Deciphering structure of metallic nanoparticles using reverse Monte Carlo method, molecular dynamics and machine learning

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Understanding the relation between structure and properties in nanostructured materials remains a challenge. The choice of experimental techniques that provide atomic-level information in non-periodic materials is still extremely limited, and a large variety of unique structural motifs that can be expected in nanosized systems complicates significantly the interpretation of experimental data. Among the few techniques that are suitable for structural characterization of nanomaterials, invaluable is X-ray absorption spectroscopy. In particular, extended X-ray absorption fine structure (EXAFS) contains unique information about the atomic arrangements around the absorbing metal atoms. The accuracy of conventional approaches for EXAFS data analysis is, however, limited, when they are applied to such intrinsically heterogeneous, disordered materials as metallic nanoparticles (NPs). Information about the strong asymmetry of bond-length distributions, distant coordination shells and many-atom distribution functions can contain the key answers regarding the 3D structure and structure-properties relationship in NPs, but cannot be accounted for adequately in conventional EXAFS fitting. To approach this problem, we employ advanced techniques, based on modeling of NPs 3D geometry and ab-initio EXAFS simulations. We use reverse Monte Carlo (RMC) method to reconstruct particle 3D structure and asymmetric bond length distributions.[1] In the common cases, when the experimental EXAFS data alone do not contain sufficient information for RMC procedure to reconstruct NPs structure unambiguously, we employ molecular dynamics (MD) simulations.[2] Finally, we employ machine learning methods[3] to establish the relationship between structural motifs in NPs and corresponding subtle changes in EXAFS spectra: an artificial neural network is trained on theoretical EXAFS spectra, obtained in MD simulations, and can then be applied for interpretation of experimental EXAFS in mono- and bimetallic NPs. Our approach is as quick and flexible as conventional EXAFS data fitting (and thus suitable for high-throughput studies), accounts for complex shapes of bond-length distributions and contributions of distant as RMC, and yields physically reasonable structure models as MD approach.

References:

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