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Proton conductivity of $Sr_2P_2O_7$ – based ammonium polyphosphate composite electrolytes for intermediate temperature fuel cells

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Fuel cells have attracted worldwide attention since they produce clean energy with high efficiency. Therefore, fuel cells are expected for practical use as power sources in vehicles, portable electronic devices, etc.

Recently, fuel cells operating at around room temperature (PEMFCs) and high temperatures, exceeding 873 K (SOFCs), have been extensively studied. However, fuel cells operating at intermediate temperatures ranging 523~873 K have been hardly studied. Intermediate temperature fuel cells will give many advantages: (1) high energy efficiency by using waste heat, so called energy cogeneration, (2) suppression of CO poisoning for electrode, (3) reduction of manufacturing cost by using no ceramic materials, etc. Despite of having these advantages, intermediate temperature fuel cells have not reported so far because of few electrolytes having sufficient proton conductivity.

In this work, we report the preparation and proton conductivities of $Sr_2P_2O_7$ – based ammonium polyphosphate composite electrolytes for intermediate temperature fuel cells.

$x NH_4PO_3$ / $Sr_2P_2O_7$ composites (x =2.0~4.0) were prepared by using NH_4PO_3 and $Sr_2P_2O_7$. NH_4PO_3 was prepared by reaction of 85%H_3PO_4 and P_2O_5, $(NH_2)_2CO$, followed by sintering at 573 K for 20 h under NH_3 gas flow [1]. $Sr_2P_2O_7$ was prepared by dehydrating $SrHPO_4$ at 923 K under Ar atmosphere. From X-ray diffraction patterns, the obtained NH_4PO_3 and $Sr_2P_2O_7$ corresponded to NH_4PO_3 Form. I and α-$Sr_2P_2O_7$, respectively [2, 3]. $x NH_4PO_3$ / $Sr_2P_2O_7$ composites (x =2.0~4.0) were prepared by milling NH_4PO_3 and $Sr_2P_2O_7$ in the molar ratio required to form $x NH_4PO_3$ / $Sr_2P_2O_7$ composites (x =2.0~4.0) and pressing into pellets, followed by sintering at 673 K for 10 h under NH_3 gas flow. X-ray diffraction patterns of the obtained composites exhibited no chemical reaction of NH_4PO_3 and $Sr_2P_2O_7$.

The proton conductivities of $x NH_4PO_3$ / $Sr_2P_2O_7$ composites (x =2.0~4.0) were measured under dry Ar and Ar humidified at 293 K atmospheres by an impedance analyzer using Pt / C electrodes. The measured frequencies and temperatures were in the range of 0.1 Hz to 1 MHz and 423 K to 573 K, respectively.

Arrhenius plots of proton conductivities for $x NH_4PO_3$ / $Sr_2P_2O_7$ composites (x =2.0~4.0) under dry Ar and Ar humidified at 293 K atmospheres during heating-cooling cycles are shown in Fig. 1(a) and (b), respectively. Proton conductivities of these composites drastically increased with increasing x, regardless of the atmosphere. This result exhibits that NH_4PO_3 mainly contributes to proton conduction and that $Sr_2P_2O_7$ serves as matrix. Moreover, it is found that proton conductivities of these composites containing lower x drastically increased at lower temperatures under Ar humidified at 293 K atmosphere, indicating that water molecules adsorbed on Sr should enhance the proton conduction. It is clear from Fig.1 that 4.0 NH_4PO_3 / $Sr_2P_2O_7$ composites under dry Ar and Ar humidified at 293 K atmospheres exhibited high proton conductivities of 58 mS / cm and 82 mS / cm, respectively at 523 K, which are sufficient high for intermediate temperature fuel cells.

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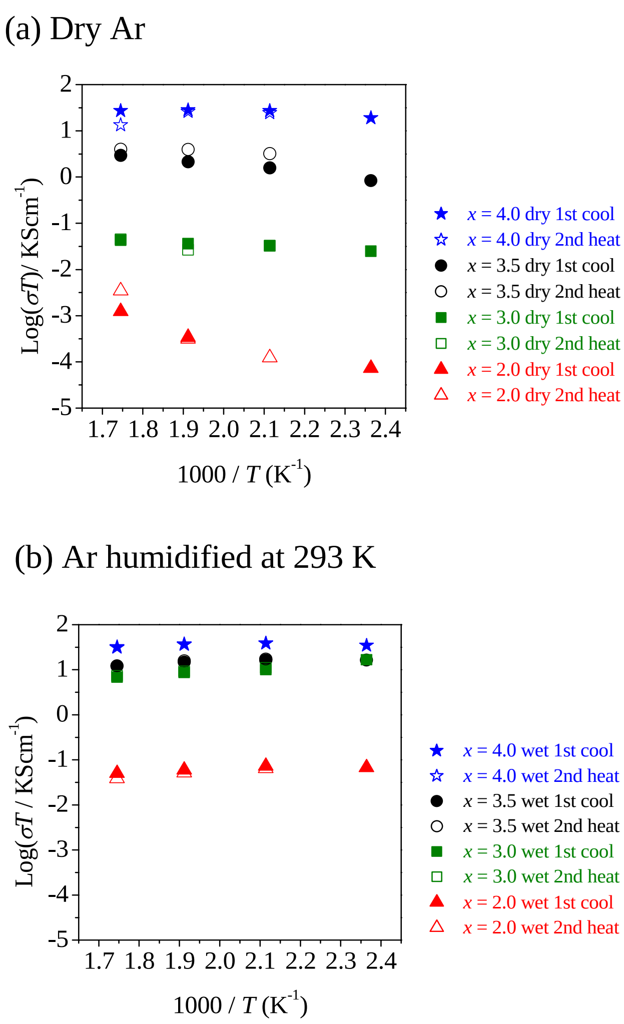

Fig. 1. Proton conductivities of $x NH_4PO_3$ / $Sr_2P_2O_7$ composites (x =2.0~4.0) under (a) dry Ar and (b) Ar humidified at 293 K atmospheres at 423~573 K.