

Controlling the Activity and Stability of Pt-Based Electrocatalysts By Means of the Lanthanide Contraction - DTU Orbit (08/11/2017)

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In order to reduce the Pt loading at the cathode of proton exchange membrane fuel cells (PEMFCs) more active and stable catalysts are needed to drive the oxygen reduction reaction. Most research has focussed on achieving this by alloying Pt with Fe, Co, Ni or Cu [1,2]. However, these compounds typically degrade under PEMFC conditions, due to dealloying. Alloys of Pt and lanthanides may be inherently less prone to dealloying under reactions conditions, due to their negative enthalpy of formation [2-4].

Herein we present a systematic study on the trends in activity of seven novel Pt-lanthanide electrodes (Pt₅La, Pt₅Ce, Pt₅Sm, Pt₅Gd, Pt₅Tb, Pt₅Dy and Pt₅Tm). The materials are highly active, presenting a 3 to 6-fold activity enhancement over Pt [3-5], amongst the most active polycrystalline Pt-based catalyst ever reported. Moreover, our recent study showed that Pt_xGd is highly active in the nanoparticulate form [6]. On the bulk alloys, a Pt overlayer with a thickness of few Pt layers is formed onto the bulk alloys by acid leaching (Fig. 1A) [3-5]. The ORR activity versus the lattice parameter obtained by X-ray diffraction measurements follows a volcano relationship (Fig. 1B). Furthermore, we explain the trends in stability, and present the lattice parameter as a new descriptor that controls both the activity and stability of these materials. Using the lanthanide contraction we demonstrate that the electrocatalytic performance can be engineered by tuning the Pt-Pt distance.

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Authors: Escribano, M. E. (Intern), Malacrida, P. (Intern), Vej-Hansen, U. G. (Intern), Tripkovic, V. (Intern), Velazquez-Palenzuela, A. A. (Intern), Schiøtz, J. (Intern), Rossmeisl, J. (Intern), Stephens, I. (Intern), Chorkendorff, I. (Intern)

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