initiated, deposition is rapid, particularly with amorphous silica near wellbores (Arnórsson & Gudmundsson, 2003).

Calcite is typically found in geothermal systems with temperatures of $\sim 140-300^{\circ}$ C and where fluids have high concentrations of dissolved CO₂ (Simmons & Christenson, 1994). Any process that increases the temperature, decreases the salinity, decreases the confining pressure or reduces the amount of CO₂ in the system will supersaturate the fluid with respect to calcite, making it more likely to precipitate. Varying temperature profiles along fluid flow pathways in geothermal systems, particularly at operating sites using coupled production and injection wells, create 'cold' zones where highly reactive calcite can be dissolved and 'hot' zones where it can be precipitated. Precipitation of hydrothermal calcite is largely governed by boiling, dilution, and condensation (Simmons & Christenson, 1994). Bladed calcite, commonly found in hydrothermal veins, forms by precipitation from boiling fluids through the exsolution of CO₂, however rates are poorly constrained. A low supersaturation requires large fluid volumes to produce precipitated material; the minimum fluid volume required for sealing may be estimated by dividing the product of the mineral density and fracture porosity reduction by the concentration of silica in the fluid (Dobson et al., 2003).

Importantly, the amount of healing in fluid-filled fractures also depends on the initial crack dimensions and pore fluid pressure (Brantley et al., 1990; Smith & Evans, 1984). Microcracks allow pervasive fluid penetration but heal quickly, whereas macrocracks channel most fluid and heal slowly (Brantley et al., 1990). In a geothermal field where hot (>80°C), mineral-rich water favours precipitation, fracture porosity will likely be short-lived (Laubach & Diaz-Tushman, 2009). Calcite scaling may be promoted by decompression of fluid between the reservoir and wellhead, pH increase (i.e. solubility decrease) due to casing corrosion, removal of CO₂ from the aqueous fluid, or boiling in the geothermal well (Wanner et al., 2017). In general, siliceous fracture healing by diffusive flux, or sealing by minerals derived from external fluid, will be greater with depth due to increases in fluid pressure. Upward migration of cracks and fluids is rate-controlled by quartz growth at the base of the fracture (Fisher & Brantley, 1992). Euhedral growth veins remain open over several seismic-interseismic cycles, as fluid pressures remain high enough to prop open the fracture collapse. The rate of precipitation is linearly related to chemical affinity (Rimstidt & Barnes, 1980), however little data exists at high pressures and temperatures.

1.2.3 Experimental Approaches - A Brief Review

Whilst many modelling studies considering temperatures relevant to geothermal systems have been undertaken (Brantley et al., 1990; Crerar & Anderson, 1971; Doubra et al., 2017; Ellis, 1959, 1963; Fisher & Brantley, 1992; Fournier, 1989; Ghassemi & Kumar, 2007; Giggenbach, 1981; Griffiths et al., 2016; Martin & Lowell, 2000; Ngo et al., 2016; Wanner et al., 2017), particularly for quartz, fewer studies have adopted an experimental approach in a similar manner to that proposed in Section 2.

Experimental studies of mineral growth rates from fluids in porous media have been shown to be orders of magnitude slower than those on free surfaces (Godinho & Withers 2018). Reactive crystal surfaces within less permeable regions grow at a slower rate than that expected from bulk fluid composition, as saturation indices are lower than along main flow paths. These experiments are akin to 'evolution' experiments proposed in this paper (Section 2), whereby a doped fluid with a known saturation index is moved through a fracture at a constant flow rate. Our experiments aim to add temperature and pressure as added variables. Experiments have also been performed whereby a fluid, with controlled flux and saturation levels, has been flowed through simulated fractures. These experiments have been shown to lead to a decrease in mineral growth rate towards the downstream end, causing cessation of flow at the inlet where growth competition and precipitation rates were higher (Hilgers & Urai, 2002b; Hilgers et al., 2004; Lee et al., 2016). A high fluid flow velocity and a low supersaturation increase the potential to seal a vein homogeneously (Hilgers & Urai, 2002a), although it must be noted that low supersaturation levels require unreasonably high fluid volumes to fill natural veins over geological timescales (Lee et al., 1996).

Morrow et al. (2001) found that the permeability of intact granite cycled under a 2MPa pressure differential decreased with time proportional to the temperature (although below 250°C little change was observed). Fractured granites displayed higher rates of permeability decrease at a given temperature, and fracture surfaces showed increased evidence of dissolution and mineral growth with temperature and time. Martin & Lowell (2000) showed that precipitation is a function of initial permeability, heat transfer coefficient and kinetics, whereby decreased permeability (in the order of 10^{-10} to 10^{-14} , as expected in a geothermal reservoir (Lamur et al., 2017)), crack width and flow rate reduces the time taken to precipitate quartz from decades to weeks. High flow rates (10-501/s) are required to make geothermal projects successful (Schulz et al 2003) and should be managed in such way to limit accumulation on the inner surface of pipes according to experimental data. Smith & Evans (1984) also investigated the effect of pore fluid, time and temperature on crack healing. Cracks heal as silica is locally transported by diffusion via the pore fluid along the crack surface, however no change is observed in experiments run with no pore fluid, even at temperatures as high as 600°C. Thermal activation parameters for crack healing suggest that at >200°C microcracks in quartz have geologically short lifetimes.

Related experimental studies on fracture mineralisation have also been performed on quartz gouge (Karner et al., 1997) and barite (Griffiths et al., 2016). Using slide-hold-slide shear experiments, Karner et al. (1997) showed enhanced lithification rates in quartz gouge at elevated temperatures, where friction coefficients increased with both temperature and healing time. Using a combined experimental and kinetic modelling approach, Griffiths et al. (2016) showed that the rate of barite precipitation within open fractures increases dramatically from timescales of months to days as the temperature of the geothermal brine decreases, highlighting the risk of mineral precipitation at geothermal sites, where fluid temperature fluctuates due to circulation through the reservoir rock and fluid mixing around the injection well.

2.1 Design of Laboratory Equipment

A new apparatus capable of reproducing conditions at depth in the Earth's crust has been designed and built in the University of Liverpool's Rock Deformation Laboratory. Independently controlled confining and pore pressures can be modified to create effective pressures of up to ~250MPa. An internal furnace can heat samples to ~750°C during experiments, with minimal temperature gradient across the sample.

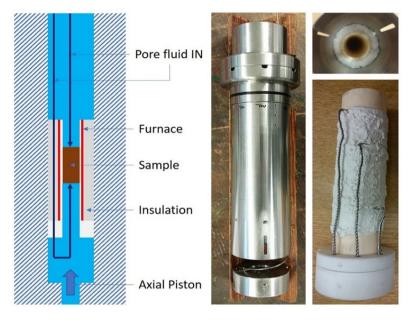


Figure 4: a) Conceptual diagram of a 20mm diameter, 50mm long fractured rock core sample, within a sample assembly with an internal furnace, within a pressure vessel. b) Photograph of the constructed sample assembly. c) the internal furnace. d) internal furnace inside the outer aluminium can, filled with alumina wool insulation.

The sample assembly (Figure 4) comprises an internal furnace, capped at both ends by macor plates both to keep the ceramic tube in place and to hold in a layer of tightly packed alumina wool insulation. This is all held in place by an external aluminium can, into which the top and bottom pistons are inserted: one fixed, the other able to move axially. The internal furnace was made by winding a coil of kanthal wire around alumina tubing. Three sections of coil were made, each of which can be provided with a controlled amount of power, using variable resistors, to provide an even heat distribution across the cored sample. The central coil forms ~one half the total length of the furnace in order to allow maximum power (and therefore heat) input to the central part of the assembly (i.e. the sample). The sample assembly has been calibrated using a hollow pyrophyllite core through which two thermocouples were used to obtain the temperature profile between the base of the downstream and the top of the upstream inputs (cf. Karner et al., 1997). Pyrophyllite has thermal properties such that volume change during heating is insignificant, and thermal conductivity is similar to that which would be experienced during experiments on many rock samples.

The sample assembly is cooled externally using aluminium ring jackets, hollowed with tracks to allow circulation of water to absorb and dissipate emitted heat. A cooling baseplate also isolates the bottom of the vessel from the force gauge, preventing high temperatures or temperature fluctuations from expanding or contracting this part of the rig. Cooling jackets also factor into safety considerations, since they are the first barrier on the outside of the vessel chamber. Gas mediums, such as argon, have a much higher specific heat capacity than silicone oil, which is typically used as the confining medium in high pressure – low temperature experiments. Hence, vessel failure would be exploited by pressured gas with a much greater force than with oil.

Safety measures have been put in place to a) prevent excessive confining pressure build-up, and b) to contain any potential explosive activity. a) Confining pressure limitation: A mechanical pressure limit of 200MPa is set on the air-driven pump, which delivers the confining medium from the reservoir to the vessel. A software pressure limit limiting the user to 250MPa is set on the control programme. A rupture disc is fitted to an elbow on the confining pressure control pump. The disc is pressure rated to \sim 300MPa; should these pressures be reached confining medium will preferentially escape through this area. Each individual component of the vessel (i.e. pressure chamber, top nut, high pressure pipework and connections) has been rated to \sim 400MPa. Each component was thoroughly checked for any damage (e.g. scratches where stress could concentrate) prior to use. b) Containment: Aluminium cooling jackets will provide a relatively soft buffer to any failure perpendicular to the vessel sides. A \sim 50mm sheet of wood between two \sim 10mm steel sheets above the vessel is in place to absorb the kinetic energy of the vessel's top nut, should it fail and be forced upwards. The vessel is housed inside a safety barrier of \sim 10mm steel sheets, sufficient to block any missiles generated as a result of an explosion (which would have already lost energy via the barriers noted in the points above). Ear defenders are to be worn at all times when the vessel is at high pressures, as rapid expulsion and expansion of gas is likely to be accompanied by a loud noise.

2.2 Summary of Planned Experiments

Two main experimental techniques are being considered initially to answer the questions outlined in Section 1: 'static' and 'dynamic' experiments. Both setups, outlined below, will use a sample configuration schematically shown in Figure 5, within the apparatus shown in Figure 4.

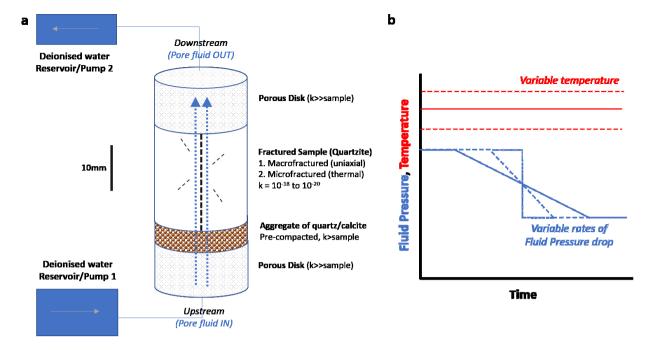


Figure 5: Schematic summary diagram of a) experimental sample setup and b) experiment types.

Planned experiments involve first fracturing the sample to create either a single microfracture, or a series of microfractures. The fractured core sample is then supersaturated with quartz- or calcite-doped pore fluid; this is best achieved by flowing deionized water through a porous disc and mineral aggregate at the upstream end (Figure 5a) in order to prevent any mineralization occurring in the high-pressure pipework. Once pore fluid pressure is equilibrated and sufficient time has been allowed for the fluid to become supersaturated with respect to the mineral in question, 'static' experiments can be performed by dropping fluid pressure instantaneously (Figure 5b) upon opening the downstream valve, allowing fluids to migrate out of the sample and into a second pore fluid reservoir. 'Dynamic' experiments can also be performed by using a servo-controlled system to induce a more controlled flow through the sample, thereby varying the rate at which fluid pressure is dropped (Figure 5b). By changing the equilibration temperature, the rate at which fluid pressure is dropped, or the absolute difference in fluid pressure, the mechanisms by which minerals will precipitate from solution is hypothesized to change (Figure 3).

These initial experiments will be performed at constant confining pressures, however in future experiments these parameters can also be changed to assess the impact these variables have on precipitation mechanisms. Reaction rates of a quartz-water system increase substantially with temperature (Rimstidt and Barnes, 1980) and this relationship must be considered in high-temperature laboratory studies involving a time-dependent phenomenon by approximating the effect of increased reactivity during the time required to reach a given temperature (Karner et al., 1997).

Once experiments have been performed, core samples will be thin sectioned and analysed using several microscopy techniques: optical petrology, scanning electron microscopy, cathodoluminescence, and electron backscatter diffraction (cf. McNamara et al., 2016). Since veins would have formed at known confining pressures, stress/strain orientations, fluid pressures and temperatures, in fractures with well constrained mechanical properties, and since degrees of fluid saturation with respect to quartz and calcite can be reasonably estimated, these microscopy techniques will hopefully aid the interpretation of vein precipitation processes occurring during quartz- and calcite-sealing of micro- and microfractures. Ultimately, the quantification of these processes aims to help improve models of fracture sealing and scaling in fractured geothermal reservoirs in order to improve production efficiency and lifespans of operating geothermal fields.

3. SUMMARY

Fracture sealing in rocks containing pore fluid has been shown experimentally to be a function of time, temperature and fracture dimensions. Sealing is also dependent on changes in fluid pressure and composition; however, this is less well understood. Using a combination of published and new experimental datasets, the evolution of quartz and calcite vein precipitation rates, microstructure and permeability during changes in fluid flux can be constrained. To help understand the processes involved in vein evolution, a new high-pressure, high-temperature triaxial deformation apparatus has been designed to simulate a range of upper crustal geothermal gradients whilst under confining pressure. Two types of experiment using fractured core samples have been designed to investigate vein growth mechanisms and resultant effects: 1. Static, where the sample is overpressured with supersaturated fluid and fluid pressure is reduced at varying rates; 2: Evolution, where supersaturated fluids are flowed through the sample at different rates. Data emerging from this new experimental setup, when considered alongside natural microstructures and other theoretical and experimental data, can be used to predict the conditions under which precipitation is most likely to increase scaling in wells or reduce reservoir permeability, thereby improving subsurface models of geothermal reservoir production or stimulation.

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Beynon et al.

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