American Mineralogist, Volume 93, pages 863–874, 2008

A new experimental thin film approach to study mobility and partitioning of elements in grain boundaries: Fe-Mg exchange between olivines mediated by transport through an inert grain boundary

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Abstract

A new experimental method is presented where transport and partitioning of elements in grain boundaries is investigated by using thin films. As a model system, the element exchange between a nanocrystalline thin film of olivine (Fo_{30}) and a San Carlos olivine single crystal (Fo_{89}), which is mediated by grain diffusion through a polycrystalline ZrO_2 layer was studied. Double layers of ZrO_2 and olivine on polished single crystals have been prepared by pulsed laser deposition and annealed in the temperature range between 900 and 1050 °C at controlled oxygen fugacity. Starting and annealed samples were analyzed by SEM, RBS, and SIMS. From the chemical analysis by RBS and SIMS, the reaction mechanism could be classified according to the reaction mechanism map of Dohmen and Chakraborty (2003). Most of the samples show a combined grain boundary/volume diffusion-controlled reaction behavior according to this scheme.

During the anneal, a linear concentration gradient for the Fe/(Fe + Mg) ratio evolved within the ZrO_2 layer, where Fe strongly prefers the ZrO_2 grain boundary compared to Mg. The depth profiles for the Fe/(Fe + Mg) ratio have been simulated in detail. From the simulation Fe-Mg volume (in olivine) and grain boundary diffusion coefficients, as well as the partition coefficient and the storage capacity for the grain boundary, have been extracted. The chemical diffusion coefficient of Fe-Mg within the ZrO_2 grain boundary at 900 °C is on the order of 1×10^{-15} m^2/s. Volume and grain boundary diffusion coefficients obtained using the new method are in good agreement with literature data. An additional observation is that grains within the polycrystalline fayalite-rich olivine layer grow by a grain rotation mechanism until an equilibrium texture is attained.

Keywords: Diffusion, kinetics, element exchange, thin films, olivine, zirconia