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Synthesis of hydrous aluminous perovskite and the substitution mechanism of aluminum

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Magnesium silicate perovskite is major constituent material in the Earth lower mantle. Recently, it was reported that a certain amount of H2O component is soluble in Al, and Fe rich magnesium silicate perovskite (Murakami et al., 2002; Litasov et al., 2002). Murakami et al., (2002) reported that 0.1-0.4 wt% water is present in Al, Fe perovskite synthesized using hydrous KLB-1 starting material. On the other hands, Litasov et al., (2002) reported that Al, Fe-perovskite synthesized using MORB contains 0.06 wt% water. This value is consistent with that reported in MgSiO3 perovskite (Higo et al., 2001). The discrepancy in the water contents in perovskites may be due to the differences of Al or Fe substitution mechanisms in Mg-perovskite synthesized using KLB-1 and MORB starting materials. We synthesized hydrous aluminous perovskite using the starting materials with various Al contents and examined the substitution mechanism of Al in MgSiO3 perovskite.

Six starting materials were prepared as mixtures of MgO, Mg(OH)2, Al2O3, SiO2. These starting materials are divided as 2 groups; one is MgSiO3-Al2O3-H2O system and the other is MgSiO3-MgAlO2.5-H2O system. Water contents of each starting materials were fixed at 10 wt%, except for one sample was 3 wt% H2O. We used the cell assembly of (Mg, Co)O, ZrO2 pressure medium and LaCrO3 heater. The fine powders of starting materials were sealed in an Au capsule. All experiments were conducted using multi-anvil press and performed at a pressure of about 27 GPa and temperatures of 1873 and 2073 K. The run durations of each temperatures were 8 and 5 hours, respectively. Recovered samples were examined by using EPMA, X-ray diffraction, and raman spectroscopy. Water contents were measured by the secondary ion mass spectrometry (SIMS).

The water content in perovskite increased drastically up to the about 4.5 mol% Al2O3 contents, where reached 0.3-0.4 wt% and became rather constant thereafter. EPMA analysis indicates that the chemical compositions of the recovered perovskites are independent of the starting material.

For discussing the substitution mechanism of aluminum, we calculated the theoretical water content of aluminous perovskite assuming all oxygen vacancies are substituted by hydroxyl, which was then compared with the actual water content in perovskite. The result shows that theoretical water content is consistent with the actual water content up to 4.5 mol% Al2O3, suggesting that H2O is in perovskite by the oxygen vacancy-hydroxyl substitution. For the Al2O3 contents larger than 4.5 mol%, howevere, theoretical water contents mismatch the actual values, suggesting that the operation of a tschermakite substitution as yielding the result that water content doesn't increase more than 4.5 mol% Al2O3.

Frost & Langenhorst (2002) reported the effect of Al2O3 on Fe-Mg partitioning between (Mg, Fe)O and magnesium silicate perovskite, and reported a non-linear relation. They proposed that the substitution of Fe3+ into the six-fold coordination site in perovskite is predominant for low Al content, whereas the substitution of a FeAlO3 component occurs at higher Al content, where Fe3+ enters into the eight-fold site, charge balanced by Al in the six-fold coordinated site. This discussion is consistent with our observation that the water contents in perovskite saturated at about 4.5 mol% Al2O3 content. The Al, Fe bearing perovskites synthesized by Murakami et al., (2002) are suggested to have a certain amount of oxygen vacancies because of the low Al content perovskite and thereafter H2O is soluble to some extent. The perovskites synthesized by Litasov et al., (2002), however, have significantly high Al contents because of use of MORB as starting material. This leads that the solution of the FeAlO3 component in perovskite prevents the formation of oxygen vacancies, and accordingly the H2O component cannot be soluble in such perovskite.