Poster Presentation

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Proton conduction mechanism and phase transition of (Rb,K)3H(SeO4)2

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The materials represented as M3H(XO4)2 (M = alkaline metal, X = S or Se) are known to exhibit high protonic conductivities at moderately high temperature. The high protonic conductivity emerges upon a structural phase transition and hydrogen bonds become directionally disordered. The protonic conduction is presumably realized through the disordered hydrogen bonds, but no experimental evidence has been reported. Meanwhile, although the mechanism of the protonic conduction is considered to be the same among this group of materials, the transition temperature (Tc) varies depending on the elements of M and X. For example, the material with M = Rb and X = Se undergoes the transition at 440 K while with M = K and X = Se the transition occurs at 390 K. Since the chemical characteristics of Rb and K are, as a principal, the same, some structural features may play crucial roles in triggering the phase transition. In order to clarify the mechanism of the proton conduction in the superprotonic phase and the relation between the crystal structure and Tc, structural studies on Rb3H(SeO4)2 at high temperature and solid solutions of Rb3H(SeO4)2 and K3H(SeO4)2 (Rb3-xKxH(SeO4)2, x=0,1,2,3) were conducted by means of single crystal neutron diffraction at FONDER at JRR-3M and SENJU at J-PARC/MLF. The proton density distribution map obtained from the high temperature neutron diffraction experiment clearly demonstrates 2-dimensional continuous spread of the proton distribution, which is considered to be the proton conduction path (figure). The structure analyses of Rb3-xKxH(SeO4)2 revealed that K ions tend to occupy one of two possible sites. As the concentration of K ion increases, the distortion of SeO4 appears to be enhanced. The variation of the distortion is consistent with the variation of the transition temperature, suggesting the close relationship between the distortion and the phase transition temperature.

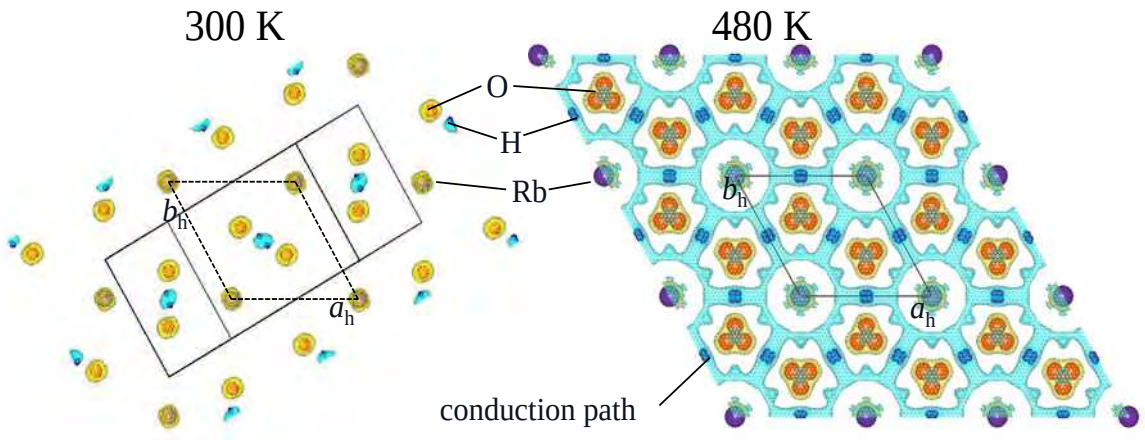

Figure Nuclear density distribution of $Rb_3H(SeO_4)_2$ at 300 K (left) and 480 K (right). The blue distribution in the right figure represents the proton density distribution.

Keywords: Proton conductor, Phase transition