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Improving the oxygen reduction reaction by strain effects: bulk and nanoparticles studies

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The sluggish kinetics of the oxygen reduction reaction (ORR) hinders the commercialization of proton exchange membrane fuel cells (PEMFC). The ORR activity could be enhanced by combining Pt with late transition 3d metals (i.e. Fe, Co, Ni, and Cu). However, such alloys are often not stable enough in the corrosive PEMFC environment and suffer from dealloying [1]. In contrast, Pt alloys containing lanthanide metals, such as Pt₅Gd, exhibits high ORR activity and stability [2]. In this work we present the structural and electrochemical characterization of mass-selected platinum-gadolinium alloy nanoparticles (Pt_xGd NPs) as potential oxygen reduction reaction (ORR) electrocatalysts. The alloy nanoparticles are synthesized for the first time using gas aggregation after sputtering of an alloy target under ultra-high vacuum (UHV) conditions [3]. The morphology of the Pt_xGd catalysts is characterized and their catalytic performance towards the ORR is assessed in acidic media using half-cell configuration. As result, the Pt_xGd 8 nm catalyst shows an outstanding performance (3,6 A (mg Pt)⁻¹), surpassing our preceding maximum activity reached with Pt_xY NPs catalysts and resulting in the highest activity reported for nanocatalysts based on an acid-leached "Pt-skeleton" structure. In addition, the optimum Pt_xGd catalyst also presents superior stability compared to Pt_xY, as suggested by the long-term stability tests under ORR potential cycling. Extended X-ray Absorption Fine Structure (EXAFS) spectroscopy measurements confirm that as-prepared Pt_xGd NPs are compressively strained relative to pure Pt and that a Pt_xGd core/Pt-rich shell structure is adopted after partial Gd leaching. We proposed that the ORR enhancement accounts for the compressive strain within the Pt shell induced by the alloyed core. Indeed, the estimated strain is used as valid descriptor of the measured ORR activity. The results herein presented confirm the suitability of Pt_xGd NPs as cathode nanocatalysts for proton exchange membrane fuel cells (PEMFCs). In addition, our investigation of sputter-clean polycrystalline Pt-lanthanide and Pt-alkaline earth alloys shows that the compressive strain also controls the activity and stability of bulk catalysts.

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