

## Water incorporation and site-specific diffusion in garnets

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We performed diffusion experiments on natural gem-quality spessartine (300 ppm H<sub>2</sub>O) and grossular (800 ppm H<sub>2</sub>O). The chemical composition of garnet influences the peak positions in infrared spectra, which are lower in wave numbers for spessartine than for grossular. The experiments were conducted on cubes of 1.5\*1.5\*1.5 mm<sup>3</sup>, at 1 atm over a temperature range of 750-1050°C under both air and various lower oxygen fugacities buffered by CO/CO<sub>2</sub> gas mixes (QFM+8 to QFM-3). Water diffusion profiles were measured by FTIR with 10 μm resolution in a slice through the center of the cube.

Water diffusion is fast in air but absent at our experimental timescales at low  $f(\text{O}_2)$  indicating that H diffusion is coupled to the very fast oxidation reaction  $\text{M}^{2+} + \text{OH}^- \rightarrow \text{M}^{3+} + \text{O}^{2-} + 0.5 \text{H}_2$ , where M is Fe or Mn. The activation energy ( $E_a$ ) of this exchange has been determined with experiments at 950, 850 and 750°C.  $E_a$  is low, (158kJ/mol) consistent with a diffusion mechanism that doesn't involve metal vacancies. An automated deconvolution program was developed to resolve the relative contributions of each FTIR peak to diffusion profiles. Absorption bands decrease at different rates during H diffusion allowing identification of "families" of bands that belong to the same point defects. Some bands grow during the experiment. Our data provide evidence for up to four different water incorporation mechanisms in garnet. Apart from H in silicon vacancies, our results suggest water related to trivalent cations, water in dodecahedral vacancies as well as water associated with minor Ti.