

Hydrogen incorporation mechanisms in olivine

Water (hydrogen) is incorporated in most of the dominant phases (including olivine) of the mantle as point defects and in considerably large amount [Ferot and Bolfan-Casanova, 2012]. The addition of hydrogen to mantle mineral phases changes mantle rheology [Chopra and Paterson, 1984; Mackwell et al., 1985; Karato et al., 1986], enhances melting [Kushiro et al., 1968; Hirschmann, 2006], and is therefore an important process that contributes to the dynamics and chemical differentiation of the Earth. Based on point defect theory and IR spectroscopy studies, multiple species of hydrogen-related defects that occupy different lattice sites exist in olivine [Karato, 2013; 2015] and each species has distinctive physical properties (e.g. mobility, bonding energy) [Karato, 2013]. Therefore, a basic starting point in studying the effect of hydrogen on mantle dynamics is to understand the mechanism of hydrogen incorporation. Various physical and chemical parameters influence the tendency of formation of different hydrogen-related defect species [Bai and Kohlstedt, 1993]. This topic had been studied for olivine (the most abundant mineral phase in the upper mantle) by many different groups [Bai and Kohlstedt, 1993; Kohlstedt, 1998; Bell et al., 2003; Balan et al., 2008, 2011; Mosenfelder, 2006] starting from the early 1990s. However, there are still confusions regarding IR peak locations and specific hydrogen incorporation mechanisms.

Several models of hydrogen incorporation in olivine have been proposed. Some invoke the role of impurities such as Ti and Sc trace elements (extrinsic mechanism, c and d above), others assume pure olivine stoichiometry (intrinsic mechanism, a and b above). However, many of previous studies (e.g. Tollan et al., 2017) did not pay attention to the real world storage capability of trace elements so their results are not directly applicable to the real Earth condition. Our current hypothesis is, when hydrogen is incorporated into mantle olivine with some trace elements, defects will first preferentially form by the extrinsic mechanisms. However, when the water fugacity increases, trace elements will be used up and the dominant hydrogen incorporation mechanism then switches to intrinsic mechanisms. This switch between two regimes has not being tested by any study systematically and we would like to conduct experiments to investigate this possibility.

On the other hand, both experimental studies and first principle calculations have been used to investigate the corresponding IR peak position of each type of hydrogen-related defects [Bai and Kohlstedt, 1993; Umemoto et al., 2011; Balan et al., 2011; Xue et al., 2017; Tollan et al., 2017]. However, disagreements between different studies are large. Due to this disagreement on peak frequencies, we would like to systematically conduct experiments to investigate the chemical environment and physical environment to identify the IR peaks corresponding to each of the four incorporation mechanisms. Experiments were performed on San Carlos olivine single crystals under Mg(FeO)-rich and Opx-rich conditions and using three different types of oxygen fugacity buffers (Ni-NiO, Re-ReO₂, Mo-MoO₂) at 4 different pressures (3, 5, 7, 10 GPa) and 1473 K. In addition, we plan to do experiments using trace element doped polycrystalline samples to investigate the effect of trace element on hydrogen incorporation. This will provide the missing information of previous studies and hopefully contribute to a complete solution to the hydrogen incorporation mechanism research.

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