Exploiting Automatic Image Processing and In-situ Transmission Electron Microscopy to Understand the Stability of Supported Nanoparticles

James P. Horwath¹, Leena Vyas¹, Dmitri N. Zakharov, Rémi Méchet, Peter W. Voorhees⁴, and Eric A. Stach¹,⁵*

¹ Department of Materials Science and Engineering, University of Pennsylvania, Philadelphia, PA USA.
² Center for Functional Nanomaterials, Brookhaven National Laboratory, Upton, NY USA
³ Department of Computer Science, University of Puerto Rico, Río Piedras, San Juan, PR, USA
⁴ Department of Materials Science and Engineering, Northwestern University, Evanston, IL, USA.
⁵ Laboratory for Research on the Structure of Matter, University of Pennsylvania, Philadelphia, PA USA.
* Corresponding author: stach@seas.upenn.edu

The activity and lifetime of heterogeneous catalysts are intimately linked with their structural stability in reactive environments. However, it can be challenging to understand and predict how reactive environments lead to nanoparticle coarsening via center of mass motion and Ostwald ripening and how evaporation can lead to mass loss.

In this work, we develop and exploit advanced data analysis tools to track the temporal evolution of nanoparticles as a function of time, temperature, and reactive environment using transmission electron microscopy. The first portion of the talk will describe our development of a fast and highly accurate image segmentation approach based on deep learning. We describe how a systematic investigation of dataset preparation, neural network architecture, and accuracy evaluation lead to a tool for determining the size and shape of nanoparticles in high pixel resolution TEM images. [1]

In the second half of the talk, we will show how we exploit this approach to generate rich data regarding the complexities of nanoparticle coarsening, ripening, and evaporation. Au nanoparticles created through colloidal synthesis approaches undergo a combination of both evaporation and diffusive mass transport. We have developed an analytical model that describes this process and shows how local and long-range particle interactions through diffusive transport affect the evaporation process. The extensive data of the evolution of several hundred particles allows us to determine physically reasonable values for the model parameters, quantify the particle size at which Gibbs-Thompson pressure accelerates the evaporation process, and explore how individual particle interactions deviate from the mean-field model. [2]

Finally, during the evaporation process, we see strong evidence of the formation of specific facets as the particles shrink. We have used kinetic Monte Carlo methods and Density Functional Theory to investigate the atomistic processes in detail, and show a correlation between evaporation the formation of {311} facets and the evaporation process [3].

References:

[3] J.P.H. and E.A.S. acknowledge support through the National Science Foundation, Division of Materials Research, Metals and Metallic Nanostructures Program under Grant 1809398. This research
used resources of the Center for Functional Nanomaterials, which is a U.S. DOE Office of Science Facility, at Brookhaven National Laboratory under Contract No. DE-SC00127044. P.W.V. acknowledges financial assistance under award 70NANB19H005 from U.S. Department of Commerce, National Institute of Standards and Technology as part of the Center for Hierarchical Materials Design. The authors would like to thank Kim Kisslinger at the Center for Functional Nanomaterials, Brookhaven National Laboratory for assistance with data collection, and Katherine Elbert and Christopher Murray from the Department of Chemistry, University of Pennsylvania for help with nanoparticle synthesis and self-assembly.