Geophysical Research Abstracts
Vol. 16, EGU2014-10677-2, 2014
EGU General Assembly 2014

Combined measurement of surface, grain boundary and lattice diffusion coefficients on olivine bi-crystals

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Diffusion along interface and grain boundaries provides an efficient pathway and may control chemical transport in rocks as well as their mechanical strength. Besides the significant relevance of these diffusion processes for various geologic processes, experimental data are still very limited (e.g., Dohmen & Milke, 2010). Most of these data were measured using polycrystalline materials and the formalism of LeClaire (1951) to fit integrated concentration depth profiles. To correctly apply this formalism, certain boundary conditions of the diffusion problem need to be fulfilled, e.g., surface diffusion is ignored, and furthermore the lattice diffusion coefficient has to be known from other studies or is an additional fitting parameter, which produces some ambiguity in the derived grain boundary diffusion coefficients. We developed an experimental setup where we can measure the lattice and grain boundary diffusion coefficients simultaneously but independent and demonstrate the relevance of surface diffusion for typical grain boundary diffusion experiments.

We performed Mg_2SiO_4 bicrystal diffusion experiments, where a single grain boundary is covered by a thin-film of pure Ni_2SiO_4 acting as diffusant source, produced by pulsed laser deposition. The investigated grain boundary is a 60° (011)/[100]. This specific grain boundary configuration was modeled using molecular dynamics for comparison with the experimental observations in the transmission electron microscope (TEM). Both, experiment and model are in good agreement regarding the misorientation, whereas there are still some disagreements regarding the strain fields along the grain boundary that are of outmost importance for the strengths of the material. The subsequent diffusion experiments were carried out in the temperature range between 800° and 1450°C. The inter diffusion profiles were measured using the TEMs energy dispersive x-ray spectrometer standardized using the Cliff-Lorimer equation and EMPA measurements.

To evaluate the obtained diffusion profiles we adapted the isolated grain boundary model, first proposed by Fisher (1951) to match several observations: (i) Anisotropic diffusion in forsterite, (ii) fast diffusion along the grain boundary, (iii) fast diffusion on the surface of the sample. The latter process is needed to explain an additional flux of material from the surface into the grain boundary. Surface and grain boundary diffusion coefficients are on the order of 10000 times faster than diffusion in the lattice. Another observation was that in some regions the diffusion profiles in the lattice were greatly extended. TEM observations suggest here that surface defects (nano-cracks, ect.) have been present, which apparently enhanced the diffusion through the bulk lattice.

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