

## Hydrogen Incorporation into Olivine at 9-15 GPa and 1100-2000°C

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The hydrogen solubility and hydrogen incorporation mechanism into  $(\text{Mg,Fe})_2\text{SiO}_4$  olivine have been studied at pressures of 9-15 GPa and temperatures of 1100-2000°C. Samples were synthesized using a multianvil apparatus from  $\text{Fo}_{88} + 15 \text{ wt.}\% \text{H}_2\text{O}$  and  $\text{Fo}_{88} + 15 \text{ wt.}\% \text{H}_2\text{O}$  mixture without controlling oxygen fugacity. Samples were studied by Fourier Transform Infrared spectroscopy (FTIR) and H ( $\text{H}_2\text{O}$ ) contents were calculated from IR spectra using the calibration by Bell et al. (2003).

The hydrogen solubility increases with pressure, but the temperature dependence is complex. The hydrogen solubility increases from 1100 to 1200°C and then decreases from 1400 to 2000°C. Maximum hydrogen solubility, equivalent to 6300 ppm  $\text{H}_2\text{O}$ , was determined in olivine at 14 GPa and 1200°C. At temperatures of 1800-2000°C the  $\text{H}_2\text{O}$  contents of olivine is relatively low (150-400 ppm). Hydrogen solubility in  $\text{Fo}_{88}$  is consistent with that in pure forsterite studied by Lemaire et al. (2004) and Smyth et al. (2006) indicating minor influence of Fe on hydrogen incorporation.

The IR spectra of olivines are composed of several OH absorption bands at 3160-3613  $\text{cm}^{-1}$ . IR spectra with the electric field vector parallel to the a-axis show the strongest absorbance, dominated by peaks at 3613 and 3579  $\text{cm}^{-1}$ . Although some structural calculations are consistent with hydrogen incorporation into octahedral vacancies in olivine (e.g. Smyth et al., 2006; Kudoh et al., 2006) we strongly suggest that majority of observed peaks (at 3533, 3548, 3566, 3579, and 3613  $\text{cm}^{-1}$ ) are due to hydrogen associated with Si-vacancies. This is consistent with microprobe analyses of the samples, which show a clear deficit of Si in olivines at the highest  $\text{H}_2\text{O}$ -contents. Minor bands at 3300-3400  $\text{cm}^{-1}$  may be due to  $\text{Fe}^{3+}$  substitution and those at 3400-3500  $\text{cm}^{-1}$  due to vacancies in Mg structural positions. The peak at 3598  $\text{cm}^{-1}$  previously attributed to Mg-vacancies should be addressed to Si-vacancies. The strongest peaks corresponding to Mg vacancies (or OH interstitials) at 3160 and 3220  $\text{cm}^{-1}$  are very weak. However, at 1800-2000°C these peaks are more pronounced. These conclusions are consistent with molecular dynamic calculations (e.g. Brodholt and Refson, 2000) and IR observations of olivines synthesized under controlled  $\text{SiO}_2$  and MgO activities (Matveev et al., 2001; 2005; Lemaire et al., 2004; Berry et al., 2005).

Significant hydrogen incorporation on Si-site vacancies in olivine is important for hydrogen diffusion models for olivine, which have been studied only for the case where hydrogen incorporation into Mg-Fe-vacancies dominates. A revision of structural refinements of hydrous olivine might be necessary. Discrimination of IR absorption bands between Si- and Mg-vacancies in the olivine structure has important implication for natural olivines in kimberlites, to determine the style of mantle metasomatism (e.g. silicate melt metasomatism, with high  $\text{SiO}_2$  activity, versus carbonatitic, with high activity of MgO), relation to their depth of origin, oxidation state, and geodynamic setting.