## Developed Nanomaterials with a Pd<sub>core</sub>-Fe-Pd<sub>skin</sub> Structure for Efficient Electrocatalytic Performance in Oxygen Reduction and Glycerol Oxidation Reactions in Alkaline Electrolytes

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## Abstract

One of the unsolved important fundamental and applied questions in heterogeneous electrocatalysis for low-temperature fuel cells and electrolyzers is about our ability to engineer a unique electrocatalyst that can simultaneously operate for both anode and cathode without changing the composition/structure or the synthesis method. Our present contribution is about a solution in the case of iron-diluted Pd nanoparticles. ORR is the common bottleneck of the pollution-free electricity production with alkaline technologies of rechargeable metal-air batteries and fuel cells. As alternative to scarcely Pt, Pd-based materials have attracted much attention for different applications in heterogeneous catalysis and not only for ORR, but also for glycerol (biomass-based byproduct) oxidation that is also common reaction (anode) for fuel cells and low-energy

input electrolyzers to produce green H<sub>2</sub>. So, being able to prepare a nanocatalyst that is simultaneously active and durable for both ORR and glycerol oxidation reaction (GOR) is highly desired. We observed that the support plays an important role and the presence of Fe (20%) dramatically improves the performance. The developed bimetallic PdFe performed efficiently and selectively ORR in alkaline media at the maximum faradaic yield of four-electron transfer (100%) and high kinetic current density  $j_k$  of 2 mA cm<sup>-2</sup><sub>Pd</sub> (1 A mg<sup>-1</sup><sub>Pd</sub>) at 0.9 V<sub>RHE</sub>, which largely exceeds the tested commercial Pd/C and the literature. For glycerol electrooxidation, the obtained peak current density of  $j_p = 2.3$  mA cm<sup>-2</sup><sub>Pd</sub> (1.1 A mg<sup>-1</sup><sub>Pd</sub>) and faradaic efficiency of FE = 83-99% outperformed the commercial Pd/C ( $j_p = 0.47$  mA cm<sup>-2</sup><sub>Pd</sub> (0.39 A mg<sup>-1</sup><sub>Pd</sub>, FE = 78%) and the majority of the reported catalysts.

Our conducted DFT calculations reveal that the high performance of the bimetallic PdFe arises from a unique structure with Pd in the core plus skin and Fe in the second layer. This particular configuration of skin surfaces has been reported for PtNi to trigger high electrocatalytic kinetics for ORR. Those findings open new possibilities in the development of multifunctional nanocatalysts for the sustainable electrochemical energy conversion and storage.

