FINAL REPORT

ENERGETICS OF MELTS FROM THERMAL DIFFUSION STUDIES

Grant #: DE-FG03-92ER14240
PI: Charles E. Lesher
Department of Geology
University of California
Davis, CA 95616
916-752-9779
916-752-0951
cemail: lescher@geology.ucdavis.edu

PROJECT OVERVIEW

Objectives: Most processes in geology are a consequence at some level of the flow of energy or mass. Heat conduction and chemical diffusion are examples of two of these sorts of flows which are driven by temperature and chemical potential imbalances, respectively. In the general case these flows may be coupled so that, for instance, a temperature gradient may result in a flow of mass as well as heat. This effect in liquids was demonstrated by Soret (1879) and bears his name. In gases or solids the phenomenon is given the general name thermal diffusion.

It was the purpose of this research program to examine the Soret effect in molten silicates under laboratory conditions. Results of these experiments are used to evaluate the form and quantitative values of many thermodynamic and kinetic properties of silicate melts over a range of temperature, pressure, and bulk composition. We published a comprehensive review and synthesis with a microscopic theoretical explanation for the effect at low pressure in silicate liquids of geological interest. We conducted experimental investigations of molecular diffusion in the absence of a thermal gradient through experiments involving dissolution of solid silicates in molten silicate and interdiffusion of species between miscible silicate liquids. Collectively these results enable us to construct a more comprehensive model of molecular diffusion in magmatic liquids. We have applied this model to problems of magma mixing and crustal assimilation.

An important area of research not anticipated at the start of this program was the investigation of the sintering behavior of ceramic and metal nanopowders at high pressure. The application of high pressure significantly increases the driving force for densification without significant coarsening or grain growth. The resulting compacted materials have, among other notable properties, enhanced ductility and superplasticity, superior hardness and toughness, and optical transparency for normally opaque material. In recent years we have worked on a large number of nanocrystalline ceramics (Al2O3, A1N, Si3N4, SiC) and metals (W, Fe, Ni), and nanocomposites (Al2O3-SiC, Al2O3-ZrO2, Al2O3-diamond). This pioneering work has contributed fundamentally to nanomaterials fabrication and commercialization of high pressure consolidation techniques. This fruitful avenue of research was only possible because of the innovations resulting from DOE funding in support of the Experimental Petrology and Materials Synthesis Laboratory at UC Davis.

Noteworthy technical developments during DOE support are the design and testing of the MA6/MA8 multianvil high pressure/temperature equipment, of the advanced sample assemblies for the conventional piston cylinder device, and of a rapid-
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quench cold-seal apparatus. This development work has increased the accessibility of this sort of equipment to U.S. laboratories due to close collaborative efforts with our research colleagues and with Rockland Research Corporation.

Relevance to the DOE mission: Thermal diffusion and thermal migration studies of silicate liquid are relevant to the DOE mission to understand the properties and dynamics of magmas. Besides contributing to the data base of thermodynamic and transport properties as a function of magma composition for such systems, these studies contribute to our understanding of the formation of ore reserves in magmatic cumulate environments; i.e. an understanding of chromite and the platinum group element deposits which are of economic importance to catalytic conversion. Obviously, thermochemical and transport properties of magmas which are recovered from the thermal diffusion experiments are relevant to the DOE mission to acquire thermochemical and transport properties of materials in general.

The high pressure equipment development effort is certainly relevant to DOE interest in exotic materials synthesis, especially with reference to nanocrystalline materials. An industry collaboration has begun in this lab as a consequence of the availability of this equipment. These technical development efforts are also benefiting the material synthesis community in general by making multianvil equipment and new techniques accessible.

Project Output: Thirty-six papers were published and/or reported at national and international meetings during the renewal project period, which began in 1990 at the Lamont-Doherty Earth Observatory before the PI moved to UC Davis. These include a number of papers focusing on thermal diffusion and its applications in the earth sciences, including a comprehensive review of our research in silicate systems. We have shown that the Soret effect is large in silicate liquids, sensitive to liquid composition and speciation, informative about thermodynamic and transport properties, and useful in predicting trace element partitioning behavior. We have shown the compositional sensitivity of isothermal chemical interdiffusion, emphasized the contrast with self diffusion, and applied the results to the selective contamination of natural magmas. We have also been productive in the emerging areas of materials science research, especially in the fabrication of new nanocrystalline ceramics. This grant provided partial support for three theses.

BIBLIOGRAPHY

Papers:

Abstracts, Extended Abstracts, Notes, etc.:


THESIS RESEARCH SUPPORTED


DEPARTMENT OF ENERGY

PATENT CERTIFICATION

INTERIM [ ] FINAL [ ]

DOE PRIME CONTRACT/GRANT NO.: DE-FG03-92ER14240

SUBCONTRACT NO.: ______________________________________

CONTRACTOR/GRANTEE: University of California, Geology Department

ADDRESS: One Shields Avenue, Davis, CA 95616-8605

If the appropriate response is "NONE," that response must be entered in the space provided. A separate list may be used to supply the information required provided the list is specifically referenced in the space provided.

Contractor/Grantee hereby certifies that:

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2. The following is a complete list of technical reports prepared during the course of the work under this contract/grant and the DOE office to which the reports were sent:

3. There were no subcontracts or purchase orders involving research, development and demonstration except as follows: (If such subcontract or purchase order was issued, please include a copy.)

4. No inventions or discoveries were conceived or first actually reduced to practice in performance of work under this contract/grant other than the following: (Certification includes does not include all subcontracts):

   Contractor Docket No. Title Inventor(s) Date Reported to DOE DOE Docket No.

5. The completion date of this contract/grant is: January 31, 1997

6. The following period is covered by this patent certification:

   From: January 1, 1992 To: January 31, 1997

   Date: November 5, 1998

   Signature: Charles Lesher

   Typed/Printed Name: Charles Lesher

   Title: Principal Investigator

   Telephone No.: 530-752-9779

Please submit directly to:
William C. Dorough, Patent Attorney,
U.S. Department of Energy, Office of Chief Counsel
Intellectual Property Law Division
P. O. Box 608, L-376
Livermore, California, 94550
(510) 422-4367