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# Unveiling the local composition of Pd-Hg alloy nanoparticles by Scanning Transmission Electron Microscopy and Spectroscopy

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Hydrogen peroxide  $H_2O_2$  is an important chemical product extensively used in many industrial processes such as paper bleaching, textile industry, metallurgy and wastewater treatment [1]. Like other common chemicals, hydrogen peroxide is mainly produced in centralized large-scale facilities via a complex batch synthesis method, the anthraquinone process. Alternatively, hydrogen peroxide can be synthesized in an electrochemical device, such as a fuel cell or electrolyzer, via direct electroreduction of oxygen [2]. This synthesis method would allow more decentralized production at smaller scale close to the consumption points and therefore lead to a cost reduction. Furthermore, renewable energy power sources can be employed to run such electrochemical devices providing a system for energy storage.

Hg-based alloys have been identified by DFT calculations as new, highly active and stable catalysts for the electrochemical reduction of oxygen to  $H_2O_2$  and confirmed experimentally [3,4]. Pd-Hg nanoparticles have been discovered as electrocatalysts exhibiting the highest reported mass activity [4]. In order to understand the outstanding performance of this catalyst, knowledge of the formation of the bimetallic compound and the elemental distribution at the atomic level is important.

Here we present an advanced Electron Microscopy characterization of a new Pd-Hg alloy nanoparticle electrocatalyst for the electrochemical reduction of oxygen to  $H_2O_2$ .

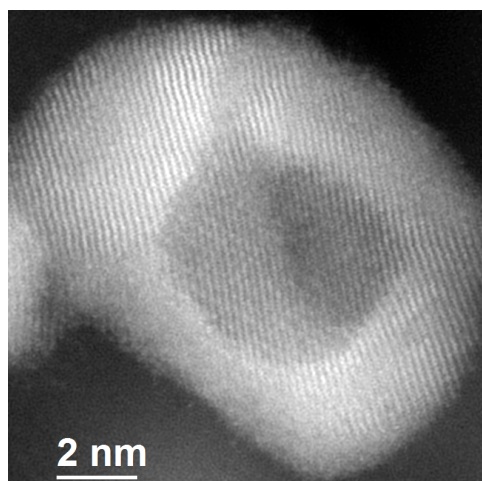
Pd-Hg nanoparticles have been synthesized by electrochemical deposition of Hg on carbon-supported Pd nanoparticles. The sample has been studied by means of high resolution Scanning Transmission Electron Microscopy (STEM) in a  $C_5$ -corrected FEI Titan 80-300. Furthermore, X-Ray Energy Dispersive Spectroscopy (EDX) spectrum imaging performed on a FEI Tecnai OSIRIS has been employed in order to map the distribution of the elements at the nanoparticle level.

Figure 1 shows a high resolution STEM micrograph of a Pd-Hg nanoparticle where a clear core-shell structure is visible. Since Hg is electrodeposited on Pd, the bright contrast shell is rich of the heavy metal Hg, while the core results composed of pure Pd. This is confirmed by the STEM-EDX spectrum image shown in Figure 2a-d. Both Pd and Hg are present in the outermost region of the particle as also clarified by the EDX linescan in Figure 2e. The Hg signal is peaked on the shell region of the particle where the alloy is formed, while it drops on the core region with an opposite trend with respect to the Pd signal.

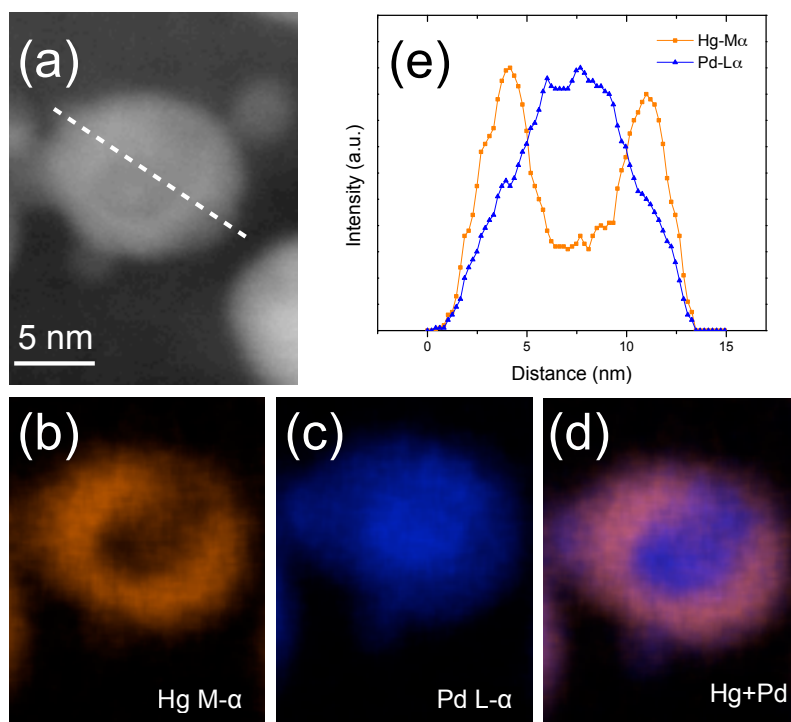
In this work, STEM-EDX mapping has been used to study in detail the elemental distribution of bimetallic Hg-Pd nanoparticles. A core-shell type of structure has been unveiled with the formation of an alloy on the shell region in agreement with the theoretical predictions, making STEM-EDX spectrum imaging a powerful technique to better understand the mechanisms for the enhanced activity of these catalysts [5].

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**Figure 1** High resolution HAADF-STEM micrograph of a Pd-Hg nanoparticle.



**Figure 2** (a) STEM micrograph of a nanoparticle and (b-d) corresponding Hg, Pd and combined X-Ray elemental maps. (e) Normalized EDS intensity line profiles along with the white dashed line drawn on (a).