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

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A thesis submitted for examination for the degree of
Doctor of Philosophy in Chemical Engineering

October 2014
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 SiO$_2$(aq) (H$_4$SiO$_4$). The hydrogen atoms from H$_4$SiO$_4$ can dissociate and release H$_3$SiO$_4^-$ 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$^\circ$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$^\circ$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.
Acknowledgements

Firstly, I would like to thank God for the strength and wisdom to complete my doctorate program. I would like to express my appreciation to those who have greatly contributed and assisted me to complete this research. In particular I would like to acknowledge:

A/Prof. Yung Ngothai, School of Chemical Engineering, The University of Adelaide, my principal supervisor, for the encouragement and support, guidance, discussions, ideas and the opportunity to carry out research in this field. Thank you for the friendly atmosphere while conducting this research.

A/Prof. Brian O’Neill, School of Chemical Engineering, The University of Adelaide, my co-supervisor, for the guidance and assistance in the calculations and design of the flow-through hydrothermal cell, experimental, as well as dissolution kinetics and thermodynamics.

Prof. Allan Pring, School of Chemistry and Physics, The University of Adelaide, and The South Australian Museum, my co-supervisor, for the expertise in the area of mineralogy and geochemistry, as well as access to XRD equipment and laboratories in the SA museum.
A/Prof. Joel Brugger, School of Earth and Environmental Sciences, The University of Adelaide and South Australian Museum, my co-supervisor, for the guidance and discussions in geochemistry, modelling and Geochemist Workbench simulations. As well to Dr. Barbara Etschmann for the valuable input in experimental and modelling.

Mr. Jason Peak, Mr Michael Jung and Mr. Jeffrey Hiorns, the Chemical engineering workshop team, for assistance in putting together the experimental apparatus, maintenance and making sure the apparatus were working well.

Dr. Benjamin Wade and Aoife McFadden, from the Adelaide Microscopy Centre, for their assistance and training in using the XL30 scanning electron microscope, ICP-MS, and the MicroMeritics Gemini VII with nitrogen adsorption for BET surface area determination.

Mr. John Stanley from the School of Earth and Environmental Sciences for the XRF analyses on the granite samples.

Gujie Qian, Fang Xia, Kevin Li and Jing Zhao for their support in experimental and expertise in software and laboratory equipment.

The research described in this paper has been funded by the Department of Primary Industries and Resources of South Australia (PIRSA) and Geodynamics.

This research would not have been possible without the support of all the people mentioned above.
I would like to dedicate this thesis to my beloved parents, Dr. Tjahjono Koentjoro, MPH, PhD and Dr. Lina Koerniaawati, MPH, and my sister Yael Esthi Nurfitri Kuncoro, SpKK. I thank them for all the love and support in the whole process of completing this study. And of course to my lovely wife, Elita Santosaaputri, B.Med.Sc., BMBS, for her love, support, patience, and understanding, also in accompanying my late night laboratory works. I would also like to dedicate this thesis to my parents-in-law, Drs. Andreas Santoso, MBA and Yusnita Santoso for their support and encouragement. Finally, to my baby boy, Evan Amadeo Kuncoro, my pride and joy, for his presence, smile and laughter.

NOTE:
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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


Table of Contents

Declaration ................................................................................................................................. ii

Summary ................................................................................................................................... iii

Acknowledgements .................................................................................................................... vii

List of Publication ....................................................................................................................... x

Table of Contents ........................................................................................................................ xi

List of Figures .............................................................................................................................. xvii

List of Tables ............................................................................................................................... xxiv

Chapter 1  Introduction and Motivation ..................................................................................... 1

Chapter 2  Literature Review ....................................................................................................... 4

  2.1 Electricity Demand in Australia ......................................................................................... 4

  2.2 Geothermal Energy ............................................................................................................. 7

    2.2.1 Hydrothermal/volcanic system .................................................................................... 8

    2.2.2 Engineered geothermal system ................................................................................... 11

    2.2.3 Geothermal energy in Australia .................................................................................. 15

  2.3 Geochemical Reactions ....................................................................................................... 23

    2.3.1 Thermodynamics ........................................................................................................ 24

    2.3.2 Kinetics of silica dissolution ....................................................................................... 26

    2.3.3 Kinetics of silica scaling ............................................................................................. 31

  2.4 Geochemical Modelling ...................................................................................................... 35

    2.4.1 Transition-state theory applied to silica dissolution ................................................ 36

    2.4.2 Non-linear regression and curve fitting ....................................................................... 42
2.4.2.1 Non-linear rate equation ................................................................. 42
2.4.2.2 Solving non-linear regression using SigmaPlot ............................... 43

2.5 Previous Fluid-Rock Interaction Studies ................................................. 44

2.5.1 Temperature effects ............................................................................ 45
2.5.2 pH effects ............................................................................................ 49
2.5.3 Ionic strength effects ........................................................................... 52
2.5.4 Pressure ............................................................................................... 53
2.5.5 Reactor design ..................................................................................... 55

Chapter 3 Field Sampling at Habanero 3 Well, Cooper Basin ....................... 61

3.1 Materials and methods ........................................................................... 61
3.2 Preliminary results and discussions ......................................................... 62

3.2.1 Habanero well outlet fluid chemistry .................................................. 63
3.2.2 Habanero rock analysis ....................................................................... 68

Chapter 4 Experimental Techniques ............................................................ 72

4.1 Materials .................................................................................................. 72
4.2 Sample Preparation .................................................................................. 73
4.3 Mineral Characterisation and Surface Area Basis ...................................... 74
4.4 Liquid Phase Analyses ............................................................................ 78

Chapter 5 Fluid - Rock Interaction Experiments .......................................... 80

5.1 Static System ............................................................................................ 81

5.1.1 Equilibrium silica concentration in water at various temperature ......... 83

5.1.1.1 Materials .......................................................................................... 83
5.1.1.2 Start-up procedures ........................................................................... 84
5.1.1.3 Experimental procedures and sampling ........................................... 84
5.1.4.1 Materials and experimental .............................................. 107
5.1.4.2 Results and discussion .................................................. 108
5.1.5 Influence of pH on granite dissolution kinetics ....................... 111
5.1.5.1 Materials and experimental ........................................... 111
5.1.5.2 Results and discussion .................................................. 112
5.2 Flow-through System ............................................................ 118
5.2.1 Initial design of the hydrothermal cell .................................. 118
5.2.2 Description of the updated geothermal cell .......................... 122
5.2.3 Materials and preparation of the geothermal cell .................. 126
5.2.4 Sample basket preparation ................................................. 126
5.2.5 Experimental procedures .................................................. 127
5.2.6 Results and discussion ...................................................... 131
5.2.6.1 Recycled system .......................................................... 131
5.2.6.2 Non-recycled flow-through system .................................. 150
5.3 High Pressure Flow-through System ........................................ 157
5.3.1 Description of the high pressure flow-through cell .................. 157
Appendix F  Mass Transfer Coefficient .................................................................291

F.1  Static system ........................................................................................................291

F.2  Flow-through system ..........................................................................................292

Appendix G  Darby’s Bore Water Analyses .................................................................295
# List of Figures

| Figure 2.1 | Australia’s sources for power generation .................................................. | 5 |
| Figure 2.2 | A coal fired power plant .................................................................................. | 5 |
| Figure 2.3 | Forecast of Australia’s electricity demand ................................................... | 6 |
| Figure 2.4 | Australia’s greenhouse gas emissions in 2004 .............................................. | 6 |
| Figure 2.5 | Plate boundary map ......................................................................................... | 9 |
| Figure 2.6 | Location of geothermal power plants in the world .......................................... | 10 |
| Figure 2.7 | Geothermal growth projection ......................................................................... | 10 |
| Figure 2.8 | Enhanced Geothermal System (EGS) thermal extraction process ..................... | 14 |
| Figure 2.9 | Binary cycle power plant ................................................................................. | 15 |
| Figure 2.10 | Schematic diagram of the Birdsville Geothermal Power Station (Queensland Environmental Protection Agency) .................................................. | 17 |
| Figure 2.11 | Australia’s geological temperature at 5 km depth ......................................... | 18 |
| Figure 2.12 | Comparison of electricity cost produced from hot dry rock versus other resources .................................................................................................................. | 19 |
| Figure 2.13 | Australian map showing geothermal exploration licenses, applications, and gazettal areas .................................................................................................................. | 20 |
| Figure 2.14 | Geothermal exploration licenses in the South Australian region ................... | 21 |
| Figure 2.15 | Tenement map of the Cooper Basin geothermal system ................................ | 22 |
| Figure 2.16 | Equilibrium solubilities of silica phases ........................................................ | 26 |
| Figure 2.17 | Steps involved in silica polymerization .......................................................... | 32 |
| Figure 2.18 | Rate of monomeric silica disappearance as a function of time at various temperatures (Brown, 2011b) ............................................................... | 34 |
| Figure 2.19 | Silica polymerisation as a function of pH (Brown, 2011b) ............................... | 34 |
| Figure 2.20 | Schematic of free energy maximum through which reactants must pass to become products .................................................................................................................. | 36 |
Figure 2.21  Schematic of the formation of an activated complex on a surface of K-feldspar grain reacting with an acid aqueous solution .......................................................41
Figure 2.22  Schematic representation of the Gibbs free energy change associated with the hydrolysis of K-feldspar in an acid aqueous solution ........................................41
Figure 2.23  The effect of pH on the dissolution rate of quartz ........................................50
Figure 2.24  The effect of pH on the dissolution rate of albite ........................................51
Figure 2.25  Compilation of quartz solubility data at saturated vapour pressure and at 1000 bars (Worley, 1994) ........................................................................................................54
Figure 2.26  Pressure effect in quartz dissolution using the correlation by Fournier and Potter II (1982a) ........................................................................................................55
Figure 2.27  Stirred autoclave reactor used by Robinson (1982) ...........................................56
Figure 2.28  Titanium continuous-flow stirred tank reactor used by Worley (1994) ..........58
Figure 2.29  Experimental apparatus used by Azaroual & Fouillac (1997) .......................59
Figure 3.1  Back-scattered SEM image of Habanero 3 well rock sample from various points...
.............................................................................................................................69
Figure 3.2  XRD pattern of Habanero 3 rock sample after ultrasonically cleaned ..........70
Figure 4.1  Surface of granite particles after ultrasonically cleaned (100-200 μm) ..........74
Figure 4.2  Relationship between BET surface area and particle size ..................................77
Figure 5.1  Stainless steel autoclave with Teflon liners .....................................................82
Figure 5.2  Photograph of static Teflon experimental set up ...............................................83
Figure 5.3  Experimental results showing silica concentration at temperatures 120°C, 140°C, 160°C, 170°C, 200°C, and 220°C .................................................................85
Figure 5.4  Linearization of equilibrium silica concentration from experimental data .......86
Figure 5.5  Equilibrium silica concentration at various temperatures compared with literature .................................................................................................................87
Figure 5.6  Equilibrium concentrations of dissolved Na, K, and Al at various temperatures ....
.......................................................................................................................................88
Figure 5.7  Reactive silica concentration profile for four different temperatures (160°C, 170°C, 200°C, 220°C) .........................................................................................91
Figure 5.8  Approximation of equilibrium silica concentration (showed by the dashed lines) .................................................................93
Figure 5.9  Arrhenius plot for 160°C, 170°C, 200°C and 220°C .................................................................96
Figure 5.10  Concentration of dissolved Na, K, and Al from granite dissolution in water at 160°C .................................................................98
Figure 5.11  Concentration of dissolved Na, K, and Al from granite dissolution in water at 170°C .................................................................98
Figure 5.12  Concentration of dissolved Na, K, and Al from granite dissolution in water at 200°C .................................................................99
Figure 5.13  Concentration of dissolved Na, K, and Al from granite dissolution in water at 220°C .................................................................99
Figure 5.14  Reactive silica concentration profile for three different particle size (40 – 60 µm, 100 – 200 µm, and 200 – 400 µm) at 200°C in water ...........................................101
Figure 5.15  Comparison of silica dissolution rate constants (k_i) in pure water at various temperatures using geometric surface area basis ........................................104
Figure 5.16  Comparison of silica dissolution rate constants (k_i) in pure water at various temperatures using BET surface area basis ........................................105
Figure 5.17  Concentration of dissolved Na, K, and Al from granite dissolution in water at 200°C using 40 – 60 µm particle size ...........................................106
Figure 5.18  Concentration of dissolved Na, K, and Al from granite dissolution in water at 200°C using 200 – 400 µm particle size ...........................................106
Figure 5.19  Comparison of granite dissolution profile in water and 250 ppm NaCl solution at 200°C in autoclave .........................................................108
Figure 5.20  Concentration of dissolved K and Al from granite dissolution in 250 ppm NaCl at 200°C using 100 – 200 µm particle size ...........................................110
Figure 5.21  Comparison of granite dissolution profile in water and pH 4 buffer solution at 200°C in autoclave .................................................................112
Figure 5.22  Comparison of granite dissolution profile in water and pH 7 buffer solution at 200°C in autoclave .................................................................113
Figure 5.23  Comparison of granite dissolution profile in water and pH 10 buffer solution at 200°C in autoclave

Figure 5.24  Comparison of granite dissolution profile in water and pH 13 buffer solution at 200°C in autoclave

Figure 5.25  Comparison of dissolution rate constants in pH buffer solutions at 200°C in autoclave using geometric surface area basis

Figure 5.26  Comparison of dissolution rate constants in pH buffer solutions at 200°C in autoclave using BET surface area basis

Figure 5.27  Air-cooled flow loop

Figure 5.28  Temperature profile of the hydrothermal cell (water)

Figure 5.29  Temperature profile of the hydrothermal cell (NaCl solution)

Figure 5.30  Photo of sample baskets (from left to right: 0.7 g sample, 3 g sample, 7 g sample)

Figure 5.31  Updated geothermal cell

Figure 5.32  Geothermal flow-through cell

Figure 5.33  Dissolution in water with varying sample weight

Figure 5.34  Concentration of dissolved Na, K, and Al in water with 0.7 g granite sample

Figure 5.35  Concentration of dissolved Na, K, and Al in water with 3 g granite sample

Figure 5.36  Concentration of dissolved Na, K, and Al in water with 7 g granite sample

Figure 5.37  Dissolution in 250 ppm NaCl with varying sample weight

Figure 5.38  Concentration of dissolved K and Al in 250 ppm NaCl solution with 0.7 g granite sample

Figure 5.39  Concentration of dissolved K and Al in 250 ppm NaCl solution with 3 g granite sample

Figure 5.40  Concentration of dissolved K and Al in 250 ppm NaCl solution with 7 g granite sample

Figure 5.41  Comparison between dissolution in water and NaCl with 0.7 g granite sample
Figure 5.42  Comparison between dissolution in water and NaCl with 3 g granite sample .................................................................138
Figure 5.43  Comparison between dissolution in water and NaCl with 7 g granite sample .................................................................138
Figure 5.44  SEM images (0.7 g sample) of the rock sample surface: (a) starting rock (b) after 4 days experiment (c) after 7 days experiment (d) after 14 days experiment (e) after 28 days. Experiments performed in pure water .............................................142
Figure 5.45  Backscatter SEM images of the granite (0.7 g sample) showing comparison of etch pits on albite and K-feldspar phase: (a) after 1 day experiment (b) after 14 days experiment (c) after 28 days. Experiments performed in pure water ..........143
Figure 5.46  SEM images (0.7 g sample) of the rock sample surface: (a) after 1 day experiment (b) after 28 days. Experiments performed in 250 ppm NaCl .................................................................144
Figure 5.47  Backscatter SEM images of the granite (0.7 g sample) showing comparison of etch pits on albite and K-feldspar phase: (a) after 3 days experiment (b) after 30 days. Experiments performed in 250 ppm NaCl solution .................................................................145
Figure 5.48  ED spectrum for quartz .........................................................................................................................................................146
Figure 5.49  X-ray diffraction results for 14 and 28 days rock-fluid interaction periods for 0.7 g granite sample ..............................................................................................................................................147
Figure 5.50  X-ray diffraction results for 14 and 28 days rock-fluid interaction periods for 3 g granite sample ..............................................................................................................................................147
Figure 5.51  X-ray diffraction results for 14 and 28 days rock-fluid interaction periods for 7 g granite sample ..............................................................................................................................................148
Figure 5.52  X-ray diffraction results for 14 and 28 days rock-fluid interaction periods in 250 ppm NaCl solution using 0.7 g granite sample ..............................................................................................................................................148
Figure 5.53  X-ray diffraction results for 14 and 28 days rock-fluid interaction periods in 250 ppm NaCl solution using 3 g granite sample ..............................................................................................................................................149
Figure 5.54  X-ray diffraction results for 14 and 28 days rock-fluid interaction periods in 250 ppm NaCl solution using 7 g granite sample ..............................................................................................................................................149
Figure 5.55  Comparison of cumulative silica concentration after 28 days replacing fluid ....151
Figure 5.56  Comparison of cumulative Na, K and Al concentration after 28 days replacing water.................................................................152
Figure 5.57  Comparison of cumulative K and Al concentration after 28 days replacing 250 NaCl solution ..........................................................................................................................152
Figure 5.58  SEM images of the granite surface (a) starting sample, (b) after 7 days, and (c) after 28 days replacing water.................................................................154
Figure 5.59  Backscatter SEM images of the granite showing (a) starting sample, (b) after 7 days interaction, and (c) 28 days replacing water.................................155
Figure 5.60  SEM images of the granite surface (a) after 7 days and (b) after 28 days replacing 250 ppm NaCl solution .................................................................156
Figure 5.61  Backscatter SEM images of the granite showing (a) after 7 days interaction and (b) 28 days replacing 250 ppm NaCl solution.................................156
Figure 5.62  High pressure flow though cell..........................................................................................................................157
Figure 5.63  Effect of pressure on silica dissolution................................................................................................................159
Figure 5.64  Silica precipitation on the diaphragm of a back-pressure regulator ..........160
Figure 6.1  Constraints on the initial system in React for granite dissolution at 250°C to obtain the reacted minerals individually........................172
Figure 6.2  Specified reactants and kinetic reactions at 250°C to obtain the reacted minerals individually..................................................................................................................172
Figure 6.3  Constraints on the initial system in React to obtain the equilibrium silica concentration at various temperatures .................................................173
Figure 6.4  Specified reactants in React to obtain the equilibrium silica concentration at various temperatures ..........................................................173
Figure 6.5  Equilibrium silica concentration from quartz dissolution at different temperature generated by GWB .................................................................174
Figure 6.6  Constraints on the initial system in React for granite dissolution at 250°C ....176
Figure 6.7  Specified reactants and kinetic reactions at 250°C for 0.7 g sample size ..........176
Figure 6.8  Specified reactants and kinetic reactions at 250°C for 3 g sample size ..........177
Figure 6.9  Specified reactants and kinetic reactions at 250°C for 7 g sample size ..........177
| Figure 6.10 | Concentrations of fluid components over time fitted using GWB to determine the reaction rate constant for 0.7 g sample size | 178 |
| Figure 6.11 | Concentrations of fluid components over time fitted using GWB to determine the reaction rate constant for 3 g sample size | 179 |
| Figure 6.12 | Concentrations of fluid components over time fitted using GWB to determine the reaction rate constant for 7 g sample size | 179 |
| Figure 6.13 | Silica concentrations from different sample sizes after data fitting using GWB | 180 |
| Figure 6.14 | Amount of various minerals dissolved in water using 0.7 g sample size | 181 |
| Figure 6.15 | Amount of various minerals dissolved in water using 3 g sample size | 181 |
| Figure 6.16 | Amount of various minerals dissolved in water using 7 g sample size | 182 |
| Figure 6.17 | Constraints on the initial system for silica precipitation from water | 184 |
| Figure 6.18 | Specified surface area and kinetic reactions for silica precipitation from water | 185 |
| Figure 6.19 | Changes in the amount of silica in water as the fluid cools | 185 |
| Figure 6.20 | Constraints on the initial system in React for silica precipitation from 250 ppm NaCl solution | 186 |
| Figure 6.21 | Changes in the amount of silica in 250 ppm NaCl as the fluid cools | 187 |
List of Tables

Table 2.1  Major geothermal producers ................................................................. 8
Table 2.2  Physical data from six EGS reservoirs ................................................... 11
Table 2.3  Physical data from six EGS reservoirs (cont.) ........................................... 12
Table 2.4  Fluid composition from six EGS reservoirs ............................................. 13
Table 2.5  Rate constants $k_\nu$ (mol/cm$^2$.s) for dissolution of silica minerals in water at various temperatures ................................................................. 39
Table 2.6  Compiled global activation energy ......................................................... 48
Table 3.1  Habanero 1 water analysis ................................................................. 63
Table 3.2  Habanero 1 water analysis (cont.) .......................................................... 64
Table 3.3  Habanero 1 gas analysis ..................................................................... 65
Table 3.4  Habanero 3 production well head condition ........................................... 65
Table 3.5  Habanero 3 gas analysis ..................................................................... 65
Table 3.6  Habanero 3 water analysis .................................................................. 66
Table 3.7  Habanero 3 gas analysis by AMDEL ................................................... 67
Table 3.8  Habanero 3 water analysis by AWQC .................................................. 67
Table 3.9  Quantitative XRD analysis using TOPAS ........................................... 71
Table 3.10 XRF analysis ..................................................................................... 71
Table 4.1  Particle size distribution ..................................................................... 77
Table 4.2  Densities of minerals ......................................................................... 78
Table 4.3  Specific surface area of Habanero 3 granite ($A_s^*$) ............................ 78
Table 5.1  Constants from curve fitting model using SigmaPlot for 100 – 200 µm sample size ................................................................. 91
Table 5.2  Constants from SigmaPlot in terms of (molal) and $k^#$ (s$^{-1}$) for 100 – 200 µm sample size ................................................................. 92
Table 5.3  Dissolution rate constants for four different temperatures based on geometric surface area ................................................................. 95
| Table 5.4 | Dissolution rate constants for four different temperatures based on BET surface area | 95 |
| Table 5.5 | Constants from SigmaPlot for various particle sizes at 200°C | 102 |
| Table 5.6 | Constants from SigmaPlot in terms of (molal) and $k^\#$ (s$^{-1}$) in water for various particle sizes at 200°C | 102 |
| Table 5.7 | Dissolution rate constant for various particle size distribution based on geometric surface area | 102 |
| Table 5.8 | Dissolution rate constant for various particle size distribution based on BET surface area | 102 |
| Table 5.9 | Constants from SigmaPlot for granite dissolution in water and NaCl solution at 200°C | 109 |
| Table 5.10 | Calculated equilibrium silica concentration (molal) and $k^\#$ (s$^{-1}$) in water and NaCl solution at 200°C | 109 |
| Table 5.11 | Granite dissolution rate constant in water and NaCl solution based on geometric surface area | 109 |
| Table 5.12 | Granite dissolution rate constant in water and NaCl solution based on BET surface area | 109 |
| Table 5.13 | Composition of pH buffers and calculated pH at 200°C | 111 |
| Table 5.14 | Constants from SigmaPlot for various pH | 115 |
| Table 5.15 | Calculated equilibrium silica concentration (molal) and $k^\#$ (s$^{-1}$) in water for various pH | 116 |
| Table 5.16 | Dissolution rate constant for various pH buffer solutions based on geometric surface area | 116 |
| Table 5.17 | Dissolution rate constant for various pH buffer solutions based on BET surface area | 116 |
| Table 5.18 | Data to calculate flow velocity in the hydrothermal cell (water) | 123 |
| Table 5.19 | Average values between thermocouples (water) | 124 |
| Table 5.20 | Data to calculate flow velocity in the hydrothermal cell (NaCl) | 124 |
| Table 5.21 | Average values between thermocouples (NaCl solution) | 124 |
Table 5.22 Various liquid/rock ratios ................................................................. 139
Table 5.23 Constants from SigmaPlot for various liquid/rock ratios (water) ......... 139
Table 5.24 Calculated equilibrium silica concentration (molal) and $k^w$ ($s^{-1}$) in water for various liquid/rock ratios ................................................................. 139
Table 5.25 Dissolution rate in water for various liquid/rock ratios based on geometric surface area ............................................................................................ 139
Table 5.26 Dissolution rate in water for various liquid/rock ratios based on BET surface area ................................................................................................. 140
Table 5.27 Constants from SigmaPlot for various liquid/rock ratios (250 ppm NaCl) ............... 140
Table 5.28 Calculated equilibrium silica concentration (molal) and $k^w$ ($s^{-1}$) in 250 ppm NaCl for various liquid/rock ratios ................................................................. 140
Table 5.29 Dissolution rate in 250 ppm NaCl for various liquid/rock ratios based on geometric surface area ............................................................................................ 140
Table 5.30 Dissolution rate in 250 ppm NaCl for various liquid/rock ratio based on BET surface area ................................................................................................. 141
Table 5.31 Dissolved granite (% wt) in pure water after 7 and 28 days ................. 151
Table 5.32 Dissolved granite (% wt) in 250 ppm NaCl after 7 and 28 days .......... 151
Table 5.33 $U_T/U_{TS}$ values for a range of Reynolds numbers ................................. 163
Table 5.34 Comparison of mass transfer rate and reaction rate constant for static system at different temperatures ................................................................. 164
Table 5.35 Comparison of mass transfer rate and reaction rate constant for static system with different particle size ................................................................. 164
Table 5.36 Comparison of mass transfer rate and reaction rate constant for flow-through system ................................................................................................. 164
Table 6.1 Rate of silica release from mineral used as basis .................................... 168
Table 6.2 Activation energy and pre-exponential factor values used for modelling .... 168
Table 6.3 Quartz dissolution rate constants calculated for various temperatures ....... 168
Table 6.4 Albite dissolution rate constants calculated for various temperatures ...... 169
Table 6.5 K-feldspar dissolution rate constants calculated for various temperatures .... 169
<table>
<thead>
<tr>
<th>Table</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Table 6.6</td>
<td>The composition of granite used in the model</td>
</tr>
<tr>
<td>Table 6.7</td>
<td>Log K calculated by Rxn for quartz, albite and K-feldspar</td>
</tr>
<tr>
<td>Table 6.8</td>
<td>Reacted minerals calculated by React</td>
</tr>
<tr>
<td>Table 6.9</td>
<td>Reaction rate constant values from literature and GWB modelling</td>
</tr>
<tr>
<td>Table 6.10</td>
<td>Component mass based on the calculated active surface area</td>
</tr>
<tr>
<td>Table A.1</td>
<td>160°C data with initial guesses of $C^\infty = 100$ and $k^# = 0.001$</td>
</tr>
<tr>
<td>Table A.2</td>
<td>Summary of the Gauss-Newton result with the least square of the residual</td>
</tr>
<tr>
<td>Table B.1</td>
<td>Initial sample used and pH after experiment – Run I</td>
</tr>
<tr>
<td>Table B.2</td>
<td>Initial sample used and pH after experiment – Run II</td>
</tr>
<tr>
<td>Table B.3</td>
<td>Experimental results showing equilibrium reactive silica concentration at different temperatures</td>
</tr>
<tr>
<td>Table B.4</td>
<td>Experimental results showing equilibrium silica concentration (ICP-OES) at different temperatures</td>
</tr>
<tr>
<td>Table B.5</td>
<td>Dissolved major cation data for various temperatures in pure water – Run I</td>
</tr>
<tr>
<td>Table B.6</td>
<td>Dissolved major cation data for various temperatures in pure water – Run II</td>
</tr>
<tr>
<td>Table B.7</td>
<td>Average Dissolved major cation data for various temperature in pure water</td>
</tr>
<tr>
<td>Table B.8</td>
<td>Standard deviation cation data for various temperature in pure water</td>
</tr>
<tr>
<td>Table B.9</td>
<td>Initial sample used and pH after experiment (160°C) – Run I</td>
</tr>
<tr>
<td>Table B.10</td>
<td>Initial sample used and pH after experiment (160°C) – Run I</td>
</tr>
<tr>
<td>Table B.11</td>
<td>Reactive silica data in pure water at 160°C</td>
</tr>
<tr>
<td>Table B.12</td>
<td>Dissolved major cation data in pure water at 160°C – Run I</td>
</tr>
<tr>
<td>Table B.13</td>
<td>Dissolved major cation data in pure water at 160°C – Run II</td>
</tr>
<tr>
<td>Table B.14</td>
<td>Average dissolved major cation data in pure water at 160°C</td>
</tr>
<tr>
<td>Table B.15</td>
<td>Standard deviation dissolved major cation data in pure water at 160°C</td>
</tr>
<tr>
<td>Table B.16</td>
<td>Initial sample used and pH after experiment (170°C) – Run I</td>
</tr>
<tr>
<td>Table B.17</td>
<td>Initial sample used and pH after experiment (170°C) – Run I</td>
</tr>
<tr>
<td>Table B.18</td>
<td>Reactive silica data in pure water at 170°C</td>
</tr>
<tr>
<td>Table B.19</td>
<td>Dissolved major cation data in pure water at 170°C – Run I</td>
</tr>
<tr>
<td>Table B.20</td>
<td>Dissolved major cation data in pure water at 170°C – Run II .................................. 231</td>
</tr>
<tr>
<td>Table B.21</td>
<td>Average dissolved major cation data in pure water at 170°C ........................................ 232</td>
</tr>
<tr>
<td>Table B.22</td>
<td>Standard deviation dissolved major cation data in pure water at 170°C ......................... 232</td>
</tr>
<tr>
<td>Table B.23</td>
<td>Initial sample used and pH after experiment (200°C) – Run I .................................... 233</td>
</tr>
<tr>
<td>Table B.24</td>
<td>Initial sample used and pH after experiment (200°C) – Run II .................................. 233</td>
</tr>
<tr>
<td>Table B.25</td>
<td>Reactive silica data in pure water at 200°C ................................................................. 234</td>
</tr>
<tr>
<td>Table B.26</td>
<td>Dissolved major cation data in pure water at 200°C – Run I ........................................ 234</td>
</tr>
<tr>
<td>Table B.27</td>
<td>Dissolved major cation data in pure water at 200°C – Run II .................................... 235</td>
</tr>
<tr>
<td>Table B.28</td>
<td>Average dissolved major cation data in pure water at 200°C ...................................... 235</td>
</tr>
<tr>
<td>Table B.29</td>
<td>Standard deviation dissolved major cation data in pure water at 200°C ....................... 235</td>
</tr>
<tr>
<td>Table B.30</td>
<td>Initial sample used and pH after experiment (220°C) – Run I .................................... 236</td>
</tr>
<tr>
<td>Table B.31</td>
<td>Initial sample used and pH after experiment (220°C) – Run I .................................... 237</td>
</tr>
<tr>
<td>Table B.32</td>
<td>Reactive silica data in pure water at 220°C ................................................................. 237</td>
</tr>
<tr>
<td>Table B.33</td>
<td>Dissolved major cation data in pure water at 220°C – Run I ........................................ 238</td>
</tr>
<tr>
<td>Table B.34</td>
<td>Dissolved major cation data in pure water at 220°C – Run II .................................... 238</td>
</tr>
<tr>
<td>Table B.35</td>
<td>Average dissolved major cation data in pure water at 220°C ...................................... 239</td>
</tr>
<tr>
<td>Table B.36</td>
<td>Standard deviation dissolved major cation data in pure water at 220°C ....................... 239</td>
</tr>
<tr>
<td>Table B.37</td>
<td>Initial sample used and pH after experiment (40 – 60 µm) – Run I ................................ 240</td>
</tr>
<tr>
<td>Table B.38</td>
<td>Initial sample used and pH after experiment (40 – 60 µm) – Run II ................................ 241</td>
</tr>
<tr>
<td>Table B.39</td>
<td>Reactive silica data in pure water (40 – 60 µm) at 200°C ............................................ 241</td>
</tr>
<tr>
<td>Table B.40</td>
<td>Dissolved major cation data in pure water (40 – 60 µm) – Run I .................................. 242</td>
</tr>
<tr>
<td>Table B.41</td>
<td>Dissolved major cation data in pure water (40 – 60 µm) – Run II .................................. 242</td>
</tr>
<tr>
<td>Table B.42</td>
<td>Average dissolved major cation data in pure water (40 – 60 µm) ................................ 243</td>
</tr>
<tr>
<td>Table B.43</td>
<td>Standard deviation dissolved major cation data in pure water (40 – 60 µm) ................ 243</td>
</tr>
<tr>
<td>Table B.44</td>
<td>Initial sample used and pH after experiment (200 – 400 µm) – Run I ......................... 244</td>
</tr>
<tr>
<td>Table B.45</td>
<td>Initial sample used and pH after experiment (200 – 400 µm) – Run II ......................... 244</td>
</tr>
<tr>
<td>Table B.46</td>
<td>Reactive silica data in pure water (200 – 400 µm) at 200°C ........................................ 245</td>
</tr>
<tr>
<td>Table B.47</td>
<td>Dissolved major cation data in pure water (200 – 400 µm) – Run I ............................ 245</td>
</tr>
<tr>
<td>Table B.48</td>
<td>Dissolved major cation data in pure water (200 – 400 µm) – Run II ............................ 246</td>
</tr>
</tbody>
</table>
Table B.49  Average dissolved major cation data in pure water (200 – 400 µm) .................. 246
Table B.50  Standard deviation dissolved major cation data in pure water (200 – 400 µm) ...... ......................................................................................................................................................................................... 247
Table B.51  Initial sample used and pH after experiment in 250 ppm NaCl at 200°C – Run I ..... .................................................................................................................................................................................................................................................. 247
Table B.52  Initial sample used and pH after experiment in 250 ppm NaCl at 200°C – Run II .................................................................................................................................................................................................................................................. 248
Table B.53  Reactive silica data in 250 ppm NaCl at 200°C.................................................... 248
Table B.54  Dissolved major cation data in 250 ppm NaCl at 200°C – Run I......................... 249
Table B.55  Dissolved major cation data in 250 ppm NaCl at 200°C – Run II......................... 249
Table B.56  Average dissolved major cation data in 250 ppm NaCl at 200°C......................... 250
Table B.57  Standard deviation dissolved major cation data in 250 ppm NaCl at 200°C ...... 250
Table B.58  Initial sample used and pH after experiment in pH 4 buffer – Run I ................. 251
Table B.59  Initial sample used and pH after experiment in pH 4 buffer – Run II ................. 252
Table B.60  Reactive silica data in pH 4 buffer at 200°C ....................................................... 252
Table B.61  Initial sample used and pH after experiment in pH 7 buffer – Run I .................. 253
Table B.62  Initial sample used and pH after experiment in pH 7 buffer – Run II .................. 253
Table B.63  Reactive silica data in pH 7 buffer at 200°C ....................................................... 254
Table B.64  Initial sample used and pH after experiment in pH 10 buffer – Run I ................ 254
Table B.65  Initial sample used and pH after experiment in pH 10 buffer – Run II ............... 255
Table B.66  Reactive silica data in pH 10 buffer at 200°C ....................................................... 255
Table B.67  Initial sample used and pH after experiment in pH 13 buffer – Run I ................ 256
Table B.68  Initial sample used and pH after experiment in pH 13 buffer – Run I ............... 256
Table B.69  Reactive silica data in pH 13 buffer at 200°C ....................................................... 257
Table C.1  Experimental data using 0.7 g sample in pure water - Run I .............................. 258
Table C.2  Experimental data using 0.7 g sample in pure water – Run II............................. 259
Table C.3  Average values from Run I and Run II for 0.7 g sample in pure water................. 259
Table C.4  Reactive silica data for 0.7 g sample in pure water ............................................ 260
Table C.5  Dissolved major cation data for 0.7 g sample in pure water – Run I ................. 260
Table C.6  Dissolved major cation data for 0.7 g sample in pure water – Run II .................. 261
Table C.7  Average Dissolved major cation data for 0.7 g sample in pure water .................. 261
Table C.8  Standard deviation cation data for 0.7 g sample in pure water .......................... 262
Table C.9  Experimental data using 3 g sample in pure water – Run I .............................. 262
Table C.10 Experimental data using 3 g sample in pure water – Run II .............................. 262
Table C.11 Average values from Run I and Run II for 3 g sample in pure water ................. 263
Table C.12 Reactive silica data for 3 g sample in pure water ............................................ 264
Table C.13 Dissolved major cation data for 3 g sample in pure water – Run I ................... 264
Table C.14 Dissolved major cation data for 3 g sample in pure water – Run II ................... 265
Table C.15 Average dissolved major cation data for 3 g sample in pure water .................. 265
Table C.16 Standard deviation major cation data for 3 g sample in pure water ................... 266
Table C.17 Experimental data using 7 g sample in pure water – Run I .............................. 266
Table C.18 Experimental data using 7 g sample in pure water – Run II .............................. 267
Table C.19 Average values from Run I and Run II for 7 g sample in pure water ................. 267
Table C.20 Reactive silica data for 7 g sample in pure water ............................................ 268
Table C.21 Dissolved major cation data for 7 g sample in pure water – Run I ................... 268
Table C.22 Dissolved major cation data for 7 g sample in pure water – Run II ................... 269
Table C.23 Average dissolved major cation data for 7 g sample in pure water .................. 269
Table C.24 Standard deviation major cation data for 7 g sample in pure water .................. 270
Table C.25 Experimental data using 0.7 g sample in 250 ppm NaCl solution – Run I ........ 271
Table C.26 Experimental data using 0.7 g sample in 250 ppm NaCl solution – Run II .......... 271
Table C.27 Average values from Run I and Run II for 0.7 g sample in 250 ppm NaCl .......... 272
Table C.28 Reactive silica data for 0.7 g sample in 250 ppm NaCl solution ......................... 272
Table C.29 Dissolved major cation data for 0.7 g sample in 250 ppm NaCl solution – Run I ........ .......................................................... 273
.......................................................... 273
Table C.30 Dissolved major cation data for 0.7 g sample in 250 ppm NaCl solution – Run II .......................................................... 273
.......................................................... 273
Table C.31 Average dissolved major cation data for 0.7 g sample in 250 ppm NaCl solution .......................................................... 274
Table C.32  Standard deviation major cation data for 0.7 g sample in 250 ppm NaCl solution ................................................................. 274
Table C.33  Experimental data using 3 g sample in 250 ppm NaCl – Run I................................. 275
Table C.34  Experimental data using 3 g sample in 250 ppm NaCl – Run II................................. 275
Table C.35  Average values from Run I and Run II for 3 g sample in 250 ppm NaCl.............. 276
Table C.36  Reactive silica data for 3 g sample in 250 ppm NaCl solution................................. 276
Table C.37  Dissolved major cation data for 3 g sample in 250 ppm NaCl solution – Run I........ 
................................................................................................................................................................. 277
Table C.38  Dissolved major cation data for 3 g sample in 250 ppm NaCl solution – Run II ...... 
................................................................................................................................................................. 277
Table C.39  Average dissolved major cation data for 3 g sample in 250 ppm NaCl solution...... 
................................................................................................................................................................. 278
Table C.40  Standard deviation major cation data for 3 g sample in 250 ppm NaCl solution ..... 
................................................................................................................................................................. 278
Table C.41  Experimental data using 7 g sample in 250 ppm NaCl solution – Run I.............. 279
Table C.42  Experimental data using 7 g sample in 250 ppm NaCl solution – Run II.............. 279
Table C.43  Average values from Run I and Run II for 7 g sample in 250 ppm NaCl.............. 280
Table C.44  Reactive silica data for 7 g sample in 250 ppm NaCl solution................................. 280
Table C.45  Dissolved major cation data for 7 g sample in 250 ppm NaCl solution – Run I....... 
................................................................................................................................................................. 281
Table C.46  Dissolved major cation data for 7 g sample in 250 ppm NaCl solution – Run II ...... 
................................................................................................................................................................. 281
Table C.47  Average dissolved major cation data for 7 g sample in 250 ppm NaCl solution...... 
................................................................................................................................................................. 282
Table C.48  Standard deviation major cation data for 7 g sample in 250 ppm NaCl solution ..... 
................................................................................................................................................................. 282
Table D.1  Silica concentration in pure water and 250 ppm NaCl solution after 7 days...... 283
Table D.2  Na, K and Al concentration in pure water after 7 days ....................................... 283
Table D.3  K and Al concentration in 250 ppm NaCl solution after 7 days ....................... 284
Table D.4  Silica concentration in pure water and 250 ppm NaCl solution after 28 days ... 284
Table D.5  Silica concentration in pure water and 250 ppm NaCl solution after 28 days (cont.) .................................................................................................................. 285
Table D.6  Na, K and Al concentration in pure water after 28 days ........................................ 285
Table D.7  Na, K and Al concentration in pure water (cont.) after 28 days (cont.) ............... 286
Table D.8  K and Al concentration in 250 ppm NaCl solution after 28 days ......................... 287
Table D.9  K and Al concentration in 250 ppm NaCl solution (cont.) after 28 days .......... 288
Table E.1  Sample information after 6 hours interaction at different pressures (Run I) ..... 289
Table E.2  Sample information after 6 hours interaction at different pressures (Run II) ..... 289
Table E.3  Silica concentration after 6 hours interaction at 40 bar ..................................... 290
Table E.4  Silica concentration after 6 hours interaction at 100 bar .................................... 290
Table E.5  Silica concentration after 6 hours interaction at 200 bar .................................... 290
Table F.1  Mass transfer coefficient data for static system ................................................. 292
Table G.1  Darby’s bore water analysis March 2007 ............................................................ 295
Table G.2  Darby’s bore water analysis August 2008 .......................................................... 296
Table G.3  Darby’s bore water analysis August 2008 (cont.) ............................................. 297
Table G.4  Darby’s bore water analysis August 2008 (cont.) ............................................. 298
Table G.5  Darby’s bore water analysis November 2008 ..................................................... 299
Table G.6  Darby’s bore water analysis November 2008 (cont.) ......................................... 300