Learning the nanocatalyst structure “on the fly” using neural networks

Janis Timoshenko, Nicholas Marcella, Yang Liu (*Department of Material Science and Chemical Engineering, Stony Brook University, Stony Brook, NY 11794, USA*), Yuewei Lin (*Computational Science Center, Brookhaven National Laboratory, Upton, NY 11973, USA*), Deyu Lu (*Center for Functional Nanomaterials, Brookhaven National Laboratory, Upton, NY 11973, USA*) and Anatoly I. Frenkel (*Department of Material Science and Chemical Engineering, Stony Brook University, Stony Brook, NY 11794, USA; Division of Chemistry, Brookhaven National Laboratory, Upton, NY 11973, USA*)
janis.timoshenko@gmail.com

The unique catalytic properties of metallic nanoparticles (NP) are a result of interplay between NPs core sites and undercoordinated surface sites, the quantum confinement effects that modify the electronic properties of metal atoms, and the interactions between metal sites and support and/or adsorbates. Understanding of NPs atomic structure and its relation to the NPs properties is thus important for design of novel catalytic materials. *In-situ* studies are an essential element in such investigations, since the atomic structure of nanosized catalysts can change dramatically in reaction conditions. X-ray absorption spectroscopy is one of a few methods that are useful in this case, due to its sensitivity to the chemical state of absorbing atoms and to the types and arrangements of atoms around the absorber, and its suitability for *in-situ* and *in-operando* studies.

While EXAFS spectroscopy is widely used in NPs structure studies, much less attention has been paid to the information encoded in X-ray absorption near edge structure (XANES). Analysis of XANES data has several advantages. First, XANES is less sensitive to disorder, which affects severely EXAFS quality and complicates EXAFS data interpretation. Secondly, XANES is more sensitive to the 3D geometry of the environment around absorbing atoms. Better signal-to-noise ratio in XANES region of absorption spectra also means that spectra can be collected with better time-resolution, for more diluted samples, on strongly attenuating support materials and/or in complex experimental setups. The main challenge that hinders the usage of XANES for quantitative analysis is the lack of methodology that would allow one to extract structural information from experimental data.

Recent developments in data-enabled discovery methods provide a key to this problem. To correlate XANES features with the descriptors of 3D local structure of metallic NPs, we employed machine learning and ab-initio XANES calculations. Here we demonstrate the potentiality of this method on the example of XANES study of monometallic (Pt, Ag and Cu), as well as bimetallic (PdAu) particles. We use theoretical site-specific XANES spectra, calculated by ab-initio codes for a broad range of structure models, to train an artificial neural network (NN). The trained NN is then used to extract structural information from the experimental XANES data.

By relying on NN method, we obtain from XANES data unique information on the size and shape of investigated metallic NPs, effective interatomic distances and alloying motifs within bimetallic systems. We expect that the proposed approach will be especially beneficial to follow *in situ* the changes in the environments of metal atoms in a wide range of catalytically relevant systems.

We acknowledge support of the U.S. Department of Energy grant No. DE-FG02-03ER15476.