Aberration-Corrected STEM Study on Pt_{0.8}Ni De-alloyed Nanocatalysts for Proton Exchange Membrane Fuel Cells

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Proton exchange membrane fuel cells (PEMFCs) are promising energy conversion devices for transport and stationary applications. Pt nanoparticles are currently used as the catalyst to promote the kinetics of the hydrogen oxidation and oxygen reduction reactions in the anode and cathode of the fuel cell, respectively. However, alloys of Pt with base metals are being investigated to replace Pt on the cathode as a way to improve the efficiency of the fuel cell, and reduce cost [1].

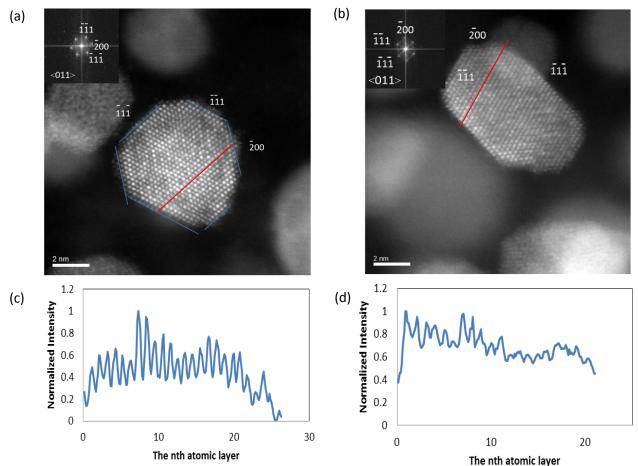
In this work Pt-Ni nanoparticles were de-alloyed by acid leaching to produce 5.8 nm size Pt_{.8}Ni catalyst nanoparticles. In order to better understand the relationship between the elemental distribution and the nanoparticle shape and structure, the nanoparticles were characterized by aberration-corrected scanning transmission electron microscopy (STEM), using high-angle annular dark-field (HAADF) imaging [2]. The Pt and Ni compositional distribution of the de-alloyed nanoparticles was investigated using EDS mapping in STEM mode. In order to better understand the three dimensional shape of the nanoparticles and the carbon support, 3-D electron tomography of the nanoparticles was performed in a JEOL JEM ARM 200F. A total of 61 STEM images were collected over a tilt range of -60 to +60 degrees, with a 2° tilting step. The final tilt series were aligned, reconstructed and visualized using Inspect 3D and Amira 4.1, respectively.

Figure 1 shows two aberration-corrected HAADF STEM images of the de-alloyed Pt_{0.8}Ni nanoparticles projected along the [110] beam direction. Although most of the particles exhibit a truncated octahedron shape with {111} and {200} facets (Fig. 1a), there are also some particles with long {111} facets (Fig. 1b). The absence of superlattice reflections in the FFTs (insets) shows that Pt and Ni are in solid solution, forming a face-centered cubic structure. However, as shown in Figures 1c and 1d, the nanoparticles exhibit regions of bright and dark contrast, which indicate that the composition of Pt and Ni is not uniform throughout the particle. This heterogeneous distribution is a result of the de-alloying process. In addition, nanoparticles exhibit Pt enriched surfaces, as shown by EDS mapping (Fig. 2). Finally, 3-D electron tomography confirms that most of the particles exhibit a truncated octahedron shape (Fig. 3).

References

- [1] S. C. Ball et al. ECS Transactions, 11(1) (2007), p.1267.
- [2] S. Chen, et al. J. Phy. Chem. C. 113(3) (2009), p.1109.

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The nth atomic layer

Figure 1. (a) and (b) Aberration corrected HAADF images of Pt_{0.8}Ni nanoparticles and corresponding FFTs (insets). (c) and (d) normalized intensities across the nanoparticle (along the

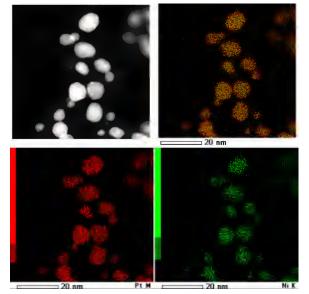


Figure 3. 3D reconstructed image of the Pt_{0.8}Ni nanoparticles

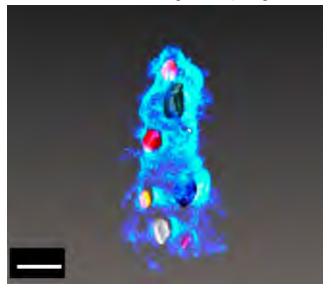


Figure 2. HAADF STEM image and EDS mapping of Pt_{0.8}Ni nanoparticles