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### **Orthopyroxene: the Most Hydrous Nominally Anhydrous Upper Mantle Phase?**

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According to a recent study by Mierdel et al. [1], orthopyroxene, the second most abundant phase in the upper mantle, can incorporate up to almost 1 wt % H<sub>2</sub>O at high P and T. The data indicate a rapid decrease in storage capacity with increasing pressure from 1.5 to 3.5 GPa, corresponding to the well-known trend of decreasing Al content with increasing pressure in opx. This could have important implications for stabilizing hydrous partial melts, commonly thought to be responsible for the Earth's asthenosphere [1]. However, the OH concentrations measured by Mierdel et al. conflict with several previous studies that obtained much lower amounts, at comparable Al contents (up to ~5 wt % Al<sub>2</sub>O<sub>3</sub>, e.g. [2-5]). To investigate this discrepancy, we conducted new experiments on a suite of 10 natural orthopyroxene starting materials with variable Fe/Mg ratios (Mg# 77.5-99.5) and Al contents (0.13-4.76 wt % Al<sub>2</sub>O<sub>3</sub>). The experimental conditions were 900 °C and 1.5 or 3 GPa, with fO<sub>2</sub> fixed at NNO. Run durations were chosen so as to achieve equilibrium according to available diffusion data for H in opx. Retrieved crystals were analyzed for OH and major element concentrations, taking special care to check for concentration gradients. Whereas the rims of some crystals are characterized by Fe loss, there is remarkable consistency in OH concentrations among different crystals from the same charge, and a lack of gradients in OH concentration. Differences in IR spectra compared to the starting materials, combined with electron probe data, indicate that the coordination environment of Al associated with H in most samples changes from dominantly tetrahedral towards octahedral coordination at high pressure. We obtained a maximum concentration of about 650 ppm H<sub>2</sub>O at 3 GPa in opx with 4.76 wt % Al<sub>2</sub>O<sub>3</sub> (compared to 400 ppm at 1.5 GPa in the same material). This contrasts with concentrations of 1420-6400 ppm H<sub>2</sub>O measured by Mierdel et al. for Fe-free opx with similar Al concentrations. The discrepancy between Mierdel et al.'s results and ours, as well as those of other studies on both Fe-free and Fe-bearing opx [2-5], remains unresolved. One possibility is that the diffusion of H in orthopyroxene occurs by multiple mechanisms, as in olivine. One mechanism clearly operates rapidly enough to eliminate concentration gradients in our samples, to within the 50-micron resolution of our technique. However, it is possible that an additional, slower mechanism is involved in the achievement of reversible equilibrium, perhaps leading to higher OH contents such as those observed by Mierdel et al. On the other hand, the extreme variations in Al concentration (up to 50%) and OH concentration (up to 40%) in crystals grown under the same conditions by Mierdel et al. suggest the possibility that rapid growth under non-equilibrium conditions could trap metastable concentrations of point defects. [1] Mierdel et al., 2007, Science, 315, 364-368 [2] Rauch and Keppeler, 2002, CMP, 143, 525-536 [3] Koga et al., 2003, G3, 4. DOI: 10.1029/2002GC000378 [4] Stalder, 2004, EJM, 16, 703-711 [5] Stalder et al., 2005, CMP, 150, 473-485