

Effect of Core Composition of Pd alloy on the Single Cell Performance in high temperature PEMFC

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High temperature polymer electrolyte membrane fuel cell (HT-PEMFC), which used the reformed gas from natural gas, has been attracted as a combined heat and power system, for the stationary application due to the high tolerance to CO and no humidification control [1]. One of the drawback of HT-PEMFC is a use of large amount of Pt in the electrode because of the poisoning by phosphoric acid [1,2]. In this regards, the development of non-Pt catalyst having similar activity to the Pt catalyst based on the economic precious elements for the HT-PEMFC is essential.

In this presentation, the core-shell structures of Pd-based alloy catalysts, such as PdNi, PdCu and PdNiCu cores, respectively, with PdIr shell, were prepared *via* modified polyol process and their galvanic displacement reactions were investigated.

The carbon supported Pd-alloy core nanoparticles with different compositions were prepared by a modified polyol method [4] in the microwave reactor at 250 °C with Pd(NH₃)₄Cl₂·H₂O, NiCl₂·6H₂O and CuCl₂·2H₂O. Three compositions of core nanoparticle are PdNi, PdCu and PdNiCu, and pre-formed materials are named as PdNi/C, PdCu/C and PdNiCu/C, respectively. For the formation of PdIr shell, the as-prepared PdNiCu/C was dispersed in the deionized water to conduct the galvanic displacement using K₂PdCl₄, H₂IrCl₄ and sodium citrate. For comparison purpose, PdNi@PdIr/C and PdCu@PdIr/C were prepared by the same procedures. The total metal contents of three catalysts are ca. 50wt%.

The peak positions for (111) plane for Pd of fcc structure from the all three catalysts shifted to the higher 2θ value from the XRD (not shown), which could suggest that smaller Ni,

Cu, and Ir atom co-existed in the interstitial sites of Pd crystal lattice.

To estimate the effect of core composition on oxygen reduction reaction (ORR) activity, the linear scan voltammetry was conducted. The mass activity for ORR based on total mass of Pd and Ir of PdNi@PdIr/C, PdNiCu@PdIr/C and PdCu@PdIr/C is 3.3, 10.7 and 1.2 mA/mg, respectively, which indicates the coexistence of Ni and Cu with Pd in the core has great positive effect on the ORR activity. It might be attributed to the change of d-band center of Pd by electronic effect and/or compressive strain to the surface atoms, Pd and Ir [5]. As shown in Fig. 1, the PdNiCu@PdIr/C catalyst showed the superior performance in the below 400mA/cm². The MEA performance is aligned to the mass activity for ORR of the catalysts: PdNiCu@PdIr/C > PdNi@PdIr/C > PdCu@PdIr/C. The MEA data indicate that an improved activity in the core-shell catalyst is realized in the single cell level for the HT-PEMFC and suggest the core compositions of core-shell catalysts is very essential for activity control.

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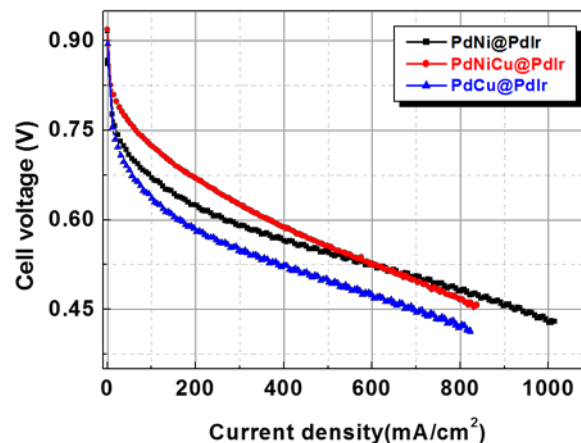


Fig.1 Single cell performance of PdNi@PdIr/C, PdNiCu@PdIr/C and PdCu@PdIr/C catalysts

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