

## FLUID-ROCK INTERACTIONS IN DEEP-SEATED GEOTHERMAL RESERVOIRS

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### ABSTRACT

*Extending the depth of hydrothermal resources, and development of engineered reservoirs for Hot Dry Rock geothermal heat extraction, complement 'conventional' geothermal energy utilization, yet water-rock interaction processes and the physical character of induced/natural fracture systems in Deep-Seated Geothermal Reservoirs (DSGR) are poorly understood. Dissolution of granite and quartz, with pure water, has been investigated at sub- and supercritical conditions to evaluate deep geothermal reservoir dynamics. In terms of dissolution behavior of granite and quartz, the supercritical region may be subdivided into two apparent phases, comprising 'liquid-like' and 'vapor-like' regions. The solvent properties of supercritical fluid in the liquid-like region are similar to those of subcritical water, whilst dissolution reactions in the vapor-like supercritical state are weak.*

*SEM-Cathodoluminescence (SEM-CL) imaging techniques reveal chronological relationships between secondary (vein/replacement) quartz and (primary) rock-forming minerals, which aid differentiation of (multiple) hydrothermal events, and interpretation of physico-chemical changes in the active Te Kopia (New Zealand) geothermal system. SEM-CL, integrated with fluid inclusion/petrographic data, "fingerprints" quartz from specific alteration events, constrains the timing of fluid inclusion trapping and nature of fluid migration, which is important for understanding the pressure, temperature and chemical evolution of the hydrothermal system.*

### 1.0 INTRODUCTION

Utilization of active geothermal systems for electricity generation is limited by (a) resource temperatures or field pressures; (b) a decline during production in reservoir mass output,

temperature etc.; or (c) environmental / cultural concerns. Extending the depth of hydrothermal resources by developing engineered geothermal reservoirs complement 'conventional' resource utilization. At Kakkonda (Japan), well WD-1a encountered temperature / pressure conditions that exceeded the critical point for water (374°C, 22MPa), with the temperature at ~3.5km depth being >500°C (Ikeuchi *et al.*, 1996). Few other wells have been drilled to >370°C (and they encountered little permeability), so fluid-rock interactions / character of fracture systems in deep-seated geothermal reservoirs (DSGR) are poorly understood.

Granite is a likely reservoir rock for DSGR systems. Fluid-rock/quartz interactions are important to understand, because precipitation of quartz in deep parts of a hydrothermal system may decrease permeability to such an extent that there is little fluid convection. Researchers have attempted to model deep (>3-4km) geothermal reservoir conditions, and the mechanisms for crack generation (e.g. Willis-Richards and Wallroth, 1995), and studied fluid-rock interactions using a variety of batch-type vessels over a range of high temperature-pressure conditions (e.g. Savage *et al.*, 1992; Bignall *et al.*, 2000a), yet more information is required concerning multi-component (chemical) reactions in a dynamic (flow) regime.

Silica solubility is a complex function of temperature and fluid density (White and Mroczek, 1998). At saturated vapour pressure, the dissociation capacity of hot water is directly related to ion product ( $K_w$ ), which has a maximum value around 350°C. Dissociation capacity decreases with lower ion product, above ~360°C, producing conditions favourable for mineral (i.e. quartz) deposition. Vapour pressure decreases with an increase in salt concentration. Such factors influence interaction processes, and the degree to which dissolution-deposition reactions may proceed. Here, we

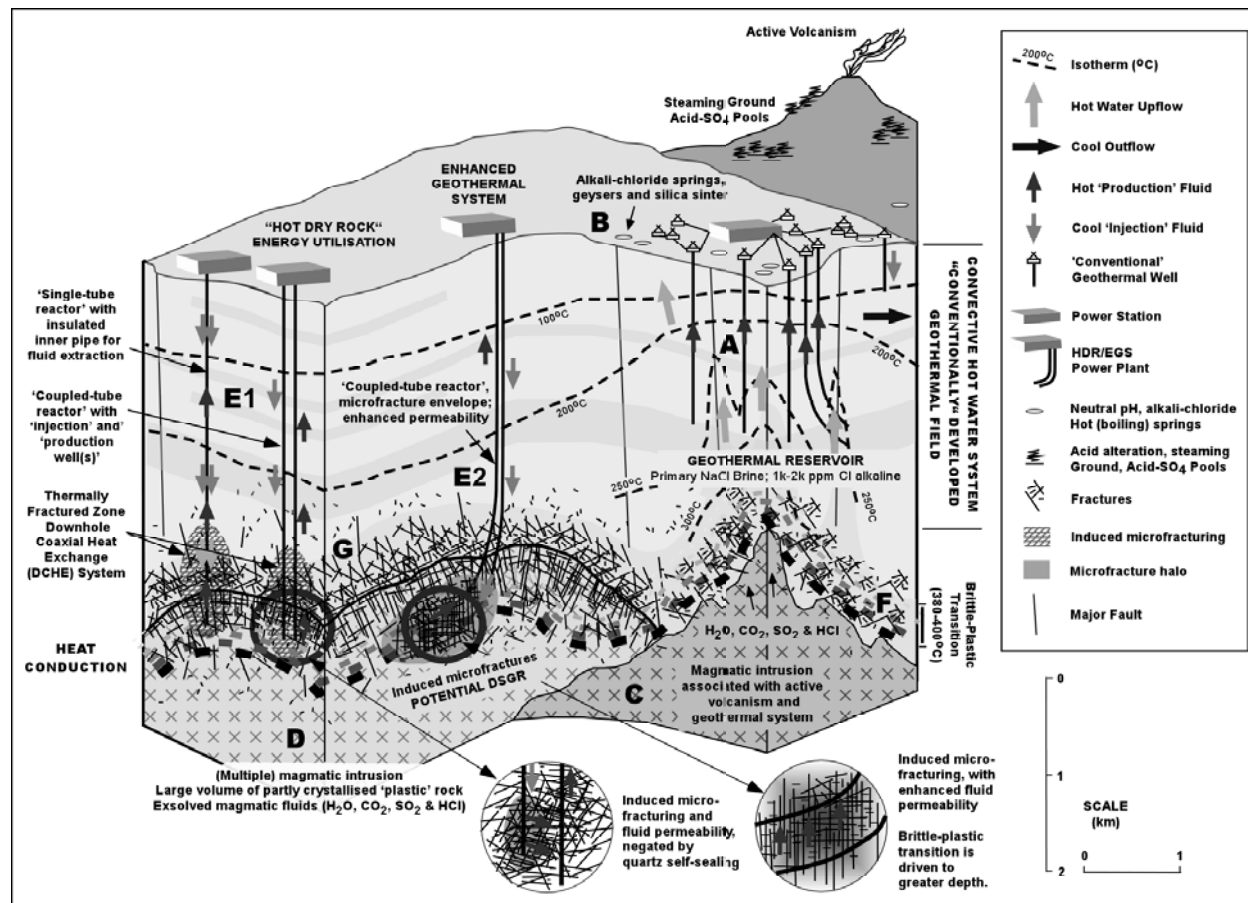


Figure 1. Schematic representation of the development and utilisation of a hypothetical Deep-Seated Geothermal Reservoir (DSGR).

detail findings of our experimental programmes, which aim to deduce granite/mineral dissolution and fracture characteristics up to supercritical temperature-pressure conditions, in a granite-hosted DSGR.

## 2.0 SCHEMATIC MODEL OF DSGR

Figure 1 shows a schematic magmatic-hydrothermal system, utilized for “conventional” electricity generation, or as a DSGR. In most developed geothermal systems, the main upflow zone (at “A”) or reservoir is tapped by <2.5-3km wells, sited to intersect permeable structures (which supply fluids to surface features, “B”), with a convective region overlying a deeper conductive zone. Multiple magmatic intrusions (“C”) are the ‘heat source’ for many high-temperature geothermal systems, (e.g. in the Philippines; Reyes, 1990), and therefore potential host rocks for artificial geothermal energy extraction (“D”; either using an injection

well(s)/production well(s) approach (“E1”), or by creation of a network (‘cloud’) of microfractures (“E2”) that extend the depth of the reservoir to high temperature/pressure conditions, below the brittle-plastic transition at ~370-400°C in granitoid rocks; “F”). The core (“G”) of the intrusion is partly crystallized, and exsolved fluids that accumulate near the top of the pluton may produce fracturing, brecciation and/or expulsion of fluid into overlying rocks. Countering processes that increase permeability is self-sealing by quartz and other minerals (in veins and breccia matrix; e.g. at “H”).

## 3.0 DISSOLUTION/FRACTURING EXPERIMENTS

### 3.1 Methodology

Hydrothermal dissolution/fracturing experiments were conducted using distilled water, with (30 x 15 x 3 mm, impurity-free) artificial quartz, and

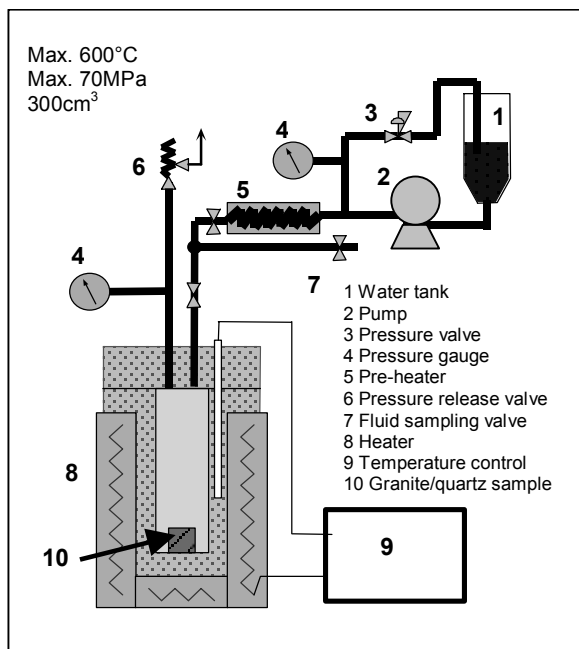


Figure 2. Schematic illustration of our batch-type autoclave system.

(polished, 1 cm<sup>3</sup> cube) medium grained lidate Granite (Fukushima Prefecture, Japan). The modal composition of lidate Granite is 37% quartz, 34% plagioclase and 22% K-feldspar (plus subordinate biotite, hornblende and accessory minerals).

Our batch autoclave has a main body of hastelloy-C (other parts from SUS-316), and incorporates a hot-water injection unit, which allows solution to be injected into the autoclave to maintain pressure conditions (Fig. 2). The autoclave has an effective volume of 30cc. The maximum temperature and pressure in our experiments was 600°C and 60 MPa respectively, with a heating rate of 10°C/min. Experiments were undertaken for up to 120 hours, and then slowly cooled to room temperature (to avoid quench damage). Reacted solutions were analysed by ICP (HITACHI P4000), to determine reaction rates, with sample weight change, petrology and morphological characteristics also examined.

### 3.2 Results

#### *Dissolution Experiments*

Quartz solubility is a complex function of temperature and fluid density, and dependent on

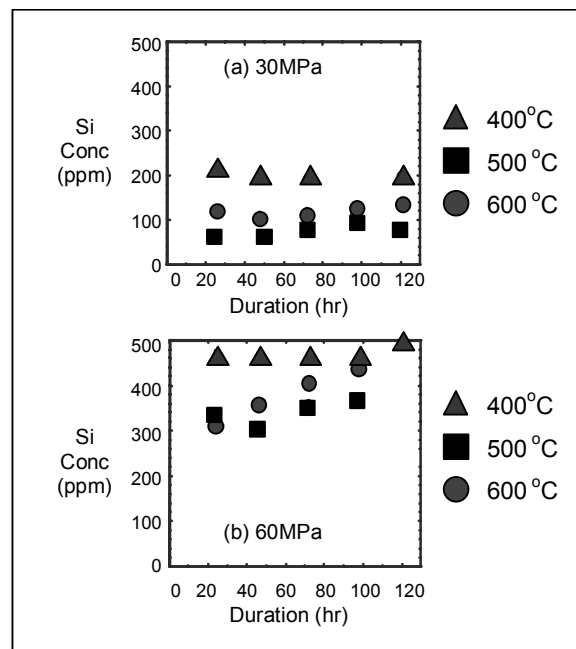


Figure 3. Plot of Si concentration and reaction duration, for quartz dissolution at: (a) 30 MPa, and (b) 60 MPa.

Kw in pure water under saturated vapour pressure conditions. The highest Kw (value) for a given pressure (under 100MPa) occurs in the range of 300°C - 400°C. In our experiments, the Si concentration of reacted (residual) fluids, after quartz dissolution in pure water, at 30 MPa and 60 MPa, for 400, 500 and 600°C is shown in Figure 3. The high Si concentration of fluid demonstrated at 30MPa/400°C (compared to 500°C and 600°C) is an effect of a greater ionic product. The Si concentration of reacted fluids in our 60MPa/600°C experiments rapidly reaches equilibrium, whilst Si concentrations for reaction experiments undertaken at 400°C and 500°C take >100 hours to reach an equilibrium condition.

The rate of quartz dissolution is greatest when specific volume of the hydrothermal (reactive) fluid is small, and dissolution rate decreases with an increase in specific volume, so that the rate expression of quartz dissolution, can be described by following n-th order kinetic equation:

$$r = k V^n$$

where  $r$  is dissolution rate,  $k$  is apparent kinetic constant,  $V$  is specific volume, and  $n$  is apparent reaction order. At low specific volume, the slope

of lines approximating change of dissolution rate is minus unity (Fig. 4), which means dissolution rate is proportional to water density. At higher specific volume slopes are steeper, indicating that the fluid loses its capability as a solvent. The dissolution of granite in our autoclave experiments is represented by Si-concentration of reacted fluid (Fig. 5). High Si-concentrations were determined at conditions less than the critical temperature for pure water, but low Si-concentrations are indicated at high temperature/low pressure conditions. Water-rock interactions (i.e. granite dissolution) occur from 300°C to 450°C, but apparently weaker in the high temperature regime.

**Quartz/Granite Fracturing**

Batch-autoclave experiments were conducted from 250°C to 600°C; 0.1MPa to 60MPa; for 24 to 120 hours in a water-quartz/granite system, to deduce the range of T-P conditions at which quartz and granite fracturing might occur in a DSGR system (Hirano et al., 2002). We found, that no fracturing of quartz occurred under dry conditions (0.1 MPa, even up to 600°C), but quartz was fractured in the presence of water at 3MPa/600°C. The boundary of quartz fracturing is coincident with the subcritical, vapour-phase for pure water (Fig. 6). At elevated temperatures the phase boundary conditions differentiating  $\alpha$ - and  $\beta$ -quartz approximates regions of unfractured and fractured quartz. The temperature-pressure conditions conducive for granite fracturing are shown in Figure 6. Petrographic examination reveals that the fractures predominantly occur in quartz, and not in feldspar or other primary mineral phases.

**Flow Reactor Experiments**

Experiments were undertaken using a flow reactor autoclave, that allows fluid-rock interactions at conditions up to 600°C and 60MPa, and 0-15cm<sup>3</sup>/m flow rate, in three 1-m Hastelloy-C autoclave units (Fig. 7). In the latter flow-reactor experiments, distilled water is pre-heated and injected into the pressurized autoclave system; with the temperature in each reactor tube controlled by independently operated heaters. In our 350°C (inlet)/30MPa experiment, granite is leached near the ‘inlet’ of Reactor A (where temperatures are subcritical), with dissolution of quartz and feldspar. In Reactor B, the internal temperature is set at 350°C (inlet) and 420°C (outlet), and in this

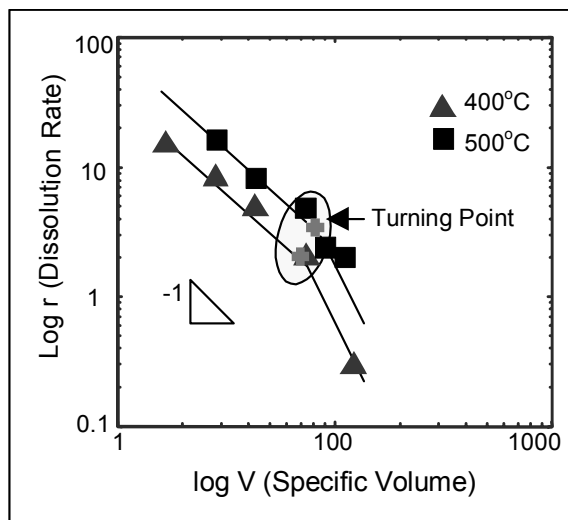


Figure 4. Relationship between specific volume and rate of quartz dissolution

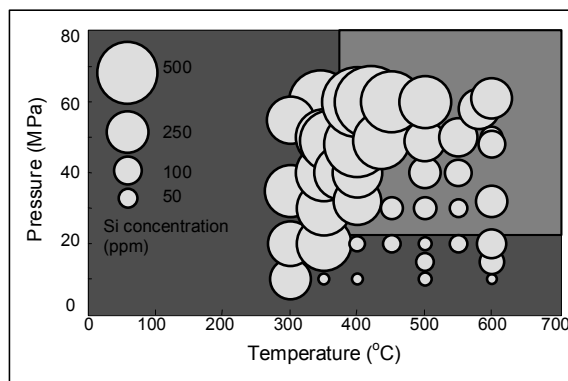


Figure 5. Dissolution rate of lidate Granite, indicated by Si- concentration in residual fluid (from batch autoclave experiments).

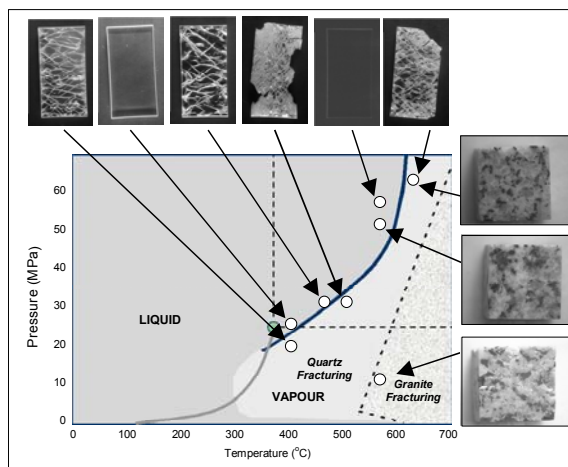


Figure 6. Phase relationships for pure water, showing the temperature-pressure regions identified in our batch-autoclave experiments that are conducive for quartz- and granite-fracturing.

region there is a strong weight gain, with precipitation of quartz, lesser albite and sericite. The occurrence of quartz, which has been examined by SEM-CL, is attributed to retrograde quartz solubility at about 380-400°C, (after Fournier, 1985), with quartz precipitation occurring upon heating of the ‘hydrothermal’ fluid.

#### 4.0 SCANNING ELECTRON MICROSCOPY - CATHODOLUMINESCENCE (SEM-CL) STUDY

Mineral non-stoichiometry, lattice defects or incorporation of trace elements into a crystal structure can generate CL of varying wavelength and intensity, which are useful for interpreting hydrothermal processes (Götze *et al.* 2001). SEM-CL has applications in alteration studies, by revealing CL-distinct hydrothermal quartz overprinting, or vein formation chronology. We used SEM-CL to characterise precipitation textures in quartz from our flow autoclave experiments (Fig. 8; Bignall, *in prep*). However, in this paper, we highlight use of SEM-CL to characterise precipitation and overgrowth textures in hydrothermal quartz from the Te Kopia geothermal field (New Zealand; after Bignall *et al.*, 2002).

At Te Kopia, regional faulting has exposed rocks that were once >300 m below the ground surface, and influenced field hydrology, surface manifestations and hydrothermal alteration (Bignall and Browne, 1994), with overprinting of quartz-illite-adularia assemblages by amorphous silica, kaolinite, cristobalite and/or alunite (Bignall, 1994). Sub- to euhedral quartz crystals (up to 4.5 cm long) occur on the scarp and contain liquid-rich fluid inclusions that homogenize at  $205 \pm 15^\circ\text{C}$  (<0.4 wt% NaCl equivalence), which grew at depths of >170 m below ground surface. Electron back-scattered and transmitted light images show the quartz are fractured and partly covered by crusty, amorphous silica, but reveals no other distinguishing features, whilst SEM-CL indicates the quartz crystals have a complex history of formation.

#### 4.1 Methodology

Carbon coated, 100  $\mu\text{m}$ -thick, double polished sections were prepared of the quartz, and examined by SEM-CL using the Department of

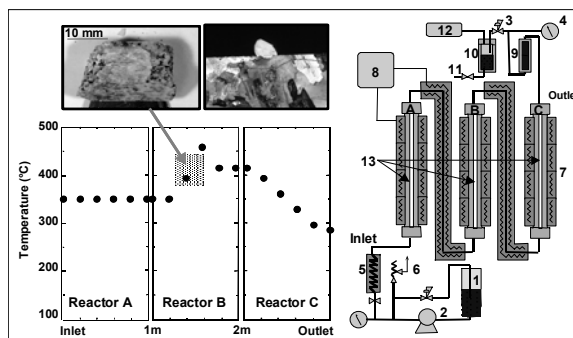


Figure 7. Flow reactor/autoclave system for fluid-rock interaction experiments, up to 600°C/60MPa.

Key: 1 water tank; 2 pump; 3 pressure control valve; 4 pressure gauge; 5 pre-heater; 6 pressure release valve; 7 heater; 8 temperature control unit; 9 separator; 10 fluid sampling chamber; 11 gas chromatography; 12 sampling valve; 13 granite

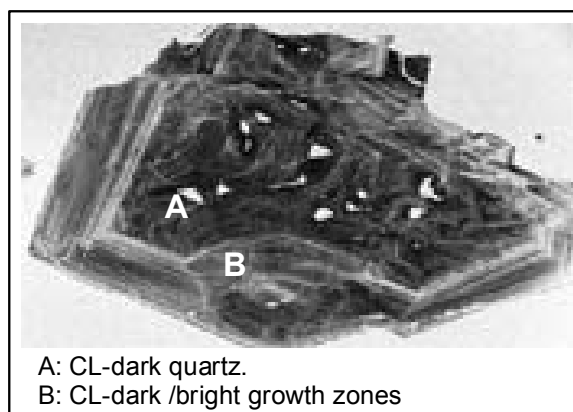


Figure 8. SEM-CL image of zoned quartz, obtained from our flow-reactor experiment (see Fig. 7).

Geoscience and Technology’s Hitachi-S2460N SEM-EDX instrument, which is equipped with an Oxford Mini-CL detector and photomultiplier. The quartz thick-sections were analyzed at 15 kv with a beam current set from 10-30 nA (to obtain optimal contrast in observed luminosity), which produces grey-scale “CL-dark”, “CL-grey” or “CL-bright” images.

#### 4.2 Results

SEM-CL imaging, combined with fluid inclusion thermometry, reveals evidence of multi-stage crystal growth, at different temperature-pressure conditions. Most of the quartz has CL-dark luminescence, but in places precipitated

concentric CL-dark and CL-bright growth zones of variable intensity (Fig. 9). SEM-CL shows that the crystals are fractured and partly dissolved along grain boundaries, with subsequent quartz deposition into open space.

The crystal core is composed of CL-dark quartz (containing fluid inclusions with  $T_h$  values of  $215 \pm 5^\circ\text{C}$ ), which is inferred to have grown within the Te Kopia geothermal reservoir (>320 m below the present ground surface), under stable physico-chemical conditions. The edge of the crystal is composed of alternating micron-scale bands of CL-dark and CL-bright quartz (containing fluid inclusions with  $T_h$  values of  $210 \pm 50^\circ\text{C}$ ), which precipitated during a later period of crystal growth, coinciding with fault uplift, with repeated movement on the Paeroa Fault providing pathways for pulses of hot reservoir fluids to move through the system, leading to precipitation of CL-bright quartz from cooling reservoir fluids, and/or inflow of cool condensate. The physical fracturing that took place underlines the complex history of repeated microfracturing, and resealing (through quartz deposition) that occurred during crystal growth.

As well as crystal *growth* textures, we find SEM-CL evidence of partly *dissolved* CL-dark quartz, with corroded crystal edges, as a result of the quartz being out of chemical equilibrium with reservoir fluids, and subsequent overprinting by CL-bright quartz. Low pH fluids in the near-surface setting have also acted to round the quartz crystals, and coat them with CL-grey amorphous “silica residue”.

## 5.0 DISCUSSION

Fluid-rock interactions in hydrothermal systems are intrinsically linked to water-rock ratio, the surface area of permeable channels and the nature of those surfaces, and chemical composition of the reactive fluid. The character of hydrothermal solutions, however, and the nature of their fluid-rock interactions also vary according to physical (T-P) conditions. The dissolution of quartz has an important role on fluid-rock interaction processes in geothermal reservoirs at temperature-pressure conditions exceeding the critical point for water. Our experiments indicate, below about  $300^\circ\text{C}$ , that granite dissolution is mainly controlled by the degradation of feldspar, whilst the dissolution of quartz is the dominant phase controlling

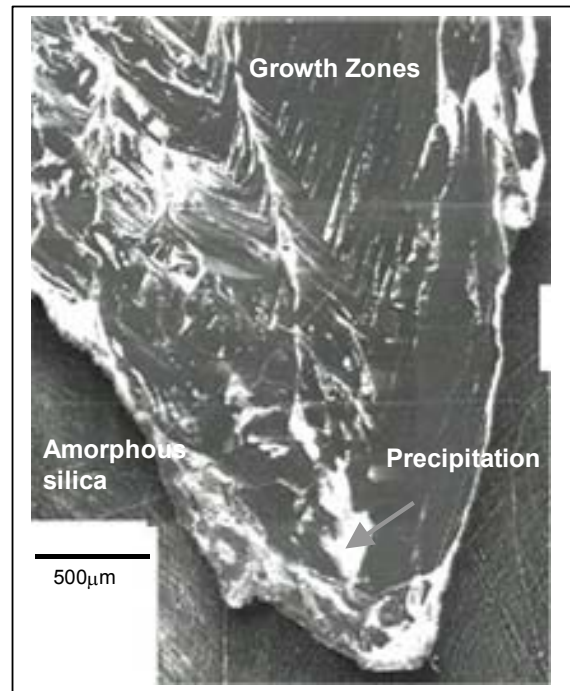


Figure 9. SEM-CL (mosaic) image of quartz from the Te Kopia geothermal field (cut along c-axis). Image reveals concentric growth zones, CL-bright quartz precipitation into open spaces, dissolution along crystal edges, and covering by CL-grey amorphous silica.

interaction processes at higher temperature. Our dissolution experiments also indicate that the solubility of granite at supercritical water conditions is significantly lower than in the subcritical region.

At high-pressure, supercritical conditions, hydrothermal fluids show greatest potential as a solvent, similar in behaviour to a ‘subcritical liquid’. In contrast, weaker water-rock interactions occur at high-temperature, supercritical conditions. The supercritical region has been regarded as a homogeneous state, but our granite/quartz dissolution experiments show that the supercritical region can be divided into two sub-phases (after Tsuchiya *et al.*, 2001), based on variations in the intensity of the water-rock interactions (Fig. 10). We demonstrate that there are two temperature-pressure regions, *within* the supercritical region - one liquid-like and the other vapour-like. The boundary, between the two subtly different ‘sub-phases’ is based on the T-P conditions that define the inflection points for the rate of quartz dissolution, and Si-concentration data from granite dissolution experiments.

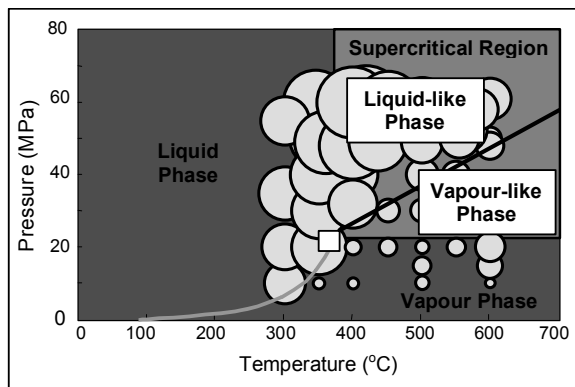


Figure 10. Liquid- and vapour-like phases, within the supercritical region. Circles indicate granite dissolution.

Temperature and (particularly) pressure are important, but dissolution rate in supercritical water is also greatly dependent on fluid density. In a developed DSGR system, the nature of induced fractures will change in shape and form, by the interaction of injected fluid, which mixes with in-situ (hydrothermal) fluid and hot reservoir rocks. The reduced solubility of granite (and by implication its silicate mineral phases) at supercritical water conditions, and corresponding weak water-rock chemical interaction, suggests that heat extraction from DSGR at supercritical conditions may overcome potential plugging of fluid flow paths due to precipitation, and hence provide more manageable operating conditions, compared to a system developed in a subcritical regime, where higher dissolution/precipitation rates are anticipated.

## 6.0 CONCLUSIONS

DSGR's have potential to provide renewable energy resources, from rock masses which are hotter and/or deeper than those reached by conventional geothermal development. Key factors for utilization of DSGR's are the creation, and maintenance of a large surface area for heat exchange, at conditions where weak water-rock interaction might preserve the heat exchange surface without plugging. Supercritical water is not a homogeneous phase, but can be subdivided into two 'sub-phases', one liquid-like and the other a vapour-like. The vapour-like region, at relatively low pressure, supercritical conditions, has the greatest potential to satisfy criteria necessary for efficient utilization of a supercritical DSGR.

SEM-CL has applications in mineral and geothermal exploration programmes. SEM-CL is an effective tool to reveal chronological and physico-chemical processes in (active / fossil) hydrothermal systems. Our SEM-CL study reveals microtextures in Te Kopia quartz that are unobservable by transmitted or reflected light microscopy, backscattered electron imaging or secondary electron imaging. SEM-CL may "fingerprint" quartz, it is useful tool in relating fluid inclusions to a corresponding alteration stage, and invaluable for understanding the temperature-pressure and chemical evolution of the hydrothermal system, and an important component of our study to understand quartz microfracturing and chemical processes affecting rock permeability in deep-seated geothermal reservoirs.

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