Electrochemical Properties and Durability of Electrocatalysts Supported on SnO₂ T. Tsukatsune¹, Y. Takabatake¹, Z. Noda², A. Hayashi^{2,3,4} and K. Sasaki^{1,2,3,4} Kyushu University ¹Department of Hydrogen Energy System, ²International Research Center for Hydrogen Energy, ³WPI-I2CNER, ⁴NEXT-FC Motooka 744 Nishi-ku, Fukuoka 819-0395, Japan

Introduction

For polymer electrolyte fuel cells, Pt-based electrocatalysts supported on carbon black are widely used. However, on the long-term use under the instantaneous high potential at the cathode, carbon corrosion followed by agglomeration and detachment of Pt catalyst particles from carbon can occur, resulting in the decrease in electrochemical surface area (ECSA) and oxygen reduction reaction (ORR) activities.

Therefore, we have developed SnO_2 as an alternative support, which is an oxide semiconductor with high electronic conductivity and thermochemically stable under the cathode conditions ^[1].

We have already shown that SnO_2 electrocatalysts have high durability, even though their initial activities were still lower. Then, the aims of this study are clarification of factors to decrease ORR activities and optimization of electrocatalysts preparation processes.

Experimental

Co-precipitation procedure was applied for preparation of SnO_2 and $Sn_{0.98}Nb_{0.02}O_2$, and then H_2PtCl_6 or $Pt(acac)_2$ were used as starting materials to impregnate Pt particles.

To evaluate electrochemical activities of these electrocatalysts, half-cell measurements were applied. ECSA was determined from hydrogen desorption region obtained in the cyclic voltammetry (CV) measurements. ORR activities were characterized by kinetic current obtained in rotating disk electrode (RDE) measurements.

Durability of the electrocatalysts was evaluated by the change in ORR activities and ECSAs. The electrocatalysts were subjected to the potential cycling between 1.0 V_{RHE} and 1.5 V_{RHE} according to Fuel Cell Commercialization Conference of Japan (FCCJ)^[2].

The concentrations of impurities in electrocatalysts were examined to clarify the impact of difference in preparation conditions (Pt precursors and heat treatment temperature) on ORR activities. The mixture of ultrapure water and electrocatalysts powder was boiled at 100°C to dissolve the impurities into water, and the filtrate was used for ICP analysis.

Results and Discussion

Fig. 1 and Fig. 2 show FESEM images of Pt/SnO_2 prepared by using $Pt(acac)_2$ and H_2PtCl_6 , respectively. These images clearly confirm homogeneous dispersion of Pt particles of ca. 2-3 nm.

 Pt/SnO_2 and $Pt/Sn_{0.98}Nb_{0.02}O_2$ showed high durability: they could keep up their kinetic currents compared to that of Pt/Vulcan. In addition, they showed almost no change in specific activities up to 60000 cycles as shown in Fig. 3, while that of Pt/Vulcan continuously decreased. This indicates that the decrease in ORR activities of Pt/SnO_2 and $Pt/Sn_{0.98}Nb_{0.02}O_2$ is mainly caused by the loss of ECSAs, although there are probably other factors to decrease ORR activities attributed to the corrosion of support materials (carbon) in the case of Pt/Vulcan. These results suggest that SnO_2 support can be a candidate to solve the problem on carbon corrosion at the cathode.

As mentioned above, initial ORR activities of SnO_2 electrocatalysts were still lower at this time. Therefore, to decrease impurities in electrocatalysts and to optimize their ORR activities, we have tried to improve catalysts preparation methods and have successfully increased ORR activities so far. We will report details of our results in the presentation.



Fig. 1 A FESEM image of Pt/SnO₂ (made by Pt(acac)₂)



Fig. 2 A FESEM image of Pt/SnO₂ (made by H₂PtCl₆)



Fig. 3 Change in specific activities during the potential cycles (1.0 - 1.5 $V_{\text{RHE}})$

References

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