EXPERIMENTAL APPROACH TO UNDERSTANDING WATER-ROCK INTERACTIONS IN ENHANCED GEOTHERMAL SYSTEMS

G. BIGNALL, N. HIRANO, N. YAMASAKI, N. TSUCHIYA & K. NAKATSUKA

Department of Geoscience and Technology, Graduate School of Engineering
Tohoku University, Aoba 01, Aramaki, Aoba-ku, Sendai, 980-8579, JAPAN

SUMMARY - Batch- and flow-reactor autoclave systems were used to investigate granite and quartz dissolution processes, with pure water, under sub- and supercritical conditions (up to 600°C, 60MPa), to evaluate geothermal reservoir dynamics in a potential, deep-seated, enhanced geothermal system. Development of engineered reservoirs for Hot Dry Rock (HDR) geothermal energy extraction depend on the maintenance of (fracture) surfaces areas for heat exchange, yet precipitation of quartz (as a result of retrograde solubility at **>380°C),** albite and other minerals occur, which effectively ‘plug’ the system. The supercritical region, above the critical point for water **(~ 374°C,** 22 MPa), is not a homogeneous state, but comprises ‘liquid-like’ and ‘vapour-like’ regions, based on the dissolution behaviour of granite and quartz. Solvent properties in the liquid-like, ‘high-pressure’ supercritical region are similar to subcritical water. In contrast, dissolution reactions in the vapour-like, ‘high-temperatureflow pressure’ supercritical region are weak, and provide optimum conditions for energy utilization of an artificial HDR reservoir.

1. INTRODUCTION

Active geothermal systems, developed for their electricity generating capability, constitute a clean energy resource, but development is invariably constrained; either by resource temperatures, field pressures, a decline in mass output during production, or by environmental/cultural concerns.

In NE Japan, production drilling at the Kakkonda geothermal field intersected Kakkonda Granite, with a rock temperature at **–3.5 km** depth **of >500°C** (Doi, ***et al.*, 1998**). Utilization of deep-seated supercritical geothermal reservoirs **(>374°C** and 22 MPa for pure water), underlying conventionally developed geothermal systems, has the potential to supply huge amounts of renewable thermal energy, by extending the depth a geothermal reservoir, or by the engineered development of an enhanced HDR/HWR (Hot Dry Rock / Hot Wet Rock) geothermal system.

Several HDR projects have been initiated in Japan (e.g. Higashi-Hachimantai, Ogachi and Hijiori) and in other countries (Figure **1).** Water-rock interactions at supercritical conditions, however, and the physical character of fracture systems in Deep-seated Geothermal Reservoirs (DSGR) remain poorly understood, and prompted our multi-group, 5-year project (Hashida ***et al.*,** 2000).

In this paper, we detail the major findings of our experimental programmes, which aim to deduce granite/mineral dissolution and fracture characteristics at sub- to supercritical conditions, in a hypothetical, granite-hosted DSGR.

2. SCHEMATIC MODEL OF DSGR

Figure 2 shows a schematic representation of our magmatic-hydrothermal system, with a convective region overlying a deeper conductive zone, which may potentially host a DSGR. In a conventional geothermal system, the main upflow zone or reservoir (e.g. at A) may be tapped by **<2.5-3km** wells sited to intersect faults/permeable features (that supply fluids to surface features, B).

Multiple magmatic intrusions **(C)** are the ‘heat source’ for many active, ‘high-temperature’ geothermal systems, (e.g. in the Philippines; Reyes, 1990), with the brittle-plastic transition in the intrusive rocks occurring at **–370-400°C (D).**

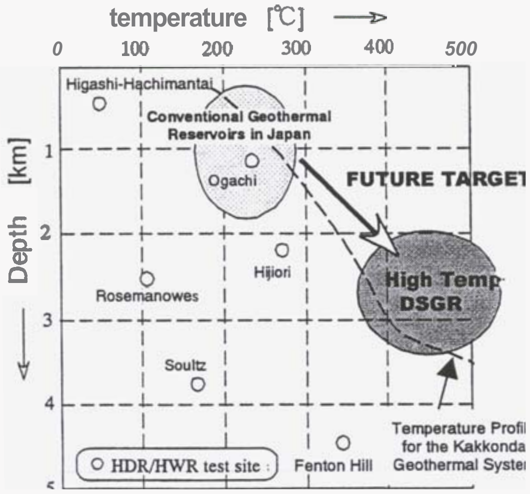

Figure 1: Temperature-depth conditions for HDR test sites in Japan and elsewhere, and potential (future?) DSGR resources.

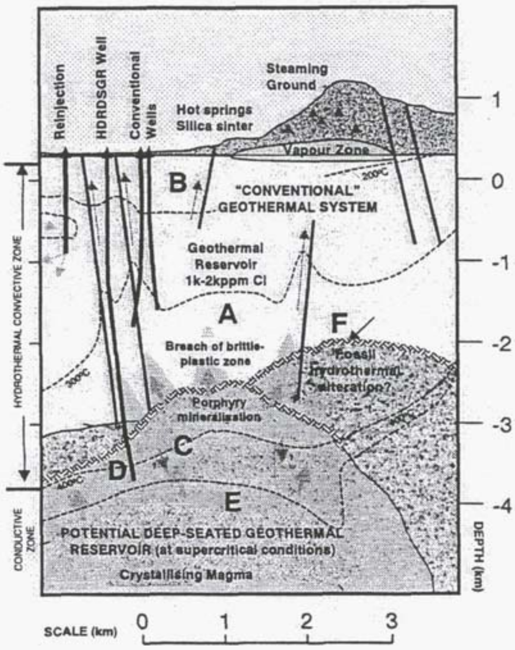

Figure 2. Model of a magmatic-hydrothermal system, with a convective region at intermediate depths and a supercritical geothermal reservoir at >3 km (see text for explanation of symbols).

Some wells have penetrated the brittle-plastic transition, but typically have little permeability.

The core (E) of the intrusion is partly crystallized, and exsolves fluids that accumulate near the top of the pluton, where high pressures may produce fracturing, brecciation and expulsion of fluid across the brittle-ductile transition into overlying rocks (Fournier, 1999). Countering processes that increase permeability is deposition of quartz and other minerals (in veins and breccia matrix; e.g. at F). The circulation of fluid at hydrostatic pressure, and resultant self-sealing, has major implications for the energy utilization of supercritical DSGR's.

3. EXPERIMENTAL PROGRAMME

Many workers have reported on their experimental and theoretical approach to understanding the dissolution of silicate minerals at elevated temperature-pressure conditions (e.g. Savage *et* al., 1992; Chigira and Watanabe, 1994).

Quartz dissolution has an important role in fluid-granite interaction processes at supercritical temperature-pressure conditions, and therefore the focus of our water-rock interaction experiments.

Savage ***et al*** (1987) demonstrated, using batch-type autoclave systems, that the dissolution of granite at <300°C was controlled mainly by the decomposition of feldspar. In contrast, Takahashi *et* al. (1988) and Wang et al. (1997; *submitted)* argue that quartz dissolution is the dominant process controlling granite dissolution at temperature conditions exceeding 300°C.

3.1 Experimental Methodology

Hydrothermal dissolution/fracturing experiments, were conducted on synthesized (impurity-free) quartz (30 x 15 x 3 mm) and medium-grained Iidate Granite (1 cm^3 cube), with distilled water. Our batch-autoclave apparatus incorporates a specially designed hot-water injection unit (Figure 3). It is necessary to collect reaction solutions for compositional analysis, to determine sample reaction rates, and the unit allows equal volumes of fresh solution to be injected into the autoclave, and maintain desired pressure conditions.

The effective volume of our batch autoclave system is $300 cm^3$, with the main body of the autoclave being made of hastelloy-C, with other parts manufactured by SUS-316. The maximum temperature and pressure in our experiments was 600°C and 60 MPa respectively, with an average heating rate of 10°C/min.

The rate of dissolution, determined during long-term experiments (ranging from 24 to 200 hours), was calculated per unit surface sample area, which was assumed to be constant. Reacted solutions were collected, and analysed by ICP (HITACHI P4000), with sample weight change, petrology and morphological characteristics also examined.

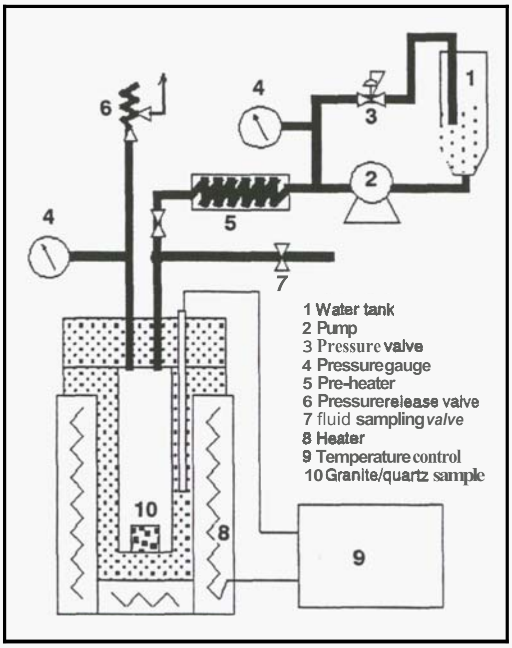

Figure 3. Schematic illustration of our batch-type autoclave system.

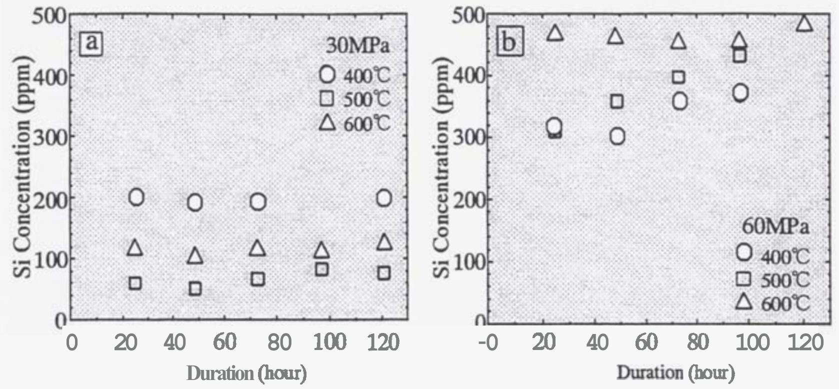

Figure 4. Relationship between Si concentration and reaction duration, for quartz dissolution at:
(a) 30 MPa, and
(b) 60 MPa
at 400, 500 and 600°C reaction temperatures.

3.2 Results

Quartz Solubility Experiments

Silica solubility is a complex function of temperature and fluid density. According to Wang et *al.* (1997), quartz solubility in pure water, under saturated vapour pressure, is dependent on the ionic product of water (Kw), which is itself a function of temperature and pressure, with the highest ionic product for a given pressure (under 100MPa) occurring in the range of 300°C – 400°C.

Si concentration of reacted fluids, after quartz dissolution in pure water, at 30 MPa and 60 MPa, for 400, 500 and 600°C is shown in Figure 4a. The high Si concentration of fluid from the 30MPa/400°C experiment, compared to 500°C and 600°C, is an effect of a greater ionic product.

Whilst the solubility/dissolution behaviour of quartz is clearly controlled by the ionic product, fluid pressure has a great influence at supercritical conditions. The Si concentration of reacted fluids from the 60MPa/600°C experiments quickly reach an equilibrium state (Figure 4b), whilst Si concentrations for reaction experiments undertaken at 400°C and 500°C have yet to reach an equilibrium condition, even after 100 hours.

Dissolution Rate

The relationship between quartz dissolution rate (0 to 24 hours) and specific volume is shown in Figure 5. With increasing specific volume, dissolution rate decreases exponentially; and therefore the specific volume is a suitable thermodynamic variable by which to demonstrate the solvent capability of a hydrothermal fluid.

The rate expression of quartz dissolution, under sub- and supercritical conditions can be described by following n-th order kinetic equation:

$$r = kV^n$$

where r is dissolution rate, k is apparent kinetic constant, V is specific volume of fluid, and n is apparent reaction order.

In Figure 6, the slope of lines approximating change of dissolution rate at low specific volume is minus unity, which means dissolution rate is proportional to water density. At higher specific volume (at supercritical conditions) the slopes are much steeper, and indicate that the fluid loses its capability as a solvent (i.e. it is less effective in dissolving the (granite) rock).

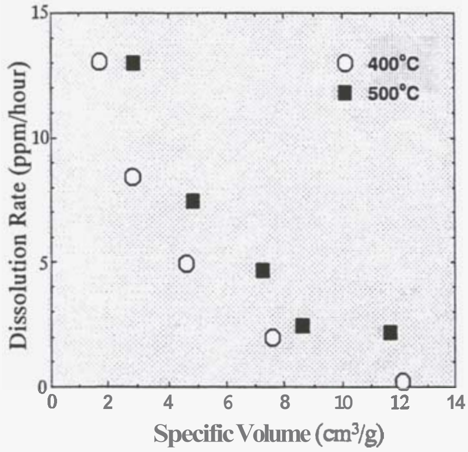

Figure 5. Relationship between specific volume of supercritical water and dissolution rate of quartz, at various (reaction) temperatures.

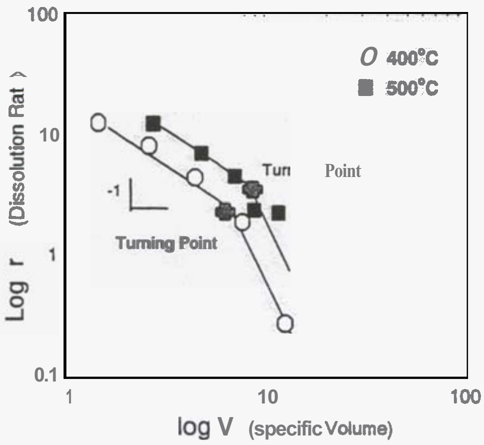

Figure 6. Logarithmic diagram showing the relationship between specific volume and dissolution rate of quartz.

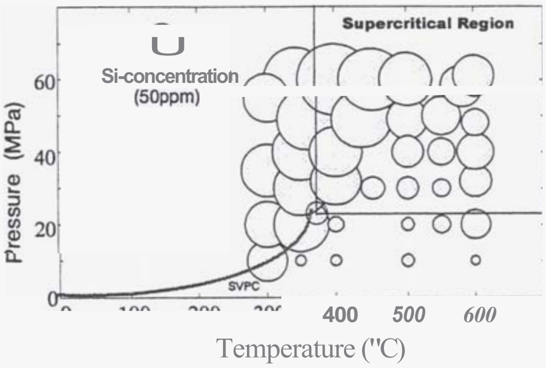

Figure 7 Dissolution of granite, indicated by Si-concentration. SVPC is the Saturated Vapour Pressure Curve of pure water.

The dissolution of granite is represented by the Si-concentration of reacted fluid (Figure 7). High Si-concentrations were determined at conditions less than the critical temperature for pure water, but have low Si-concentrations at high temperature/ low pressure conditions. Water-rock interactions (i.e. granite dissolution) occur in both the sub- and supercritical regions, from 300°C to 450°C, but chemical reactions are weaker at high temperature.

Quartz/Granite Fracturing

A series **of** batch-autoclave experiments, in pure water-quartz/granite systems, were conducted at a range of sub- to supercritical conditions (from 250°C to 600°C; 0.1MPa to 60MPa; 24 to 120 hours), to deduce the range of temperatures and pressure at which (artificial) quartz and granite fracturing might occur in a **DSGR** system.

There is no obvious fracturing of ***quartz*** under dry conditions (0.1 MPa, even up 600°C), but quartz is fractured at **3MPa/600°C,** in the presence of water. The boundary of quartz fracturing is shown in Figure **8,** and coincident with the subcritical, vapour-phase for pure water.

At elevated temperatures (>550°C) the distinction between unfractured and fractured quartz approximates the phase boundary conditions that differentiate ***a-*** and β-quartz, although hydrothermal fracturing does occur in the stability field of ***a-quartz.***

The temperature-pressure conditions conducive for granite fracturing are shown in Figure 9, although apparent discrepancies with results from triaxial high temperature/pressure experiments require further consideration (Hashida *et* al., 2001). Petrographic examination reveals that the fractures predominantly occur in quartz, and not in feldspar or other primary mineral phases.

Flow reactor Experiments

Results of 'first-stage' flow autoclave experiments, using a simple, lm-flow reactor operating at sub- to supercritical conditions (up to 450°C, 35MPa), were reported by Wang ***et al.*** (1997), Bignall ***et al.*** (2000; ***accepted).*** Recently, we constructed a 'second-generation' flow reactor that allows fluid-rock interactions at conditions up to 600°C and 60MPa, and 0-15cm^3/m flow rate, in three 1-m Hastelloy-C autoclave units (Figure 9).

In our flow-reactor experiments, distilled water is heated and injected into the pressurized autoclave system, with the temperature in each reactor tube **(A, B,** C) controlled by an independently operated heating system. In our **350°C** (inlet)/30MPa

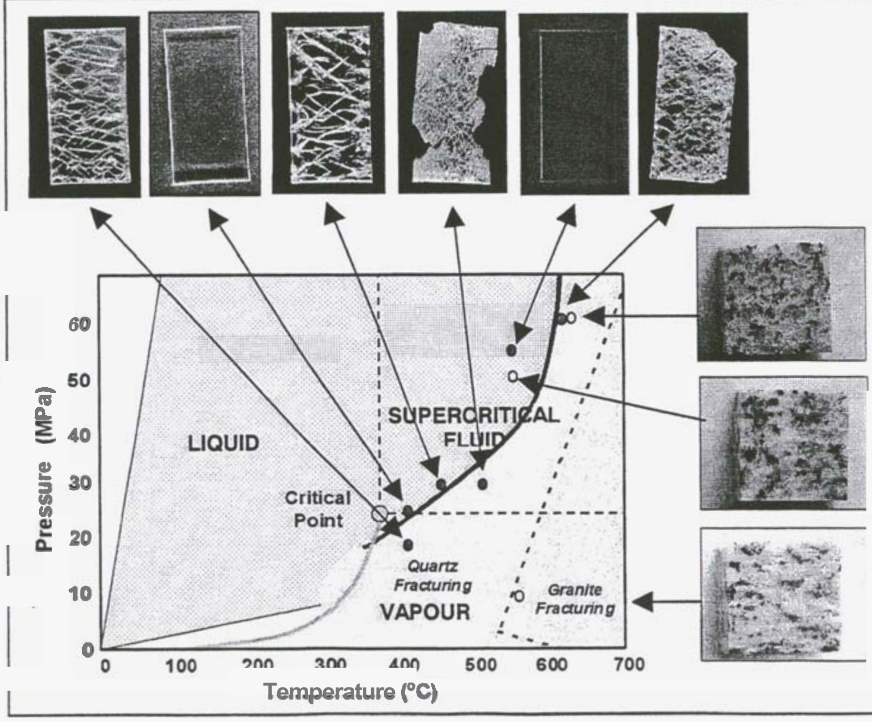

Figure 8. Phase relationships for pure water, showing the temperature-pressure regions identified in our batch-auoclave experiments that are conducive for quartz- and granite-fracturing

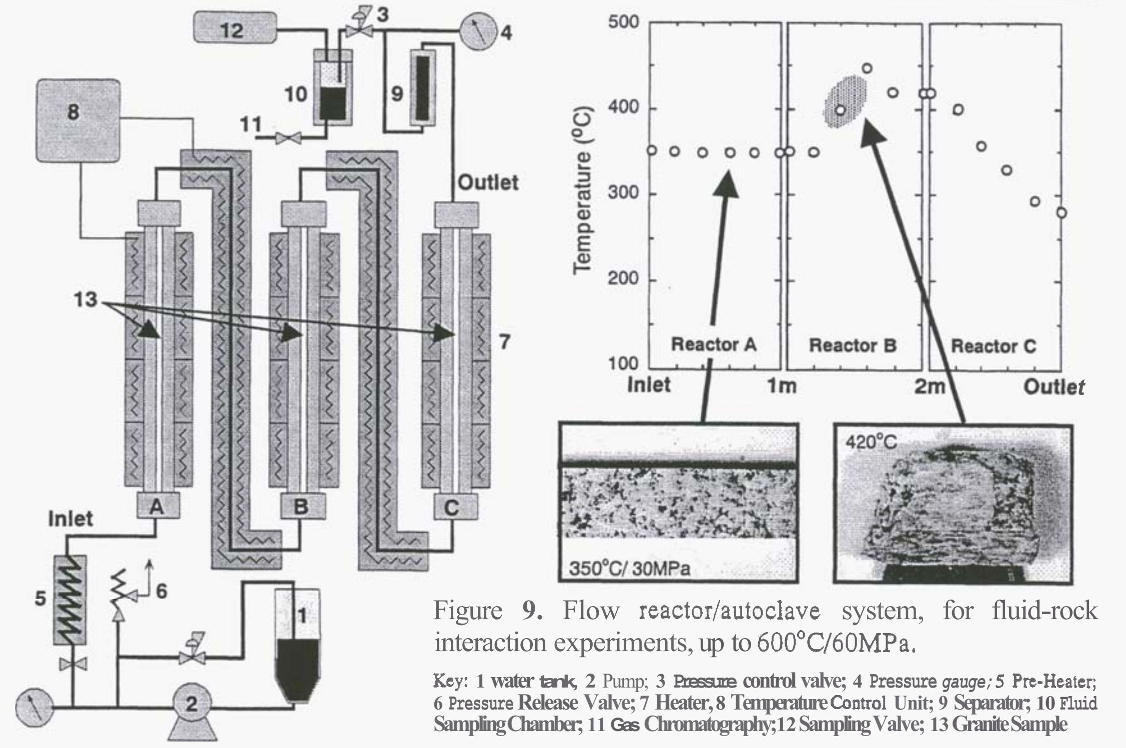

Figure 9. Flow reactor/autoclave system, for fluid-rock interaction experiments, up to 600°C/60MPa.

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 Sample

experiment (Figure 9), 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 region there is a strong weight gain, with precipitation of quartz, lesser albite and sericite.

The occurrence of quartz 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. DISCUSSION

Fluid-rock interactions are intrinsically linked to water-rock ratios in the hydrothermal system, the surface area of fracture/permeable channels and to the nature of those surfaces, as well as the 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.

At high-pressure, supercritical conditions, the fluid shows 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 (Tsuchiya et *al.*, 2001), based on variations in the intensity of the water-rock interactions (i.e. granite dissolution), and consistent with the results of Nishikawa and Morita (2000) who described density fluctuations in the supercritical region of pure water.

In Figure 10, we show the 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 (Figure 6), and Si-concentration data from granite dissolution experiments (Figure 7).

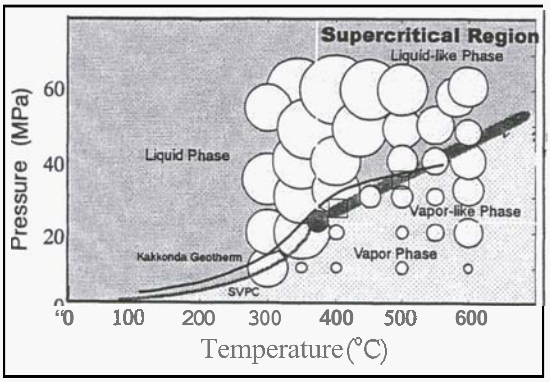

Figure 10. Liquid- and vapour-like phases, within the supercritical region. Open squares are 'turning points', indicative of change in dissolution rate (from Figure 6). Circles show Si-concentration, indicative of granite dissolution (from Figure 7).

5. CONCLUSIONS

DSGR's have the potential to provide effectively renewable energy resources, from rock masses hotter and/or deeper than those reached by present, conventional development.

Key factors for the utilization of DSGR's are the creation, and maintenance of a large surface area for heat exchange (such as a high density fracture network and/or a porous media), 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.

6. ACNOWLEDGMENTS

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