

DOE/ER/14240-75

FY 1996 PROGRESS REPORT FOR DOE GRANT DE-FG03-92ER14240

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GRANT: DE-FG03-92ER14240

TITLE: Energetics of Melts from Thermal Diffusion Studies

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Objectives: This research program characterizes mass transport by diffusion in geological fluids in response to thermal, solubility, and/or chemical gradients to obtain quantitative information on the thermodynamic and kinetic properties of multicomponent systems.

Projects and Results of the Last Year: Thermal diffusion is the phenomenon of chemical migration in response to heat flow along a thermal gradient. The details of this response are conditioned by the thermochemical properties and constitution of the substance. Silicate liquids undergo substantial thermal diffusion (Soret) differentiation, while the response in sulfide, carbonate, and aqueous fluids to an imposed temperature gradient is varied. The experimental observations of this differentiation are used to evaluate the form and quantitative values of solution parameters, and to quantify ordinary diffusion coefficients, heats of transport, and activation energies of multicomponent liquids. The diffusion, solution, and element partition coefficients determined for these geological fluids form a data base for understanding magmatic crystallization behavior and for evaluating geothermal, ore deposit, and nuclear waste isolation potentials. Thermal and isothermal diffusion experiments are conducted using the one atmosphere gas mixing apparatus, rapid-quench cold seal apparatus up to 0.3 GPa, piston cylinder device at pressures between 0.7-3.5 GPa, and a simplified multianvil device above 3.5 GPa. The electron and ion microprobes, and the infra-red spectrometer are used in the characterization of experimental run products.

Current experimental capabilities include two 1-atm gas mixing furnaces, a rapid-quench cold seal apparatus using TZM and HZM vessels, three piston cylinder devices, and a Walker-style multianvil device driven by a 1000-ton Clifton press equipped with a computer controlled hydraulic system. Automated pressure regulation systems are now also available for the piston cylinder devices. Among the research activities of the last year we conducted thermal diffusion experiments at very high pressure on magmatic silicate liquids in an attempt to use the recovered heats of transport and chemical potentials as monitors of changing speciation with pressure. We continue to study the influence of water and carbonate species on the Soret diffusion properties naturally-occurring silicate liquids. We are expanding our study of network self diffusion to include highly polymerized melts, and we are continuing our quantitative treatment of chemical diffusion based on Darken's theory of ion mobility for Pb, U, and Th. We have also initiating a comprehensive study of thermal, self and chemical diffusion of multicomponent liquids of the system CaO-MgO-SiO₂. New collaborations with colleagues in material science are exploiting our capabilities for high pressure generation to examine the sintering behavior at elevated T and P of nano-alumina, nano-titanium, and amorphous silicon carbide and various nanocrystalline composites. Densified nanocrystalline alumina-diamond composites are shown to possess improved mechanical properties and superplasticity.

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