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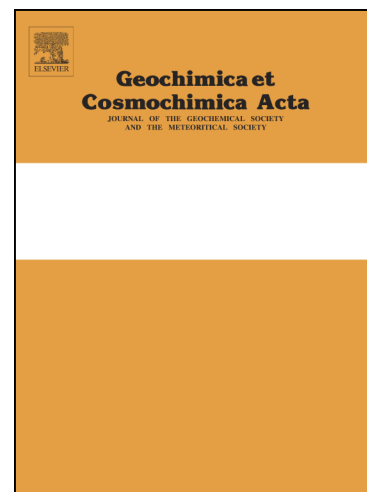
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Rates of dehydration of olivines from San Carlos and Kilauea Iki

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Abstract

The rate at which H⁺ moves into, through, and out of olivine is of great interest for the deep water cycle, assessing the fidelity of olivine-hosted melt inclusions as recorders of pre-eruptive melt compositions, and estimating the decompression rate of magma during volcanic eruptions. Here we conduct a series of experiments and observations on natural Fe-bearing olivine to better understand and quantify this H⁺ mobility, with a particular focus on the rate at which H⁺ diffuses out of olivine. We have carried out sequential dehydration experiments on single crystals of natural, Fe-bearing olivine polished and oriented along three crystallographic orientations. These efforts include four separate parts: (1) the partial hydration of polished blocks of xenolithic San Carlos olivine at 800°C and 1000°C, 1 GPa, and oxygen fugacity (fO₂) at the Ni-NiO buffer (NNO); (2) sequential dehydration of a homogeneous block of San Carlos olivine at 800°C, 1-atm, and fO₂ of NNO-2.6; (3) sequential dehydration of a polished block of an olivine phenocryst from Kilauea Iki at 800 and 1000°C, 1 atm, and fO₂ held at NNO-2.6 until the final step, which was conducted at NNO+1.9; and (4) the measurement by secondary ion mass spectrometry of hydrogen zonation profiles in an olivine phenocryst from Kilauea Iki coupled with Monte Carlo modeling to determine total H diffusivities in a natural setting. Hydrogen zonation profiles were measured in all three crystallographic directions *a*, *b*, and *c* through the uncut blocks before and after each time step for all experiments by Fourier transform infrared spectroscopy using light polarized || *a* and modeled in 3 dimensions to determine both total and peak-specific dehydration, producing over 50 sets of 3 apparent diffusivities.

The results show that the apparent diffusivity of total H⁺ varies both over time and for olivines with different defect populations, and thus H⁺ does not diffuse at inherently site-specific rates. These variations are due to rapid reactions that may occur between sites as a function of changing defect concentrations. Our experiments reveal three new phenomena: (1) H⁺ that is associated with the Ti-clinohumite defect, [Ti-2H], can enter and exit olivine at the fastest rate measured. (2) The rate of H⁺ loss from specific infrared absorption peaks, and in total, may change with progressive dehydration due to the buildup of Fe³⁺ and redistribution of defects, particularly the rehydration of [Ti-2H]. (3) The following Arrhenius laws appear to apply to total H⁺ diffusion out of most natural olivines that have been studied experimentally and naturally during dehydration: $D_a = 10^{-5.4} \exp(-130/RT)$; $D_b = 10^{-6.9} \exp(-130/RT)$; and $D_c = 10^{-6.6} \exp(-130/RT)$, where *R* is the gas constant 8.314 J/mol K; *T* is the temperature, the units of the diffusivities (*D*) are in m²/s; and the activation energy is 130 kJ/mol. This rate is slower than proton-polaron but faster than proton-metal vacancy diffusion and appears to characterize olivine of Fo~90 composition in a wide range of conditions, from self-diffusion, to melt inclusion re-equilibration, to mantle xenolith and volcanic phenocryst zonation during ascent.

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