doi:10.1017/S1431927622010947
Microsc. Microanal. 28 (Suppl 1), 2022

Improved Throughput, Statistics, and Instrument Utilization with Automated Analytical Electron Microscopy

David A. Cullen,[1*] Haoran Yu,[1] Michael J. Zachman,[1] Kimberly S. Reeves,[1] Jaehyung Park,[2] Nancy N. Kariuki,[2] Deborah J. Myers, [2] Leiming Hu,[3] Kenneth C. Neyerlin,[3] and Rangachary Mukundan[4]

1. Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, Oak Ridge, TN, USA
2. Chemical Sciences and Engineering Division, Argonne National Laboratory, Lemont, IL, USA
3. Chemistry and Nanoscience Center, National Renewable Energy Laboratory, Golden, CO, USA
4. Materials Physics and Applications Division, Los Alamos National Laboratory, Los Alamos, NM, USA
* Corresponding author: cullenda@ornl.gov

Scanning transmission electron microscopy (STEM) can provide key insights into the structures of materials at the nanoscale. However, for inhomogeneous materials, there is always a question of the statistical relevance of findings derived solely from microscopy datasets, which sample only femtograms of the source material. This is particularly true for the case of nanoparticle electrocatalysts like those used in water electrolyzers and hydrogen fuel cells. STEM is often used to quantify particle size distributions in these devices, but conventional approaches typically sample fewer than 1000 particles. Attempting to draw structure-property relationships from these limited datasets can be particularly problematic when particle size distributions vary both across and along the electrodes [1].

In this work, automated data acquisition software (Thermo Scientific Velox/MAPS) has been coupled with custom Python codes utilizing high performance computing resources to increase nanoparticle sampling by three orders of magnitude. Since the process is automated, the acquisition and analysis of these more robust datasets requires fewer labor hours than conventional approaches, while also significantly increasing instrument utilization, as the automated imaging can be performed unattended overnight. This synergy further increases throughput, allowing for multiple datasets per sample to be obtained and/or a greater number of samples to be analyzed.

This automated approach will be demonstrated on a series of membrane electrode assemblies (MEAs) used to develop accelerated stress tests (ASTs) for heavy-duty fuel cell vehicle applications [2]. One such data set is shown in Figure 1, spanning the 6 micron-thick cathode of a tested MEA. We will discuss how these larger datasets allow particle size distributions to be determined as a function of position within the electrode, enable total Pt dissolution to be estimated, and facilitate quantification and correlation of changes in surface area with electrochemical measurements.

Despite this increase in sampling, automated electron microscopy methods still sample a relatively small portion of the 25 cm^2 active fuel cell MEA area used in most lab-scale tests. Thus, these automated microscopy studies must still be complemented by a broader infrastructure of bulk particle size analysis approaches, such as micro-X-ray diffraction and small angle X-ray scattering (SAXS) [1,3]. We will present correlative SAXS data and emphasize the complementary role played by each technique in determining particle size distributions and compositions. We will then look forward to emerging edge computing methods which will allow for real-time data analysis, enabling more efficient energy-dispersive X-ray spectroscopy (EDS) mapping of individual particles, as shown in Figure 2. The incorporation of machine learning and artificial intelligence to further reduce the need for operator input and artefacts arising from overlapping particles will also be discussed [4].

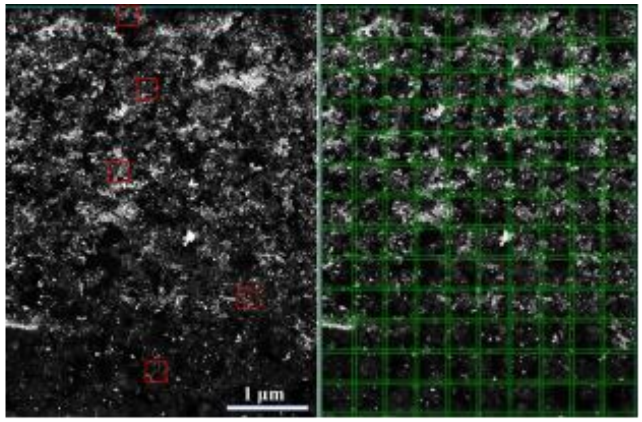

Figure 1. HAADF-STEM overview image of fuel cell cathode showing (left) conventional dataset: 5 images (red boxes) representing around 500 particles; and (right) fully automated dataset: 130 images (green boxes) representing >50k particles.

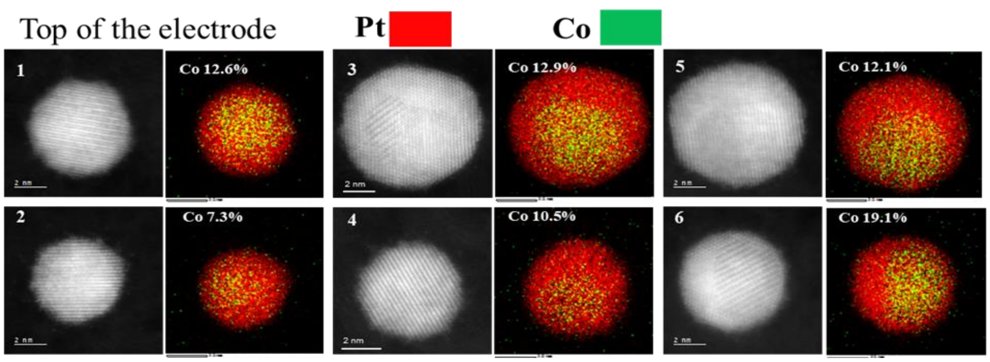

Figure 2. High-resolution HAADF-STEM and EDS spectrum images for Pt (red) and Co (green) showing the Pt-shell on the PtCo core. The label in each map shows the overall Co at.% quantified relative to Pt.

References:

[1] K. Khedekar et al., Adv. Energy Mater. **11** (2021), p. 2101794.
[2] DA Cullen et al., Nat. Energy, **6** (2021), p. 462.
[3] MC Smith et al., J. Am. Chem. Soc. **130** (2008), p. 8112.
[4] This material is based on work performed by the Million Mile Fuel Cell Truck (M2FCT) Consortium, technology managers Greg Kleen and Dimitrios Papageorgopoulus, which is supported by the U.S. Department of Energy, Office of Energy Efficiency and Renewable Energy, Hydrogen and Fuel Cell Technologies Office. Microscopy was performed at the Center for Nanophase Materials Sciences, which is a US DOE, Office of Science User Facility.. The Talos F200X S/TEM tool provided by US DOE, Office of Nuclear Energy, Fuel Cycle R&D Program and the Nuclear Science User Facilities.

https://doi.org/10.1017/S1431927622010947 Published online by Cambridge University Press