

Diffusion and solubility of nitrogen in olivine

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Earth's mantle contains ~60% of our planet's nitrogen and holds on to it tenaciously [1]. Beyond this broad constraint, however, little is known about N transport into or out of the mantle, nor about the cause and persistence of N isotopic heterogeneity over vast time periods. For these reasons, we have initiated an experimental study of the diffusion characteristics and solubility of N in the major silicate phases of the mantle, beginning with olivine.

Oriented slabs of San Carlos olivine (~1×3×3 mm) were prepared with one surface (010) polished using colloidal silica. Diffusion experiments were set up by placing olivine slabs in pressure-sealing Ag containers surrounded by ¹⁵N-labeled 4-aminobutyric acid, which is ~6 mole% ¹⁵N (use of ¹⁵N eliminates the risk of environmental contamination and enables measurement of N in olivine using nuclear reaction analysis). The Ag containers were subjected to 1 GPa pressure and 750-850°C in a piston-cylinder apparatus for several days. The aminobutyric acid is believed to have decomposed almost immediately during the high P-T treatment to graphitic residue and N-bearing C-O-H fluid, which served as the source of N diffusing into the olivine. After quenching the experiments and extracting the intact olivine slabs from the Ag containers, the polished surfaces were depth-profiled for ¹⁵N concentration using the ¹⁵N(p,α γ)¹²C nuclear reaction. The resulting diffusive-uptake profiles reveal near-surface N concentrations of 500-1000 ppm and diffusivities of ~3E-22 to 3E-21 m²/s. The small temperature range covered to date precludes accurate estimate of the activation energy for diffusion, but it is loosely constrained at 240±100 kJ/mol.

The speciation of N diffused into San Carlos olivine and the likelihood of its substitution on normal lattice sites are currently unknown. However, the reducing conditions of the experiments (elemental C present) and the high apparent solubilities probably argue against N-O species or large NH₄⁺ ions. The preliminary data suggest that diffusive transport of N in the mantle by volume diffusion may be limited to length scales of tens of centimeters in 100 MYr.

[1] Cartigny and Marty (2013) *Elements*. **9**, 359-366.