Hydrothermal alteration aspects of high heat producing granites in Australia & Europe

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In order to fully understand the relatively unstudied high heat producing granites (HHPG) of the Australian continent, this project has studied the European analogues of Cornwall, (United Kingdom), Soultz-sous-Forêts (France) and the Erzgebirge (Germany). HHPGs are characterised by their anomalous radiogenic element (U, Th and K) contents (Table 1), which is higher than average continental crust values. The Big Lake Suite in the Cooper Basin is the most investigated Australian enhanced geothermal system (EGS); Gatehouse et al., 1995).

Alteration mineralogy is a widely used feature to explore for potential ore deposits; however, this method has not yet been deployed for identifying and characterising EGS. In the current project, we are investigating alteration mineralogy of HHPG, with particular emphasis on trace element and stable isotope geochemistry. If successful, this can offer an additional approach to geothermal exploration and resource characterisation.

Table 1: Typical compositions for high heat producing granites for EGS. Sourced from T Uysal, J Van Zyl, S Bryan, unpubl. data; Charoy, 1986; Stussi et al., 2002; Forster et al., 1999; Chappell & Hine, 2006

EGS Target	Composition	Age (Ma)	SiO_2 (wt%)	K_2O (wt%)	U (ppm)	Th (ppm)
Cooper Basin granites	Biotite granites	~Mid-Carb.	-	-	10.0-30	28-144
Carnmenellis granites (Cornwall, UK)	K-feldspar megacrystic biotite granites	293-274	69-76	4.6-5.9	13-20	15-20
					(4-38)	(5-45)
Soultz-sous-Forêts (Rhine Graben, France)	K-feldspar porphyritic monzogranite	~330	67-69	~4.5	6.2-14.1	24-37
Erzgebirge (Germany)	Biotite, 2 mica & Li-mica granites	325-318	70-77	4.5-5.3	8.0-30	15-37

Keywords: Geothermal, alteration mineralogy, Cooper Basin, stable isotopes, REE, Soultz-sous-Forêts

Background

During emplacement of granitic bodies, advective heat flow can occur due to hydrothermal circulation of surrounding waters. Several studies have noted that igneous rocks enriched in radioactive elements such as U, Th and K can have a similar effect and promote hydrothermal circulation long after the intrusion has cooled (Allman-Ward, 1985, Pirajno, 2005). On circulation of these hydrothermal fluids, variable alteration products will result from original mineral phase disequilibrium with the fluid. The alteration assemblage produced depends on factors such as wall-rock and fluid composition, temperature, salinity and water/rock ratio. The variation in alteration assemblage may control the concentration of incompatible elements, including the radiogenic elements as certain alteration reactions are associated with either enrichment or depletion of these elements. A pertinent example of this was noted to have occurred in some Carnmenellis (Cornwall, UK) monazites (Poitrasson et al., 1996), whereby Th was enriched through brabantitic (Förster, 1998) cation exchange during chloritisation of biotites:

$$2REE^{3+} \rightleftharpoons Th^{4+} + Ca^{2+}$$

Incompatible elements (HFSE, REE, LILE, and B etc., including the radiogenic elements) have a low partition coefficient (K_D) (White, 2007), and hence will preferentially stay in fluid phases during crystallisation of melt and hydrothermal fluid movement. .

Hydrothermal fluids affecting surrounding wall-rocks can be classified (based on their origin), into meteoric, magmatic, marine or connate. One aspect of this project is to deduce the origin and evolution of the hydrothermal fluids involved in the alteration of HHPG and surrounding sedimentary rocks.

Analytical Work

The project results will be attained via multifaceted petrographic, geochemical and geochronological analyses. The geochronology will use Rb-Sr isochron dating of alteration-related illitic clay minerals, whereas the geochemical techniques will include both stable isotope

analysis (δ18O and δD) for hydrothermal fluid evolution and ICP-MS analysis for trace element quantification.
Qualitative and quantitative petrographic analyses will be achieved with transmitted and reflective microscopy, as well as EDS (via JEOL XL 30 SEM) and EPMA. The EPMA will be used to determine quantitatively which mineral phases encompass the elements of interest; such as incompatibles and radiogenics.

Queensland EGS targets and Cooper Basin

Queensland

Following estimated subcrustal temperature mapping and the increasing need for renewable energy sources, Queensland has been targeted by the state government as a prospective area for geothermal energy. The principal areas of interest for EGS are the Galilee Basin, Innot Hot Springs region, Hodgkinson Province, Styx Basin, Maryborough Basin and North d'Aguillar Block, Wandilla Province. These targeted areas have been based on the presence or likely presence of HHPG at depth ranging in age from the Late Devonian to Cretaceous. Many of the granite bodies emplaced within Queensland are linked to an extensional tectonic origin (Glen, 2005). Analysis of the intrusions, overlying sediments and their associated alteration zones will be performed as part of this project.

Cooper Basin

The Carboniferous Big Lake Suite granite in the Cooper Basin' represents one of the world's hottest EGS (Gatehouse et al., 1996). According to our recently acquired ICP-MS trace element data, U and Th contents in these granites reach levels of up to 144ppm, reflecting enrichment levels up to 13 times that of the upper continental crust (UCC(McLennan, 2001)). Cores taken from the granite and overlying sediments show varying degrees of alteration, with a range of incompatible element enrichment, such as U and Th. The highly altered zones have a predominant greisen-style sericite (illite) and re-precipitated quartz assemblage. We believe that this alteration may well have caused the localised enrichment in radiogenic element-bearing minerals such as illite, K-feldspar, and some accessory minerals (e.g., thorite).
The fluid history of the Cooper Basin can be deduced with the use of both crystallinity and stable isotope analyses of the illite. Illite crystallinity is a useful indicator of the temperature gradient in active geothermal systems and for locating fossil hydrothermal systems associated with ore deposition (Ji and Browne, 2000).

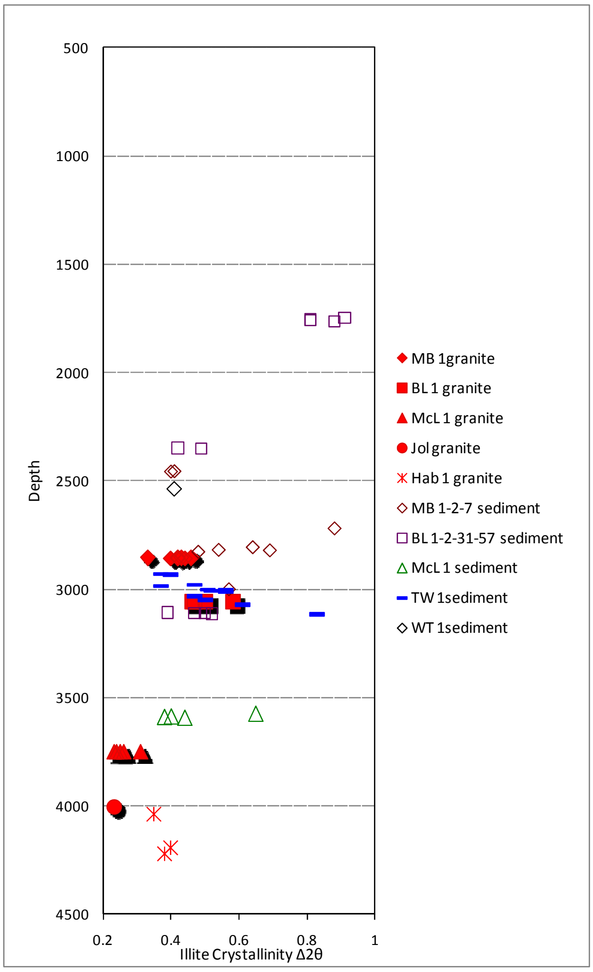

Fig 1: Graph of illite crystallinity against depth from Cooper Basin cores

Illite crystallinity is controlled by crystallisation temperature, water/rock ratio, and time available for crystallization (Arkai, 2002; Ji and Browne, 2000; Merrriman and Frey, 1999). Better-developed crystalline illites show narrower 001 basal illite peaks and have lower IC values. Such illites were formed at higher temperatures or during prolonged heating events. Higher IC values (wider peaks), on the other hand, indicate lower crystallisation temperatures and/or rapid precipitation during hydrothermal processes. Illite crystallinities are seen to progressively increase with increasing core depth, suggesting a higher crystallisation temperature and hence hydrothermal fluid temperature at depth. The granite intersected in Jolokia and McLeod 1 seems to have experienced highest reservoir temperatures.
According to our present isotopic studies of alteration-related illite within the granite and sedimentary cover, oxygen and hydrogen isotope compositions range from δ18O = -1.8‰ to +2.7‰;δD = -99‰ to -121‰ and δ18O = +2.3‰ to +9.7‰, δD = -78‰ to -119‰, respectively. Such values are much lower than those reported for many deeply buried sedimentary basins (Clauer and Chaudhuri, 1995). The calculated oxygen and hydrogen isotope compositions of

fluids in equilibrium with the illites are depleted in ^{18}O and deuterium, comparable to those of waters reported for most high-latitude sedimentary basins. Hence, stable isotope data of alteration minerals in the granite and the overlying sedimentary rocks suggest the operation of a hydrothermal system involving high latitude meteoric waters during Permo-Carboniferous extensional tectonism in the Cooper Basin region.

Europe

The European HHPG examples have all been considered to be approximately syn-genetic with extension following the late Palaeozoic Hercynian/Variscan orogeny between Gondwana and Laurasia.

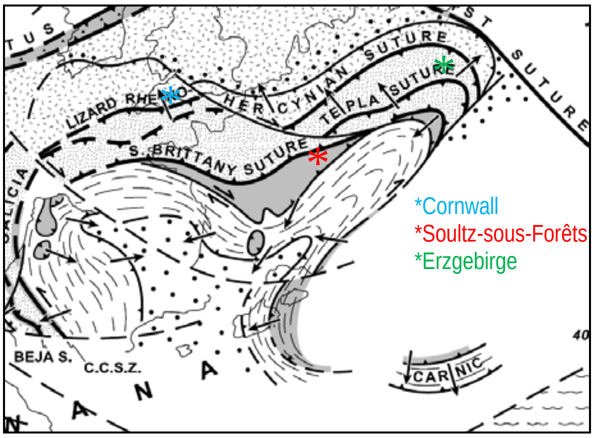

Fig 2: Possible tectonic suture configuration of Hercynian orogeny from Matte (2001)

Erzgebirge, Germany

Located at the north-western edge of the Bohemian Massif (Förster and Förster, 2000), the Erzgebirge consist of highly-evolved voluminous granites intruded into highly crystalline metamorphic rocks (Tischendorf et al., 1965). These granites are highly enriched in U and Th (Table 1) with principal radiogenic mineral phases of uraninite, U-bearing micas, xenotime, monazite-(Ce)-brabantite solid solution series minerals, and thorite (Förster, 1998).
As of yet, our analyses (Rb – Sr dating and stable isotope) have not been undertaken on Erzgebirge samples, but samples will be analysed in collaboration with Dr. Hans-Jurgen Förster (Potsdam, GeoForschungsZentrum).

Soultz-sous-Forêts, France

Located south-west of the Erzgebirge outcrops, Soultz-sous-Forêts was emplaced debatably either as an I-type subduction-related or late-stage extensional granite body (Flötmann and Oncken, 1992). Hydrothermal fluid circulation and alteration was promoted by E-W lithospheric extension (Schleicher, 1998). Petrographic studies of alteration mineralogy by Bartier et al. (2008) identified intense propylitically altered granite with newly equilibrated mineral assemblages of tosudite (chlorite-smectite), illite, chlorite and carbonates, whilst K-feldspar appears to be largely preserved.
Following further transmitted and scanning-electron microscopy performed at The University of Queensland, we identified several key alteration-related mineral phases. Altered sphene, for example, found in sample K177 (2000m – EPS1), is acting as a principal mineral phase that contains various rare earth elements (REE). Estimations of composition from semi-quantitative EDS have detected Ce, Y, Nd, La, Th and Ca with possible PO_4 or SiO_4 anion complexes.

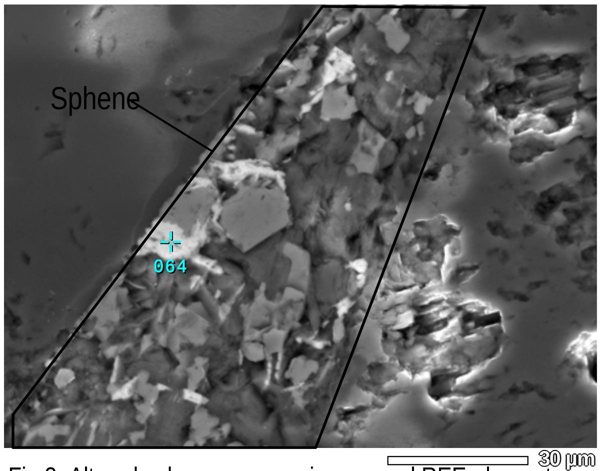

Fig 3: Altered sphene compassing several REE-element-bearing mineral phases

This may indicate a potentially high fluid/rock ratio allowing for incompatible element transport during alteration of the granitic protolith. Monazite has also been found proximal to subhedral hematite grains. A possible mechanism for this mineral association can be traced back to the formation of a redox interface with co-precipitation of U^{4+} and Fe^{3+}.

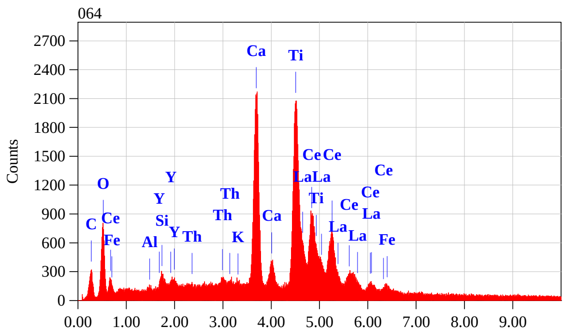

Fig 6: EDS for altered sphene found in sample K177 exhibiting intense propylitic alteration.

Cornwall, United Kingdom

Cornwall marks the far western district of the Euro-Hercynian granites with 5 major outcrops; Dartmoor, Bodmin Moor, St. Austell, Carnmenellis and Lands End. Multiple studies by Ball et al., (1979) and Charoy (1986) have found the Cornwall granites to be similarly enriched in radiogenic elements (Fig. 1) as the middle-European examples. Alteration systematics are of great importance within this system as they have been directly linked to the leaching and mineralisation of uranium. Allman-Ward (1985) noted 4 predominant alteration styles affecting the St. Austell granite; greisenisation, tourmalinisation, kaolinisation and haematisation. Uranium is likely to be transported in solution during hydrothermal alteration, as oxidising fluids will cause a loss of electrons (U^{4+} to U^{6+}) and hence increase mobility. The most likely location of the displaced uranium will be in a distal alteration zone possibly accompanied by haematite mineralisation. An example of this can be seen in Wheal Remfry where quartz-haematite veins show uranium values of up to 50ppm (Allman-Ward, 1985).

Summary

Highly altered granites in the Cooper Basin are substantially enriched in lithophile elements, particularly in Cs, Rb, Be, Th, U relative to the upper continental crust. U and Th contents are 10 and 13 times higher than those of the UCC. Some enrichment of heat-producing elements was promoted by a regional hydrothermal event leading to the precipitation of U and Th- bearing minerals such as illite, K-feldspar and accessory minerals (other than zircon). Oxygen and hydrogen isotope compositions of fluids are depleted in ^{18}O and deuterium, comparable to those of waters reported for most high-latitude sedimentary basins. European examples of enhanced geothermal systems show similar redistribution and enrichment patterns of rare earth and radiogenic elements as a result of hydrothermal alteration. This can be seen in monazites proximal to chloritised biotites in the Carnmenellis granite of Cornwall. Unlike the Cooper Basin, stable isotope data has not yet been acquired from European samples. However once performed, hydrothermal fluid origin and evolution can be deduced for these EGS. Alteration mineralogy and geochemistry of relatively shallow sedimentary sections is a potentially valuable tool to evaluate the presence of a concealed geothermal heat source in the basement of sedimentary basins.

References

Allman-Ward, P., 1985. Distribution of uranium and thorium in the western lobe of the St. Austell granite and the effects of alteration processes, High heat production granites, hydrothermal circulation and ore genesis conference, The Institute of Mining and Metallurgy, 437-458.

Aquilina, L., Pauwels, H., Genter, A., and Fouillac, C., 1997. Water-Rock interaction processes in the Triassic sandstone and granitic basement of the Rhine Graben : Geochemical investigation of a geothermal resevoir. Geochimica et Cosmochimica Acta, 6, 4281-4295.

Arkai, P., 2002. Phyllosilicates in very low-grade metamorphism: Transformation to micas. Micas: Crystal Chemistry and Metamorphic Petrology, 46: 463-478.

Bartier, D. et al., 2008. Hydrothermal alteration of the Soultz-sous-Forêts granite (Hot Fractured Rock geothermal exchanger) into tosudite and illite assemblage. European Journal of Mineralogy, 20, 131-142.

Bryan, S. E., Holcombe, R. J. and Fielding, C. R, 2001. Yarrol Terrane of the northen New England Fold Belt: forearc or backarc? Australian Journal of Earth Sciences, 48 296-316

Clauer, N. and Chaudhuri, S., 1995. *Clays in Crustal Environments. Isotope Dating and Tracing*. Springer Verlag, Heidelberg, 359.

Flötmann, T. and Oncken, O., 1992. Constraints on the evolution of the Mid Crystalline Rise –a study of outcrops west of the river Rhine. Geologische Rundschau, 81, 2, 515-543.

Förster, H-J., 1998. The chemical composition of REE-Y-Th-U-rich accesspry minerals in peralumnous granites of the Erzgebirge-Fichtelgebirge region, Germany, Part 1: The minazite-(Ce)-brabantite solid solution series. American Mineralogist, 83, 259-272.

Förster, H-J., 1998. The chemical composition of REE-Y-Th-U-rich accesspry minerals in peralumnous granites of the Erzgebirge-Fichtelgebirge region, Germany, Part 2: Xenotime. American Mineralogist, 83, 1302-1315.

Glen, R. A., 2005. The Tasminides of Eastern Australia, from: Vaughan, A. P. M., Leat, P. T. and Pankhurst, R. J., 2005. *Terrane Processes at the Margins of Gondwana*. Geological Society, London, Special Publications, 246, 23-96.

Ji, J. and Browne, P.R.L., 2000. Relationship between illite crystallinity and temperature in active geothermal systems of New Zealand. Clays and Clay Minerals, 48: 139–144.

Ledésert, B. et al., 2009. Fractures, hydrothermal alterations and permeability in the Soultz Enhance Geothermal System. C. R. Geoscience

Matte, P., 2001. The Variscan collage and orogeny (480±290 Ma) and the tectonic definition of the Armorica microplate: a review, Terra Nova 13, 122-128.

McLennan, S. M., 2001. Relationships between the trace element composition of sedimentary rocks and upper continental crust. Geochem. Geophys. Geosyst, 2.

Merrriman, R.J. and Frey, M., 1999. Patterns of very low-grade metamorphism in metapelitic rocks. In: M. Frey and D. Robinson (Editors), Low Grade Metamorphism. Blackwell Science, Cambridge, pp. 61-107.

Murray, C. G., 2003. Granites of the New England Orogen., from: Blevin, P. L., Jones, M. and Chappell, B., *Magmas to Mineralisation: The Ishihara Symposium.* Geoscience Australia Record, 2003/14, 101-108.

Pirajno, F., 2005. *Hydrothermal Processes and Mineral Systems.* Springer Link, Geological Survey of Australia, 73-157, 990-995.

Schleicher, A. M., Warr, L. N., Kober, B., Laverret, E. and Clauer, N., 2006. Episodic mineralisation of hydrothermal illite in the Soultz-sous-Forêts granite (Upper Rhine Graben, France). Contrib Mineral Petrol, 152, 349-364.

Stussi, J-M. et al., 2002. The hidden monzogranite of Soultz-sous-Forêts (Rhine Graben, France): Mineralogy, petrology and genesis. Géologie de la France, 1, 45-64.

Taylor, H. P., 1997. Oxygen and Hydrogen Isotope Relationships in Hydorthermal Mineral Deposits, from: Barnes, H. L., 1997. *Geochemistry of Hydrothermal Ore Deposits*. John Wiley and Sons, Inc, 229-288.

White, W. M., 2007. *Geochemistry*: Trace Elements in Igneous Process. John Hopkins University Press, 258-312.