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Computational Design of Novel Sustainable Catalysts for Fuel Cell Technologies

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Abstract:

Catalysts for fuel cell electrodes rely on Pt-group metals. The high price and low abundance of these active metals underpin an engineering effort for reducing the Pt load from ~mg/cm^2 down to μg/cm^2. In the context of oxide-supported metal catalysts, this implies identifying strategies to functionalize the supporting surfaces, so as to stabilize ultra-high dispersions of Pt atoms. Clear guidelines for maximizing the utilization of supported metals would help the general development of sustainable catalytic technologies for renewable energies and environmental applications with reduced precious metals contents.

In this talk, materials modeling is employed to guide the design of novel single-atom catalysts in which the metal catalyst is stabilized as single atoms. Ab-initio density functional theory calculations are combined with molecular dynamics, atomistic thermodynamics, metadynamics, and other enhanced-sampling methods to reveal specific adsorption sites on catalyst supports that provide the stabilization of single metal atoms under reaction conditions [1-3]. I will consider metal-oxide solid-solutions, nanostructured surfaces and supported nanoparticles. The calculations are used to characterize the chemical bonding and the reactivity of these active sites in a wide range of compositions and environments, ranging from model surfaces at T=0K in vacuum conditions, to realistic wet electrodes at finite temperatures.

The computational predictions are confirmed by the dedicated synthesis and characterization of model catalytic systems with applications in fuel cell technologies [4]. The combination of multi scale modelling, scanning tunnelling microscopy and photoemission spectroscopy demonstrates that Pt single atoms on cerium oxide supports are stabilized by the most ubiquitous defects on solid surfaces – monoatomic step edges [5]. Pt segregation at steps leads to stable dispersions of single Pt^{2+} ions in planar PtO_4 moieties that incorporate excess O atoms and contribute to oxygen storage capacity of the support. The reactivity of these ionic sites is estimated and compared to the one of metallic supported clusters, both in vacuum and in solution [3,6]. We identify surface step engineering and step decoration as effective strategies for maximizing the dispersion and activity of platinum-group metals.

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