Decrease of hydrogen incorporation in forsterite from CO$_2$-H$_2$O-rich kimberlitic liquid

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**Abstract**

To test if hydrogen incorporation by ionic diffusion can occur between a volatile-rich kimberlitic liquid and forsterite, results of high-pressure and high-temperature experiments using a piston-cylinder apparatus at 1200–1300 °C and 1 GPa for durations of 1 min, 5 h, and 23 h, are reported here. Kimberlitic liquid in the system CaO-MgO-Al$_2$O$_3$-SiO$_2$-CO$_2$-H$_2$O and synthetic forsterite single crystals were chosen as a first simplification of the complex natural kimberlite composition. Unpolarized Fourier transform infrared spectroscopy was used to quantify the concentrations of OH in the crystallographically oriented forsterite. Scanning electron microscopy, electron backscattered diffraction, electron microprobe analyses, and transmission electron microscopy were performed to identify the run products. After 5 and 23 h, a forsterite overgrowth crystallized with the same orientation as the initial forsterite single crystal. The kimberlitic liquid has crystallized as micrometer-scale euhedral forsterite neocrystals with random crystallographic orientations, as well as a nanoscale aluminous phase and a calcic phase. Despite theoretical water-saturation of the system and long duration, none of the initial forsterite single crystals display signs of hydration such as hydrogen diffusion profile from the border toward the center of the crystal. Most likely, the presence of CO$_2$ in the system has lowered the H$_2$O fugacity to such an extent that there is no significant hydration of the starting forsterite single crystal or its overgrowth. Also, the presence of CO$_2$ enhances rapid forsterite crystal growth. Forsterite growth rate is around 2 × 10$^8$ μm$^3$/h at 1250 °C. These experimental results suggest a deep mantle origin of the high OH content found in natural mantle-derived xenoliths transported in kimberlites, as reported from the Kaapvaal craton. In agreement with previous studies, it also points out to the fact that significant hydration must take place in a CO$_2$-poor environment.

**Keywords:** Kimberlite, water, hydrogen, diffusion, olivine, point defect

**Introduction**

Incorporation of hydrogen in nominally anhydrous minerals (NAMs), even at trace concentrations (parts per million by weight level), is known to have a significant impact on the chemical and physical properties of the rocks and minerals in Earth’s mantle such as melting temperature, electrical conductivity, seismic velocities, and rheology (e.g., Karato 1990; Mackwell et al. 1985; Hirth and Kohlstedt 1996; Demouchy et al. 2012; Gaetani and Grove 1998; Jacobsen et al. 2008). For instance, in terms of rheology, olivine controls the viscosity of the lithosphere root. The low OH$^-$ concentrations (<20 ppm H$_2$O wt, with concentration calculated using the FTIR calibration of Paterson 1982) of olivine have been proposed to increase the mantle viscosity of the South African craton, and hence could be responsible for its longevity (Peslier et al. 2010; Baptiste et al. 2012). To test if it is also the case for other cratons, concentration of hydrogen was recently measured in mantle peridotites from the Siberian craton, but the results have not confirmed the observation from the South African craton (Doucet et al. 2014). Prior to these recent studies, several publications have reported high concentrations of OH (>30 ppm H$_2$O wt) in olivine from mantle xenoliths, olivine megacrysts, and kimberlitic olivine phenocrysts from several cratons (e.g., South Africa, Siberia, and Canada; see Miller et al. 1987; Matsuysk et al. 1998; Bell et al. 2004; Matsyuk and Langer 2004; Kamenetsky et al. 2008; Peslier et al. 2010; Peslier 2010; Doucet et al. 2014). In particular, relatively high concentrations of OH in olivine (up to 150 ppm H$_2$O wt) have been reported in olivine of mantle xenoliths from the Kaapvaal craton (Peslier et al. 2010; Baptiste et al. 2012), except for the deepest samples, which are almost anhydrous (<10 ppm H$_2$O wt). Further, high concentrations of OH in olivine (54–419 ppm H$_2$O wt) have been measured in megacrysts and macrocrysts from kimberlites (Bell et al. 2004; Matsyuk and Langer 2004; Matveev and Stachel 2007) suggesting that kimberlite would be a favorable environment for important hydrogen incorporation in olivine.

Studies on Kaapvaal mantle xenoliths usually report homogeneous OH concentration profiles across olivine grains suggesting the absence of hydration or dehydration process caught-in-the-act (Peslier et al. 2010; Baptiste et al. 2012). However, the origin of hydrogen in olivines from the cratonic mantle still remains unclear. High concentration of OH in olivine in Kaapvaal xenoliths could have been inherited from (1) metasomatism, if water-saturated fluids or water-saturated