Microstructure and Conductivity of Nano-Composite Proton Conducting Membrane for Electrode-Supported Intermediate Temperature Fuel Cells

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1. Introduction

The synthetic process and proton conductivity of a nano-composite proton conducting thin film electrolyte that consists of ionic salt and mesoporous SiO_2 thin film were investigated, taking into consideration applications to intermediate temperature fuel cells operated at 150 °C ~ 300 °C. We have studied the proton conductivities of powder composites such as CsH_2PO_4/SiO_2 and $CsHSO_4/SiO_2$ in the previous works [1,2]. In this report, we focus on structural and electrical properties of $CsHSO_4/SiO_2$ thin film electrolyte. Also, the synthetic process of the microstructure of mesoporous silica film (i.e. pore size and orientation of mesopores) was studied by means of an RF magnetron spattering method.

2. Experimental

A mesoporous SiO_2 thin film was prepared by the RF magnetron spattering method. Using a powdered $FeO\text{-}SiO_2$ mixture-target, Fe-Si-O amorphous thin films (thickness: about 5 μm) were synthesized on silica, carbon and n-type Si substrates, respectively. Then, Fe components, which acted as a temperate in the mesoporous silica thin film, were removed by etching in a HCl aqueous solution (temperate method). The microstructure of the resultant silica thin film was observed by FE-SEM. $CsHSO_4/SiO_2$ thin-film composite electrolyte was prepared by the impregnation of a $CsHSO_4$ aqueous solution into the mesopores of silica thin film. The conductivity measurements were carried out in dry Ar atmosphere using a Hewlett Packard 4192A impedance analyzer in the frequency range of 10Hz-10MHz at the temperatures between 60 ℃ and 180 ℃.

3. Result and Discussion

The cross sectional SEM image of mesoporous SiO_2 thin film synthesized on a dense silica substrate is shown in Fig. 1. The SEM image indicates that the diameter of mesopores of the silica thin film is around 10 nm, and the mesopores are approximately oriented one-dimensionally toward the surface of the film, although many branch connections and meandering of the mesopores are observed. Similar microstructure was also observed in the case of the silica thin film synthesized on a dense carbon substrate, suggesting that the present temperate method can be applied to electrode materials of fuel cells.

The conductivity measurement of $CsHSO_4/SiO_2$ thin film composite electrolyte synthesized on an n-type Si electrode was conducted. The result showed that the conductivity of $CsHSO_4/SiO_2$ composite was higher than that of pure $CsHSO_4$ in the low temperature region between 60 °C and 140 °C. In the high temperature region (140 ℃~180 ℃), however, the conductivity of the composite was lower than that of pure $CsHSO_4$. The conductivity of the composite was 3×10^{-4} S/cm at 180 °C, which was lower than the conductivity of pure $CsHSO_4$ (1×10^{-2} S/cm at 180 °C). The disconnection of proton conduction network in the thin film composite electrolyte seems to be one of the reasons for the low conductivity, which can be improved by controlling the microstructure of mesopores in the silica thin film.

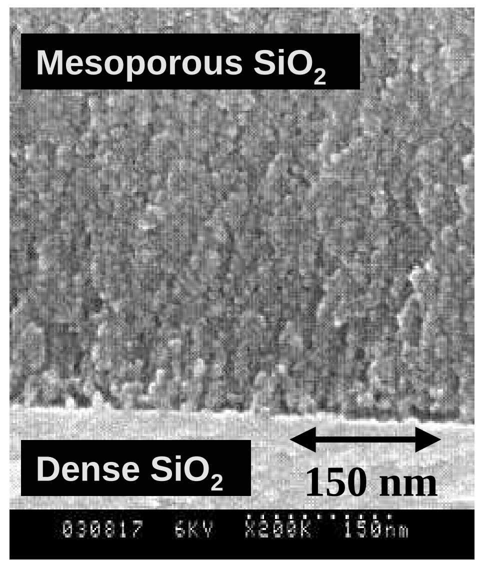

Fig. 1. The cross sectional SEM image of mesoporous SiO_2 thin film synthesized on a dense silica glass substrate.

4. Conclusion

The mesoporous silica thin film having mesopores oriented perpendicularly to the surface of the thin film was synthesized. When a proton conducting material ($CsHSO_4$) was impregnated into the mesopores of the silica thin film, the proton conductivity of the composite was increased, especially, in the low temperature region (80 ℃~140 ℃) in comparison with the conductivity of pure $CsHSO_4$. In the high temperature region (140 ℃~180 ℃), however, the conductivity of the composite was lower than that of pure $CsHSO_4$, which might be caused by disconnection of proton conduction network in the thin film composite electrolyte.

Acknowledgement

The authors acknowledge the financial support by Research and Development of Polymer Electrolyte Fuel Cell from New Energy and Industrial Technology Development Organization (NEDO) of Japan.

References

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