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Activity and Durability of Pt/tin oxide/Ketjen Black Catalyst as PEFC Cathode

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Polymer electrolyte fuel cells (PEFCs) are promised as the power sources for household use and vehicles. For the commercialization, development of the cathode catalyst with low Pt loading is required.

It is recognized that Pt/C catalyst is deteriorated at the cathode side [1]. And, loss of the carbon support by the oxidation is serious under the vehicle environment. Considering the degradation mechanisms, we reported that surface modification of carbon support with high durable material as tin dioxide is promising technique [2, 3].

In this study, tin oxide nanoparticles / Ketjen black support (x-SnO_2/KB) was prepared with various loading amounts of tin dioxide and the heat treatment temperatures, and then the ORR activity and durability were investigated after Pt loading.

x-SnO_2/KB as the catalyst support were prepared by our original recipe. Here, x is loading amounts of Tin dioxide against KB. In brief, Ketjen Black EC-300j (abbr. as KB, Ketjen Black International Co. Ltd) was dispersed in tin fluoride complex solution for 24 h. Then, it was dispersed in boric acid aqueous solution. Finally, heat treatment was carried out at 500^{o}C under Ar atmosphere.

The heat treatment temperature was changed to 600 and 700^{o}C and these are named as SnO_2/KB-y (y is the heat treatment temperature of SnO_2/KB).

Pt particles were loaded on x-SnO2/KB, SnO_2/KB-y and KB by the impregnation method [2, 3]. The prepared catalysts were characterized by TG, TEM, ICP, and CO pulse test and XPS. These are denoted as Pt/x-SnO2/KB and Pt/KB, respectively.

Pt/x-SnO2/KB Pt/SnO_2/KB-y and Pt/KB, Nafion® solution (Aldrich) and *i*-propanol was mixed ultrasonically to prepare the catalyst ink. The ink was dropped on a glassy carbon disk and was used for the working electrode to investigate the catalytic activity for oxygen reduction reaction (ORR) and the durability. A glass made electrochemical cell filled with 0.1 mol dm^{-3} $HClO_4$ aqueous solution was used. ORR activity was investigated under oxygen atmosphere by a rotating disk electrode technique.

The potential cycle durability tests were carried out between 1.0 - 1.5 V in 0.1 mol dm^{-3} $HClO_4$ aqueous solution at 60^{o}C to investigate the durability of the electrode support. To estimate the degradation of KB, the electrochemical capacitance of KB was measured by cyclic voltammetry (CV) between 0.4 – 0.7 V. Moreover, the potential pulse durability tests were carried out between 1.0 V and 0.6 V to investigate Pt durability. Electrochemical active surface area (ECA) of Pt catalysts was estimated after an arbitrary period by CV during the Pt durability test.

Chemical stability of Pt catalysts on x-SnO_2/KB and Pt/KB were investigated by ICP following immersion in 1.0 dm^{-3} $HClO_4$ aqueous solutions at 60^{o}C for 72 h.

Figure 1 shows TEM image of Pt/58-SnO_2/KB. SnO_2 nanoparticles of *ca*. 8 nm were loaded on KB surface. And, the size of Pt nanoparticles was about 2 nm for every catalysts prepared.

Specific and mass activity for ORR of Pt/x-SnO_2/ KB and Pt/KB are summarized in Table 1. The ORR activity of Pt/58-SnO_2/ KB is higher than that of Pt/KB, even though the Pt particle size is same. This suggests that ORR activity of Pt could be increased by the presence of SnO_2 nanoparticles.

Table 2 shows the durability test results. The ECA retention is increased with increasing in the loading amount of tin oxide. Based on these results, modification of KB with SnO_2 nanoparticles is effective for improvement the durability of Pt catalysts.

Effect of the heat treatment temperature on the activity as well as the durability will be also reported.

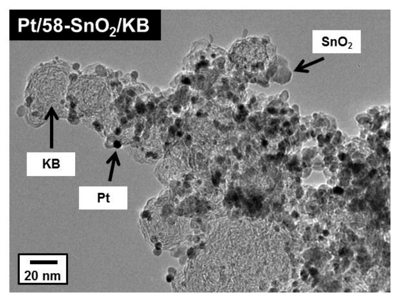

Fig. 1 Typical TEM images of Pt/58-SnO_2/KB.

Table 1 ORR activity of Pt/x-SnO_2/KB and Pt/KB.

	SA / μA cm^{-2}	MA / A g^{-1}
Pt/66-SnO_2/KB	421	325
Pt/58-SnO_2/KB	456	381
Pt/35-SnO_2/KB	322	295
Pt/17-SnO_2/KB	235	210
Pt/KB	285	281

Table 2 ECA of Pt during the durability tests before and after.

	ECA(Before) / m^2 g^{-1}	ECA(After) / m^2 g^{-1}
Pt/66-SnO_2/KB	75.3	57.2
Pt/58-SnO_2/KB	78.9	63.4
Pt/35-SnO_2/KB	72.5	45.1
Pt/17-SnO_2/KB	73.8	43.1
Pt/KB	75.2	38.9

References

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