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Hydrogen partitioning between synthetic olivine, orthopyroxene and melt

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Olivine and pyroxene are the dominant silicate phases of the Earths lithospheric upper mantle. Although nominally anhydrous, studies of natural and synthetic crystals of both phases have shown that they usually incorporate small amounts of hydrogen. As it has been shown that the incorporation of trace hydrogen places fundamental controls upon the physiochemical properties of silicates, accurately quantifying the natural abundance of hydrogen in the lithospheric upper mantle is critical before we can understand the geodynamics of this region of the Earth. Understanding how hydrogen partitions between the geologically important phases encountered at depth represents a potentially significant tool in detailing the distribution of hydrogen in the lithospheric upper mantle of the Earth.

We describe high P-T piston cylinder experiments in which we grew from a melt olivine and opx crystals which were large enough to be analysed using polarised infrared spectroscopy. Hydrogen partitioning between olivine, opx and melt was explored as a function of different physiochemical parameters. Experiments in the system albite-forsterite-H_2O yielded large olivine and opx crystals, which were accompanied by a quenched melt. Using polarized, directional IR spectroscopy we determined that olivine and opx grown in this system at 1.5GPa and cooled to a final temperature of 1320°C contain 8 and 175 ppm H_2O respectively. Combined with data for the melt yields $D_H^{ol\text{-}opx}=0.05$ and $D_H^{ol\text{-}melt}=0.0001$. The distribution of hydrogen between olivine and opx in these phenocrysts is comparable to that previously described for these silicate phases in natural spinel lherzolite xenoliths from the upper mantle [1]. Furthermore, the partitioning of hydrogen between opx and melt described here is in broad agreement with that outlined for natural opx phenocrysts and quench glass [2]. However, the partition coefficients presented here are lower than those calculated following previous experimental study [3,4].

References

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Hydrogen-mobility in grossular-andradite solid solution

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Four natural garnets of grossular-andradite solid solution with minor amounts of the pyralspite component (Gr 84 Py 2 And 14 to Gr 1 And 99) were investigated by FTIR-spectroscopy. All samples show significant amounts of hydrogen incorporated as OH-defects (0.02 - 0.15 wt% H_2O). Stepwise annealing experiments at temperatures ranging from 700° C to 1050 °C in air and in $Ar_{90\%}/D_{10\%}$ atmosphere were performed on doubly polished single-crystal slices.

Diffusion kinetics for hydrogen were obtained by numerically fitting the change in integrated IR-absorbance values with diffusion equations applied for the one-dimensional case. Activation energies for H-extraction range between 200 and 300 $kJmol^{-1}$. Samples with higher ironcontent also show faster hydrogen-loss.

In the iron-poor specimen all OH-bands have approximately the same kinetics, while the other crystals show different behaviour for single OH bands. The two specimens of intermediate composition show a shift of intensity from a band centred at 3612 cm^{-1} to a band at 3599 cm^{-1} and two new minor bands appearing at ~3400 cm^{-1} during annealing. The near end-member andradite exhibits the fastest dehydration kinetics for the lower energy bands, with medium kinetics for the higher energy bands. A very stable sharp band at 3592 cm^{-1} remains at the end of the experiment.

These results underline the complexity of OH defects in garnets. It indicates that two or three different types of OH-defects are present in the structure. A multistep diffusion process may occur during dehydration, in which a large amount of hydrogen is released very quickly at the beginning of annealing, while residual hydrogen may partially occupy newly created defects which are more stable.

This study was supported by the EU through the Human Potential Program HPRN-CT-2000-0056.