Do grain boundaries act as a water reservoir in Earth's mantle?

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Earth’s mantle is predicted to contain as much or more water as its hydrosphere, which is important because the presence of water lowers the viscosity of mantle rocks. How water is distributed within mantle rocks is therefore fundamental to understanding Earth’s geodynamic behaviour, but the picture currently remains unclear. On the grain scale, previous analyses have revealed incompatible element partitioning in grain boundaries using EPMA, and the presence of H-enriched regions close (tens of µm) to grain boundaries using synchrotron-based FT-IR. The results of such studies have been used to suggest that grain boundaries may store water in concentrations hundreds of times higher than in grain interiors. Chemical segregation at grain boundaries is generally accepted to influence grain boundary diffusivity, which in turn affects the bulk viscosity of materials deforming by diffusion creep, a mechanism which is predicted to dominate the deformation of large parts of the mantle.

This study was designed to directly image the distribution of heavy water (D2O) at the nanoscale in a synthetic peridotite sample using high-resolution secondary ion mass spectrometry (NanoSIMS), for the first time. The sample was annealed at temperature and pressure conditions typical of Earth's upper mantle (1250 °C, 0.3 GPa) for three hours, to facilitate diffusion of 2H into olivine and pyroxene grains. Preliminary NanoSIMS results suggested that the partitioning of 2H into grain boundary regions, where observed, was at least an order of magnitude lower (partition coefficient of ~101) than has previously been predicted, indicating that, for typical mantle grain sizes, grain boundaries do not act as a significant storage reservoir for water in Earth's mantle (or those of other terrestrial planets). The initial data were limited to a relatively small number of boundaries per sample. In this phase of the study, electron backscatter diffraction data has been collected from a single sample to characterise grain (mis)orientations at multiple sites suitable for NanoSIMS analyses. This is necessary because the strength of the chemical signature collected within grains and at grain boundaries using NanoSIMS is dependent on the orientations of those grains with respect to the NanoSIMS beam (matrix effects), the angle of the grain boundary with respect to the sample surface, and the misorientation angle between the two grains that comprise the boundary of interest. 2H, 16O, 16O2H and 28Si measurements will be collected from multiple boundaries by NanoSIMS using a Cs+ beam with a 100 nm diameter to quantify grain boundary partitioning. The NanoSIMS isotope profiles will be presented as 2H/28Si ratios to account for variation in measured isotope concentrations due to matrix effects. The results of the analysis will
help quantify the degree to which water undergoes grain boundary segregation in mantle rocks under equilibrium partitioning conditions.