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Controlled Synthesis of Pt_3Sn-SnOx/C Electrocatalysts for Polymer Electrolyte Membrane Fuel Cells

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Abstract : One of the greatest challenges of the implementation of polymer electrolyte membrane fuel cells (PEMFCs) is to find active and durable electrocatalysts. The cell performance is always limited by the oxygen reduction reaction (ORR) on the cathode since it is at least 6 orders of magnitude slower than the hydrogen oxidation on the anode. Therefore high loading of Pt is required. Catalyst corrosion is also more significant on the cathode, especially in case of mobile applications, where rapid changes of loading have to be tolerated. Pt-Sn bulk alloys and SnO2-decorated Pt3Sn nanostructures are among the most studied bimetallic systems for fuel cell applications. Exclusive formation of supported Sn-Pt alloy phases with different Pt/Sn ratios can be achieved by using controlled surface reactions (CSRs) between hydrogen adsorbed on Pt sites and tetraethyl tin. In this contribution our results for commercial and a home-made 20 wt.% Pt/C catalysts modified by tin anchoring via CSRs are presented. The parent Pt/C catalysts were synthesized by modified NaBH4-assisted ethylene-glycol reduction method using ethanol as a solvent, which resulted either in dispersed and highly stable Pt nanoparticles or evenly distributed raspberry-like agglomerates according to the chosen synthesis parameters. The 20 wt.% Pt/C catalysts prepared that way showed improved electrocatalytic performance in the ORR and stability in comparison to the commercial 20 wt.% Pt/C catalysts. Then, in order to obtain Sn-Pt/C catalysts with Pt/Sn= 3 ratio, the Pt/C catalysts were modified with tetraethyl tin (SnEt4) using three and five consecutive tin anchoring periods. According to in situ XPS studies in case of catalysts with highly dispersed Pt nanoparticles, pre-treatment in hydrogen even at 170°C resulted in complete reduction of the ionic tin to Sn0. No evidence of the presence of SnO2 phase was found by means of the XRD and EDS analysis. These results demonstrate that the method of CSRs is a powerful tool to create Pt-Sn bimetallic nanoparticles exclusively, without tin deposition onto the carbon support. On the contrary, the XPS results revealed that the tin-modified catalysts with raspberry-like Pt agglomerates always contained a fraction of non-reducible tin oxide. At the same time, they showed increased activity and long-term stability in the ORR than Pt/C, which was assigned to the presence of SnO2 in close proximity/contact with Pt-Sn alloy phase. It has been demonstrated that the content and dispersion of the fcc Pt3Sn phase within the electrocatalysts can be controlled by tuning the reaction conditions of CSRs. The bimetallic catalysts displayed an outstanding performance in the ORR. The preparation of a highly dispersed 20Pt/C catalyst permits to decrease the Pt content without relevant decline in the electrocatalytic performance of the catalysts.

Keywords : anode catalyst, cathode catalyst, controlled surface reactions, oxygen reduction reaction, PtSn/C electrocatalyst
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