

Fluid-Rock Interaction Studies on an Enhanced Geothermal System in the Cooper Basin, South Australia



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Declaration

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Summary

Engineered Geothermal System (EGS) has great potential to supply electricity by harnessing stored thermal energy from high temperature granitic rocks. Since reserves of coal, oil, and natural gas are being depleted at an increasing rate, this route provides opportunities to generate electrical power without producing greenhouse gas emissions or long lasting nuclear wastes, at a cost that is competitive to those generated from fossil fuels. Australia has a vast amount of thermal area, though the heat exchange occurs at a significantly greater depth (5 km) to conventional geothermal system. Clearly, the study of fluid-rock interaction is crucial and remains largely poorly addressed and known. A compounding factor is the fact that fundamental processes associated with mineral dissolution and precipitation, and the developed pressure temperature gradient remain poorly understood. Furthermore, a number of issues relating to geothermal geochemistry are required to be considered and explored to ensure safe, economic energy production from the “hot rocks”. Low pH and saline waters at temperatures exceeding 200°C are highly corrosive. Thus, it is vital to prevent the generation of scaling as the brines cool during transport to the surface.

The objectives of this study were to investigate the geochemistry, the fluid-rock interaction, and model the precipitation rate of silica. Experimental work was carried out to observe the fluid-rock interaction, including analysis on the rock to monitor the dissolved elements in the circulating fluid, and the water chemistry after the interaction. The granite samples were analysed using x-ray diffraction and results showed that the rock consist of mainly quartz, albite and K-feldspar.

This study concentrated on the dissolution rate of granite by observing the silica concentration in the liquid phase with the aid of previous dissolution rate studies of pure quartz, albite and K-feldspars (Rimstidt and Barnes, 1980; Hellmann, 1994; Worley, 1994; Brantley, 2008; Brown, 2011b). In order to investigate the fluid-rock interaction in the Cooper Basin geothermal

system (i.e. Habanero 3 well), three experimental methods at a laboratory scale were developed. To simplify the process, the gas phases were not introduced to the system. The first method allows the interaction of fluid and rock samples in a closed system where no fluid is required to be replaced (fluid mass is constant) during the experimental period. The experiment is conducted in Teflon lined autoclaves for different interaction periods and the maximum temperature chosen was 220°C due to the limitation of the Teflon liners used. This method was used firstly to obtain the equilibrium silica concentration at various temperatures. The experimental results showed good agreement with the literature values. The equilibrium silica concentrations obtained from dissolution at 120°C, 140°C, 160°C, 170°C, 200°C and 220°C for 56 days were 56 ± 3 ppm, 94 ± 6 ppm, 137 ± 6 ppm, 175 ± 7 ppm, 282 ± 11 ppm, and 350 ± 28 ppm, respectively. The second observation was the dissolution kinetics in pure water. The SigmaPlot software was used to fit the experimental data and obtain the equilibrium silica concentration and silica dissolution rate constant based on a first order global rate equation by Worley (1994). The results were compared with a compiled quartz dissolution literature values and showed good agreement, however values differ slightly due to the different materials and experimental conditions. The obtained dissolution rate constants were then regressed using the Arrhenius equation describing a kinetic rate constant with an activation energy of 64.53 kJ/mol.

A number of factors affecting the dissolution rate of granite were observed. One factor was the effect of particle size on the dissolution rate of granite. The experimental results agree with literature, which demonstrated that the dissolution rates increased with decreasing granite particle size (increasing the surface area). Another observation undertaken was the effect of electrolyte (250 ppm NaCl solution) on the granite dissolution rate. The results concluded that the dissolution rate in 250 ppm NaCl solution yielded a two-fold increase compared to that in pure water. One other observation was on the effect of pH in granite dissolution rate. The experimental results agree with the literature confirming that the increase of dissolution rates at lower pH was due to the presence of organic acid (acetic acid) in the pH buffer used. At pH above 8 the dissolved silica species that is significant is not solely $\text{SiO}_{2(\text{aq})}$ (H_4SiO_4). The hydrogen atoms from H_4SiO_4 can dissociate and release H_3SiO_4^- ion which is very soluble in

water. As the pH increase, further hydrogen dissociation is possible to form $\text{H}_2\text{SiO}_4^{2-}$ which is also soluble in water and thus increasing the silica concentration, leading to an increased dissolution rate.

The second method used a closed loop batch flow-through cell that was designed to mimic the circulation of the fluid-rock interaction hence enabling the observation of the changes in the chemical properties of the host rock and circulating fluid that may occur. This method involved two different experimental systems. The first system allows the continuous interaction of the fluid (pure water and 250 ppm NaCl solution) and rock samples at 250°C (close to the actual geothermal reservoir temperature) to study the dissolution kinetics of silica from the granite for different interaction periods. This system was also used to study the effect of fluid/rock ratio. The experimental results agree with the literature which illustrate a decrease in solid/liquid ratio (increase in fluid/solid ratio) would increase the reaction rate. The second system allows the interaction of fluid and rock samples also in a closed loop batch flow-through cell, using pure water and 250 ppm NaCl solution for 7 days and 28 days, and the fluid is consistently replaced every 24 hours for the specified interaction periods. This system was designed to accelerate the mineral dissolution to observe which minerals were more soluble. SEM results revealed that severe pitting exists on the surface of the granite, as a consequence of rapid dissolution, and it was observed that fine particles were present between and on the surface of the granite which increased the particles surface area, enhancing the dissolution rate. The SEM back-scatter images revealed albite as the more soluble phase, since more cavities were observed through the albite phases compared to the K-feldspar phases in the granite samples.

The third method involved a high pressure open loop flow through system, where fresh water is continuously injected to the system. This system was configured to observe the influence of pressure in rock water interaction. Three pressure conditions at 250°C were chosen (at vapour pressure, 100 bars, and 200 bars). The experimental results showed that the silica concentration increased with pressure, agreeing with the published literatures.

In order to validate the experimental results, the React program from the Geochemist Workbench software was used to simulate the granite dissolution reaction path and generate silica dissolution and silica precipitation rates. The simulation in React is based on the transition state theory model (Rimstidt and Barnes, 1980; Bethke, 1996). The results of the modelling showed consistent plots with the experimental results however generated different values of rate constants and equilibrium silica concentrations. React was also used to calculate the amount or rate of silica precipitation with the assumption that the aperture of the fracture was 10 cm and the surface roughness was 2. For granite dissolution in pure water, the amount of silica that may precipitate was approximately 298 mg/28 days, and in 250 ppm NaCl solution is 309 mg/28 days. From the available information, the sealing rate from granite dissolution in water was 2.30 cm/1000 years, and that in NaCl solution was 2.41 cm/1000 years.

Since this was a simplified model and only the major components of the granite were included, it may have influenced the reaction path calculated by React, affecting the silica concentration output and reaction rate. Another contributing factor may be that the active surface area of the granite in the experiment differs with the BET surface area obtained in this study. In addition, the published reaction rate constant may have different experimental conditions (e.g. different composition of minerals, particle size, duration of experiments, different reactors). As well, the input of the reaction rate constant was allowed for single minerals, and the model may not simulate the exact laboratory experimental conditions. Moreover, this study measured the dissolution of granite solely from the release of silica to the solution. Since the literature published reaction rate constant from pure minerals (e.g. albite), this reaction rate constant may not be the appropriate value to specify the albite component in the granite. In other words, the reaction path of dissolving three pure minerals in water may not be identical to the dissolution mechanism of granite with the same mineral composition. Since the model output resulted in some differences compared to the experimental results, this suggests that modelling and experiments should work together to predict more accurate outputs.

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I hope that the results of my thesis would satisfy the expectations of the people associated and provide a significant contribution to the society.

List of Publication

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