On strain in platinum and palladium nanoparticles in an electrochemical environment

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Abstract

This doctoral thesis brings together the fields of electrocatalysis and Bragg Coherent Xray Diffraction Imaging (BCDI) to shed light on the behaviours of Pt and Pd nanoparticles which are particularly relevant to energy conversion and storage technologies. BCDI emerges as a powerful technique, offering non-destructive three-dimensional structural information on single nanoparticles, such as local strain maps.

One key aspect of this work is the use of strain analysis as a valuable tool to capture adsorption and absorption phenomena. The idea is to accurately map adsorption sites under electrochemical conditions, particularly under electrode potential control.

Pt is a widely used catalyst in various reactions, including the Hydrogen Evolution Reaction, Hydrogen Oxidation Reaction, and Oxygen Reduction Reaction. Although studying the relevant electrode potential regions for these reactions proved challenging, BCDI successfully captures strain heterogeneity in Pt nanoparticles within the Electrochemical Double Layer region.

These observations demonstrate the bisulphate ion adsorption occurs at the surface of a single nanoparticle and is responsible for the increase of strain heterogeneity as a function of the electrode potential. Molecular Statics (MS) and Density Functional Theory support experimental observations by bridging the gap between the scale allowed by BCDI and the atomic scale.

Pd is known to have a high affinity for hydrogen, which absorbs into its volume, leading to a phase transition and a significant increase in its lattice parameter. These absorption mechanisms are examined in detail by monitoring single Pd nanoparticles at different electrode potentials and during phase transitions due to hydrogenation. The various experimental observations supported by MS simulation provide a basis for proposing a model to elucidate the phase transition.

While primarily focusing on fundamental investigations of model systems, this research demonstrates the potential of strain analysis through BCDI as a descriptor of adsorption and absorption at the scale of a single nanoparticle.