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Oxygen Reduction Reaction Activity for Surface-strain-controlled Pt-M(111) Model Catalyst Surfaces Prepared in UHV

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Comprehensive understanding of oxygen reduction reaction (ORR) activity enhancement mechanisms for Pt-based alloy (Pt-M) catalysts is a key for developing highly-efficient cathode catalysts for polymer electrolyte fuel cell. To clarify the effects of the alloy surface atomic structures (e.g., atomic arrangements of surface Pt atoms, Pt/M atomic ratio etc.,) on ORR activity, well-defined model catalyst study should be required. We have investigated ORR properties for the well-defined Pt-based bimetallic single crystal surface alloys prepared in ultra-high vacuum (UHV) [1]. In this study, ORR activities are investigated for Pt/M (M=Co and Zr) model catalysts prepared on Pt(111) substrate through alternative arc-plasma depositions (APDs) of Pt and M nano-layers.

The UHV-APD-EC apparatus is described elsewhere [2]. Pt and M layers were alternately deposited onto a clean Pt(111) substrate by the APDs in UHV. Total thickness of the Pt/M and thickness of the topmost-surface Pt and bottom M layers are fixed to be 6 nm, 1.6nm, and 0.4nm, respectively, and the underlying M thickness are changed. Structural analysis for the prepared PtMxnm/Pt3.6nm/M0.4nm/Pt(111) (denoted hereafter as M_xnm) samples are performed by using in-plane XRD, cross-sectional STEM. For the ORR activity evaluation, the UHV-prepared samples were transferred to an N2-purged glove box without air exposure. Cyclic voltammetry (CV) and linear sweep voltammetry (LSV) were conducted in N2-purged and O2-saturated 0.1M HClO4 in the glove-box. To discuss EC stability, square-wave potential cycling between 0.6(3s) ˜ 1.0(3s) V vs. RHE was applied.

Surface strain estimated by in-plane XRD showed that compressive and tensile strain worked on Pt-Co(111) and Pt-Zr(111) bimetallic surfaces, respectively. ORR activity enhancement well corresponds to in-plane lattice distance of the topmost Pt(111) layers estimated by the XRD. For example, the activity enhancement factor for the Co_1.6nm having ca. 2 % compressive strain is highest (13-fold vs. Pt(111)) for the Pt-Co(111) series. The results suggest that the ORR enhancements closely correlate with surface strain of the topmost Pt(111) layers induced by underlying M layers.

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