

DIFFUSION OF SIDEROPHILE ELEMENTS IN IRON-NICKEL ALLOYS AT HIGH PRESSURE AND TEMPERATURE

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Introduction: Siderophile elements are “iron-loving” elements that tend to partition themselves into iron metal. These elements all have characteristic signatures in the mantle, as well as in extra-terrestrial materials such as meteorites, some of which are assumed to represent differentiated primordial solar system material.

It is recognized that diffusion of siderophile elements in the metal phase is an important rate-limiting factor in many processes. Comprehensive understanding of siderophile diffusion in iron-rich metal, and related systems could lead to important time constraints on partitioning and models of core formation [e.g. 1-4]. It can also aid in understanding the siderophile element distribution observed in nature, in both terrestrial and extra-terrestrial materials.

We have performed experiments to determine the diffusivity of a variety of siderophile elements (Pd, Au, and Re) in a 90wt% Fe and 10wt% Ni alloy at high pressure and temperature. The end goals of this study are to provide a comprehensive set of siderophile diffusion coefficients and characterize siderophile diffusion in systems relevant to the applications mentioned above.

Experimental Methods: Multi anvil experiments have been conducted to determine the diffusivity of several siderophile elements (Au, Pd, and Re) at temperatures ranging from 1200°-1600°C and pressures ranging from 10 and 20 GPa.

Before actually performing diffusion experiments, starting materials were synthesized in a 0.75” end-loaded piston cylinder apparatus at RPI. Pure metal powders ranging in size from 0.5-3 microns were thoroughly mixed, and packed into drilled out MgO capsules. The powders were either pure Fe-Ni base (90% Fe, 10% Ni), or the same base doped with up to 2 wt% of a siderophile element. Synthesis runs were held at 1400°C and 1GPa for 60 hours. This was sufficient time to homogenize the powders into a γ phase Fe-Ni alloy, as was confirmed by microprobe analyses.

Upon completion of the synthesis runs, the new alloy rods were extracted from the MgO capsules, and sectioned into disks approximately 0.75 mm in thickness. One face of each disk was polished with SiC

paper and alumina powder down to a grain size of 0.3 microns. A doped wafer was then mated with a base composition wafer (polished faces touching), and placed into a multi-anvil assembly. The metal diffusion couple was placed directly into an MgO sleeve that was contained in an Re heater. Conditions of experiments ranged from 1200°C to 1600°C, and 10GPa. One exploratory experiment has been performed with Au at 20GPa, and 1400°C with a smaller assembly.

Table 1: Experimental conditions and measured diffusion coefficients

Run	Element	T (°C)	P (GPa)	t (hours)	D (m ² /s)
DMP1	Pd	1400	10	48	1.38E-14
DMP8	Pd	1200	10	72	1.31E-15
DMP9	Pd	1600	10	21.3	1.61E-13
DMP2	Au	1200	10	46.5	5.27E-15
DMP6	Au	1600	10	21.25	3.14E-13
DMP5	Au	1400	10	14	8.32E-14
DMP15	Au	1400	20	48	3.12E-15
DMP14	Re	1500	10	48	2.39E-14
DMP17	Re	1350	10	20	2.37E-15

Analytical Methods: Upon completion of the experiments, the samples were mounted in epoxy and sectioned, then polished with SiC paper and Alumina powder down to 0.3 microns. Diffusion profiles were measured with the JEOL 8900 electron microprobe located at the Geophysical Lab. The accelerating voltage was 15 kV and the cup current was 50nA. Analyzed spots were 15 microns apart. Count times were 30s on peaks and 15s on backgrounds. All standards used were pure metals.

Results and Discussion: The diffusion couple configuration used in these experiments is that of a semi-infinite, one-dimensional diffusion couple. It is assumed that there are no complications such as multi-component effects. It is also assumed that the resulting concentration profiles are symmetric around $x=0$,

(There is no compositional dependence of the diffusion coefficient). This type of problem has a standard solution which can be expressed as:

$$C(x,t) = \frac{C_1}{2} \operatorname{erfc}\left(\frac{x}{\sqrt{4Dt}}\right)$$

Where $C(x, t)$ is the concentration at a given point, C_1 is the concentration at the end, x is the distance from the interface (m), t is the time (s) and D is the diffusion coefficient (m^2/s). Diffusion coefficients were found using a non-linear least squares fit by a Levenberg-Marquardt method in Origin™ by Microcal™. The experimental conditions and results are listed in Table 1.

Figure 1 shows an Arrhenius plot of the 10 GPa experiments. There is approximately an order of magnitude range of diffusivities between the elements at any given temperature. The activation energy, E , is similar for all three elements (around 300 kJ/mol), as defined by the relation:

$$D = D_o \exp\left(\frac{-E}{RT}\right)$$

where D_o is a pre-exponential factor (m^2/s), R is the gas constant, T is temperature (K), and D is the measured diffusivity (m^2/s).

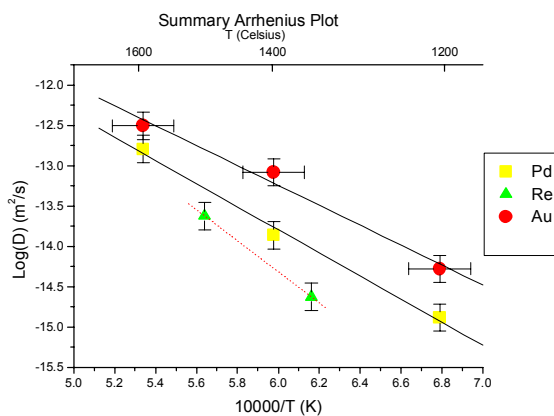


Figure 1: Arrhenius plot of 10 GPa experiments for Au, Pd, and Re

Figure 2 shows a preliminary relationship between pressure and diffusivity. The diffusivity decreases with increasing pressure, but more experiments will be needed to define the trend. The 1 GPa experiment on this plot is taken from similar piston cylinder experiments performed at the RPI lab, [5].

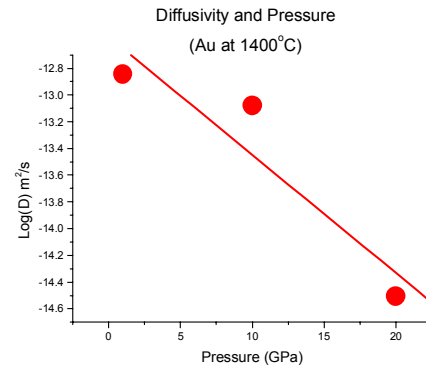


Figure 2: The effect of pressure on diffusivity of Au at 1400°C

Although the conditions studied here do not bear directly on deep planetary interiors, the preliminary results presented suggest that this method of investigation holds promise for the development of a larger diffusion database, which could be more applicable. There are, however, a few interesting calculations that can be examined from these data. For instance, by looking at both the negative effect of pressure and the positive effect of temperature, the change in diffusivity with depth in the earth can be calculated, although it may not yet be reasonable to extrapolate these values to core conditions. It is also possible to calculate approximate length scales of diffusion for these elements over a period reasonable for core formation events.

Conclusions and Future Outlook: These data represent the first in an on going study with the aim of providing a comprehensive set of diffusion data for siderophile elements in systems that are relevant for understanding planetary core formation. As we become more familiar with siderophile element behavior in Fe-Ni metal, it is planned to expand the system studied to a more realistic analog of planetary core materials. This would include the addition of light elements, such as S or O, as well as performing experiments with liquid metal.

References: [1] Fleet M.E. and Stone W.E. (1990) *Geochim. Cosmochim. Acta.* **55**, 245-253. [2] Fleet, M.E., Liu M. and Crocket J.H. (1999) *Geochim. Cosmochim. Acta.* **63**, 2611-2622. [3] Jones and Drake (1986) *Nature.* **322**, 221-228. [4] Li J. and Agee C.B. (2001) *Geochim. Cosmochim. Acta.* **65**, 1821-1832. [5] Watson H.C. and Watson E.B. (2001) *AGU Fall Meeting*, Abstract V22A-1016.