Ultra-high resolution of experimental diffusion profiles using local electrode atom probe tomography

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A major limitation in applying experimental diffusion data to geological materials, which commonly exhibit slow diffusion, is that traditional analytical techniques can only resolve diffusion profiles several tens of nm in length. As a result, experiments are commonly run at relatively high temperatures (e.g. \(\sim\)1200-1500 °C) and the retrieved Arrhenius trends are extrapolated down to more geologically relevant conditions, introducing significant error into calculations using these data. Taking the simple system of calcium in synthetic forsterite, we present the first experimentally induced diffusion profiles measured using local electrode atom probe (LEAP) tomography.

Due to it’s widespread occurrence, high thermal stability and relatively simple anisotropic properties, olivine is commonly utilized for geospeedometry in both terrestrial igneous rocks and meteorites. Previous work [1] has shown that Ca diffusion is more than one order of magnitude slower than Fe-Mg diffusion in olivine at > 900 °C, which means that Ca diffusion profiles can be used to complement Fe-Mg speedometry or provide timescale information after Fe-Mg profiles have been thoroughly re-equilibrated.

We have conducted Ca diffusion experiments along [100] in synthetic forsterite at 1 atm. from 750 – 1300 °C, with silica activity (aSiO\(_2\)) buffered by either forsterite–diopside–enstatite or forsterite–diopside–periclase. The resulting profiles were measured via SIMS depth profiling using the SwissSIMS 1280-HR and laser step scanning using the ASI resolution 193 nm laser coupled to the Thermo-Element XR sector field spectrometer housed at the University of Lausanne. Time-series experiments have been carried out at 750, 1100 and 1300 °C. Our results are in excellent agreement with the dataset of [1], and expand the range of experimental temperatures reported in that study. In contrast to other divalent cations in olivine [2], there appears to be no dependence of Ca diffusion on aSiO\(_2\). This observation could be explained by Ca ordering onto the M2 site in olivine, while M-site vacancies prefer the M1 site [3].

Following our successful reproduction of the results of [1] using SIMS, we have analyzed the 750 °C time series samples using the 4000x-HR local electrode atom probe (LEAP) house at the ScopeM facility at ETH Zürich. The retrieved diffusion coefficients are well within the error of those obtained using SIMS, demonstrating that LEAP tomography is a viable means to analyze experimentally induced diffusion profiles. The extremely high spatial resolution and relatively high sensitivity attained by LEAP tomography should enable the execution of many diffusion experiments under the same conditions to which the data are ultimately applied.