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Compositional dependence of Fe-Mg inter-diffusion in orthopyroxene

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Despite the wide range of applications for diffusion data of Fe-Mg in pyroxene, existing experimental data [1] do not fully constrain the effect of the compositional variations that natural orthopyroxene show. Different calibrations of the effect of X_{Mg} on the diffusion coefficient of Fe-Mg, $D_{Fe\text{-}Mg}$, have been suggested (e.g., [1,2]), emphasizing the need for experimental observation. We explored a new experimental approach to determine the effect of compositional parameters on $D_{Fe\text{-}Mg}$, in particular X_{Mg} and Al content.

Diffusion couples consisting of ferrosilite-rich thin films (≈ 1 μm) on gem-like quality orthopyroxene single crystals crystallographically oriented parallel to the [001] axis, were produced by pulsed laser deposition [3]. Diffusion experiments were carried out at atmospheric pressure in vertical gas mixing furnaces at temperatures between 950 and 1100 °C. The fO_2 was controlled by a flowing gas mix of CO and CO_2 and monitored throughout the experimental runs. Cross sections of the diffusion couples were analysed using combined BSE imaging and EDX mapping on FIB-lamellae with an approximate thickness of 100 nm. The calibration of this analytical approach on thin film reference samples, demonstrated that we can resolve concentration gradients within 25 nm.

During the diffusion anneal the films crystallised to a polycrystalline layer composed mostly of Fs-rich orthopyroxene and some fayalite-rich olivine crystals with sizes around 100 – 300 nm, approximately. The concentration profiles were fitted using a 1-D diffusion model that accounts for compositional dependence of the diffusion coefficients and mass balance between film and substrate. In addition, film and the substrate were treated as two separate diffusion media. With this experimental approach we are able to better constrain the effect of X_{Mg} on $D_{Fe\text{-}Mg}$. Preliminary data suggest that the diffusion rates at the X_{Mg} of the single crystal are consistent with earlier determinations; however, the compositional dependence appears to be greater than previously estimated [2].

[1] Dohmen et al. (2016) *American Mineralogist* 101.10, 2210-2221.

[2] Ganguly and Tazzoli (1996) *American Mineralogist* 79(9-10), 930-937.

[3] Dohmen et al. (2002) *Eur. J. Min.* 14, 1155-1168.