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Papers are listed under subject headings based on symposia organised for the Goldschmidt Conference in Edinburgh in August-September, 1994. Sub-headings are used for the larger symposia, and often these sub-headings cover material which overlaps several symposia. The subject headings and their research areas are briefly outlined below, and these are followed by listings of the papers in alphabetical order under each subject heading or sub-heading.

1. **Theoretical geochemistry** - applications of theoretical mineral physics to mineral chemistry and element distribution, including aspects of diffusion; ab initio methods.
2. **Experimental geochemistry** - laboratory experimental studies across the full spectrum of P-T conditions.
3. **Geochemical techniques** - developments in instrumentation and techniques; special applications and programmes.
4. **Weathering and erosion, deposition and diagenesis** - geochemical processes in low temperature rock degradation and genesis.
5. **Groundwater chemistry and palaeohydrology** - geochemical characterisation of groundwater systems and of time dependent hydrogeological changes, significance to palaeoenvironments, prognoses of future changes.
6. **Waste containment and pollutant transport** - geochemical assessment of the controls and risks of all types of waste and pollutant disposal.
7. **Ocean composition and fluxes during the Quaternary** - high resolution documentation of changes in marine geochemistry, and geochemical indicators of recent events.
8. **Ocean palaeochemistry and the evolution of the ocean basins** - the geochemistry of ocean evolution in the pre-Quaternary.
9. **Modern submarine hydrothermal processes** - controls on the geochemical reactions within sub-seafloor hydrothermal systems, and their spatial and temporal variability.
10. **Mechanisms of isotopic and chemical communication in crust and mantle rocks** - application of new microanalytical (ion and laser probe) techniques to constrain micron to millimetre scale processes in rocks, and their relevance to macroscopic studies of mass and heat transport.
11. **Partitioning of elements** - new data on partition coefficients for mineral, melt, fluid phases as determined by experiment or measurement of natural phases; including aspects of the distribution of precious metals.
12. **Dehydration, partial melting and fluid distribution in the crust** - covering both the geochemistry of high-temperature processes in the continental crust and the modification of crust undergoing subduction.
13. **Magma generation processes** - geochemical evidence of the generation and evolution of mantle-derived melts.
14. **Mantle development in space and time** - geochemical and mineralogical information on mantle constitution.
15. **Geochronometry and thermal history** - quantitative thermal histories of rocks and metamorphic belts.

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- Sackett, W
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The Himalia Ridge Formation, Alexander Island, Antarctica
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4e. *Experimental studies of mineral dissolution, interfaces and hydrothermal solutions*

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10. Mechanisms of isotopic and chemical communication in crust and mantle rocks

10a. *Small-scale evidence of geochemical processes in mantle rocks and ultrabasic complexes*

See section 14g

10b. *Spatial distribution of isotope and trace element variations in rocks and magmas, and their implications*

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14a. Experimental constraints on melting in the mantle

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14b. Source regions, extraction and evolution of basaltic melts

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14c. Mantle mineralogy and the transition zone.

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Effects of water content, temperature and pressure on actinide tracer diffusion in melts of haplogranitic composition

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Introduction

An understanding of the rates and mechanisms of U and Th diffusion in silicate melts and glasses is necessary to fully understand such diverse phenomena as U-Th decay series disequilibria, melt-crystal partitioning, and leaching of actinides from nuclear waste glasses. Uranium diffusivities have been determined in anhydrous borosilicate nuclear waste glasses (Dunn, 1987), and numerous investigators have studied release rates of actinides during leaching of glass by hydrothermal solutions (e.g. Schreiber *et al.*, 1985). Dissolved water is known to enhance diffusion of other high field strength elements in silicate melts (e.g. Harrison and Watson, 1983). In light of the abundance of water in felsic silicate melts, and the prevalence of water-rich compositions in ancient weathered natural glasses (Mungall and Martin 1994), we have investigated the effects of water content and temperature on U and Th diffusion rates in a synthetic haplogranite composition at pressures ranging from one atmosphere to 10 kbars.

Experiments

Two batches of bubble-free, homogeneous glass (one clean HPG-8 (see Holz *et al.*, 1992 for composition), the other doped with 0.1% each of U and Th) were prepared by fusing oxide and carbonate powders in a Pt crucible at 1600°C, stirring for several days with a Pt spindle, and cooling in air. For one-atmosphere anhydrous experiments, the glass was drilled and sliced into 8 mm-diameter discs 2 mm thick with polished ends. Diffusion couples were made by juxtaposing the two glasses in a Pt capsule. For high pressure experiments the clean glass was ground and tamped into Pt cylinders; in water-bearing runs double distilled water was added to give 4.5 wt. % water, after which doped glass powder was poured in, tamped, and the cylinder was welded shut. One atmosphere experiments were conducted in a box furnace; high pressure experiments were conducted in a 1/2" piston-cylinder apparatus with alumina pressure medium. One atmosphere

experiments were quenched by removing from the furnace, whereas high pressure experiments were quenched by turning off the furnace. Run products were sectioned and polished for analysis on a Cameca WDS microprobe at 25 kV accelerating voltage and 100 nA probe current, with a defocused beam. Most traverses were 1 to 3 mm wide; if the flat extremes of the diffusion profile were not at least 500 µm wide the experiment was discarded. Because there is no variation in the matrix through the couple, data were collected and processed as raw X-ray counts at 1 to 7 (usually 5) micron point spacings.

Results

Profiles were fit to an error function to minimize χ^2 ; all the products used had infinite couple geometry and to show no compositional dependence, i.e., the estimated diffusion coefficients are assumed to approximate tracer diffusion coefficients. Uncertainty was estimated by finding the range of values of D within which χ^2 gave 95% confidence of an insignificant residual. The variation of D_U and D_{Th} with temperature, water content and pressure is shown in Fig. 1 for all experiments. The logarithms of both U and Th diffusivities are linear against reciprocal temperature, and are given by the following Arrhenius relationships:

$$\begin{aligned} D_U &= 7.94 \times 10^{-3} \exp\{260 \text{ kJ mol}^{-1}/RT\} \text{ dry, 1 bar} \\ D_{Th} &= 1.92 \times 10^{-1} \exp\{318 \text{ kJ mol}^{-1}/RT\} \text{ dry, 1 bar} \\ D_U = D_{Th} &= 9.25 \times 10^{-1} \exp\{239 \text{ kJ mol}^{-1}/RT\} \\ &\quad 4.5 \% \text{ H}_2\text{O, 10 kbar (Eyring)} \end{aligned}$$

These lines are shown in Fig. 1; also shown are the diffusivities predicted by the Eyring equation $D = kT/\zeta\lambda$ (using viscosity ζ from Shaw (1972) at 10 kbar, measured viscosities from Dingwell *et al.* (1992) at 1 bar; jump distance $\lambda = 2.3 \text{ \AA}$; $k = \text{Boltzmann's constant}$). The relation we have chosen for D_U and D_{Th} in hydrous melts at 10 kbar is that predicted by the Eyring equation, because regressed lines do not differ significantly

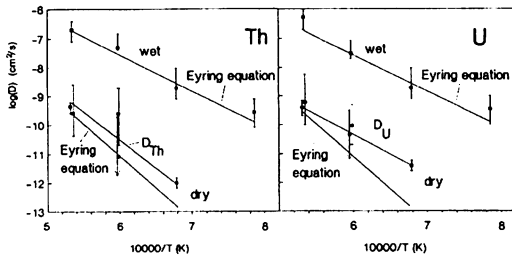


FIG. 1.

from it. This correspondence probably indicates that U and Th diffusion are controlled by the same thermally activated processes that control viscosity. Th is moderately well predicted by the Eyring equation in the anhydrous melts. However, the difference in activation energy of D_U from that of D_{Th} and viscous flow at 1 bar may indicate decoupling of D_U from melt viscosity (transition to intrinsic diffusivity). This observation suggests that U diffusivities at the low temperatures relevant to nuclear waste leaching could be very much higher than equations 2) and 4) suggest. The inference of decoupling hinges on only one measurement at 1200°C; further work will test this possibility with experiments at lower temperatures.

Oxygen fugacity was invoked by Dunn (1987) as a major control on the diffusivity of U in borosilicate glass. In our experiments, oxygen fugacity ranged from 0.21 in the 1 bar experiments to lower values presumed to lie near NNO in the 10 kbar experiments (unbuffered). Over this range in oxygen fugacities U is expected to change from a hexavalent state (uranyl ion UO_2^{2+}) at high f_{O_2} to a mix of pentavalent and quadrivalent ions at lower f_{O_2} ; this change may be responsible for the apparent transition from extrinsic to intrinsic diffusion between 10 kbar and 1 bar, since uranyl ion has a much lower charge density than do U^{4+} and U^{5+} . An alternative explanation is that at low water contents (high viscosity) we are observing the intrinsic diffusivity of U whereas at high water contents (low viscosity) the increased mobility of network formers overtakes the intrinsic diffusivity, resulting in Eyring behaviour. An interesting consequence of this hypothesis is that as a magma cools and degasses the diffusivities of U and Th may progress from being

subequal to showing differences of several orders of magnitude.

The data show that isotopic decoupling of U and Th decay series by differences in diffusivity are not expected in hydrous silicic melts over a very wide range of temperatures, pressures and water contents, but that at magmatic temperatures in water-poor magmas transport of U may be significantly faster than that of Th, resulting in a potential for U–Th fractionation.

In a reconnaissance experiment at 1400°C, 10 kbar, dry, the estimated diffusivities of both U and Th are not significantly different from those calculated at the same temperature for the 1 bar experiment. The apparent slight offset to higher values may result from the diminished melt viscosity at higher pressure, but confirmation of this suggestion would require further work.

Conclusion

Diffusion of U and Th in haplogranitic melt is Arrhenian over the temperature range 1000 to 1600°C, with diffusivities in water-rich melts predicted by the Eyring equation. At high oxygen fugacities and low water contents the diffusion of Th may remain coupled to melt viscosity, but U diffusion appears to be intrinsic, with diffusivity one to two orders of magnitude faster than the predicted Eyring diffusivity at 1200°C. U and Th may therefore become fractionated by transport processes under these conditions. Clarification of the effects of pressure and f_{O_2} over a wider range of temperatures is presently under way.

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