Towards the “Renaissance Era” in in situ/Operando Electron Microscopy: From Ultrathin (UT) Window Fluidic-Cell to Adaptive Sampling & Data Analytics

Vinayak P Dravid

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Meeting-report

Towards the "Renaissance Era" in *in situ*/Operando Electron Microscopy: From Ultrathin (UT) Window Fluidic-Cell to Adaptive Sampling & Data Analytics

Vinayak P. Dravid[1,2,3,*]

[1]*Department of Materials Science and Engineering, Northwestern University, Evanston, IL, United States*
[2]*International Institute for Nanotechnology, Northwestern University, Evanston, IL, United States*
[3]*The NUANCE Center, Northwestern University, Evanston, IL, United States*

*Corresponding author: v-dravid@northwestern.edu

Electron microscopy is often criticized as subjective, tedious, slow, complicated, and less amenable to automation. In particular, specialized *in situ* EM experiments are often considered hard to reproduce (especially in another lab) or compare with complementary- or correlate with- synergistic methods, like synchrotron (or neutron) scattering and/or super-resolution microscopy.

Inspired by the remarkable advances in high-throughout assays and related innovations in biomedicine, the ongoing work in our group is meant to dispel these notions. We are striving to demonstrate that *in situ* EM measurements can be consistent, reproducible and amenable to "round-robin" type sharing to validate important observations and cross-correlative phenomena. This is based on the synergistic approach to combine novel design and nanofabricated *in situ* stages [1, 2] with *smart* imaging [3, 4] to utilize electron dose and exposure in a commensurate manner. This approach can be tailored to "*ration*" electrons and time, both spatially and temporally, utilizing AI/ML methods. The electron rationing is greatly facilitated by the advent of direct electron detectors (DEDs) while the exposure considerations are driven by sparse imaging and AI/ML-enabled acquisitions, that are being pursued in a larger collaborative group at NU.

In the first phase of this initiative, we are working to overcome some of the major experimental shortcomings of currently available EM fluidic-stages: (i) fluidic-cells require thicker SiNx membranes (>30–40 nm) to maintain mechanical integrity and stability in use. Thicker membranes severely limit spatial resolution due to chromatic aberrations, and analytical/spectroscopic signal weak and prone to artifacts, (ii) lack of reproducibility, consistency and inability to "share" the same experiment across different collaborators and/or platform/microscopes. and (iii) lack of control over driving force (e.g. pumping of liquids), invariably resulting in unacceptable sample instability such as drift, vibrations etc.

Towards these goals, we have developed design and procedures for nanofabrication of ultrathin (UT) window fluidic-stage chips. This *in situ* chip design is inspired by the classical honeycomb framework that anchors ultrathin (<~5–10 nm) SiNx membranes, which can withstand extreme environment and severe constraints in typical S/TEMs. Yet, this novel UT window chip design offers field of view *orders* of magnitudes larger than current "best in class"- with consistency, reproducibility, and commercially viable yield. The obvious *scientific* advantages of SiNx of ~5–10 nm thickness include superb spectral and spatial resolution ("t/λ" of < ~0.3 compared to typical > 0.7), among other attributes (see separate presentations elsewhere). On the second front of data acquisition, we are combining smart imaging and tailored exposure to minimize redundancy, coupled to appropriate AI/ML-enabled decision making for adaptive and time-resolved sampling. Collectively, our approach makes it possible to ration both time and electrons to spread more efficiently to gather information that is spatially encoded (for high throughput discovery) or temporally monitored, as in *in situ* measurements under timed stimuli.

The presentation will cover emerging opportunities in innovative UT window fluidic-cells that allow for low- and core loss EELS for nanoscale discrimination of reactants and product gasses during catalysis, approaching atomic-resolution in real-(and fast) time. The presentation will also explore the feasibility of AI/ML-enabled data acquisition approach for rapid and high throughput characterization, as well as monitoring of *in situ* processes and phenomena in the temporal domain [5].

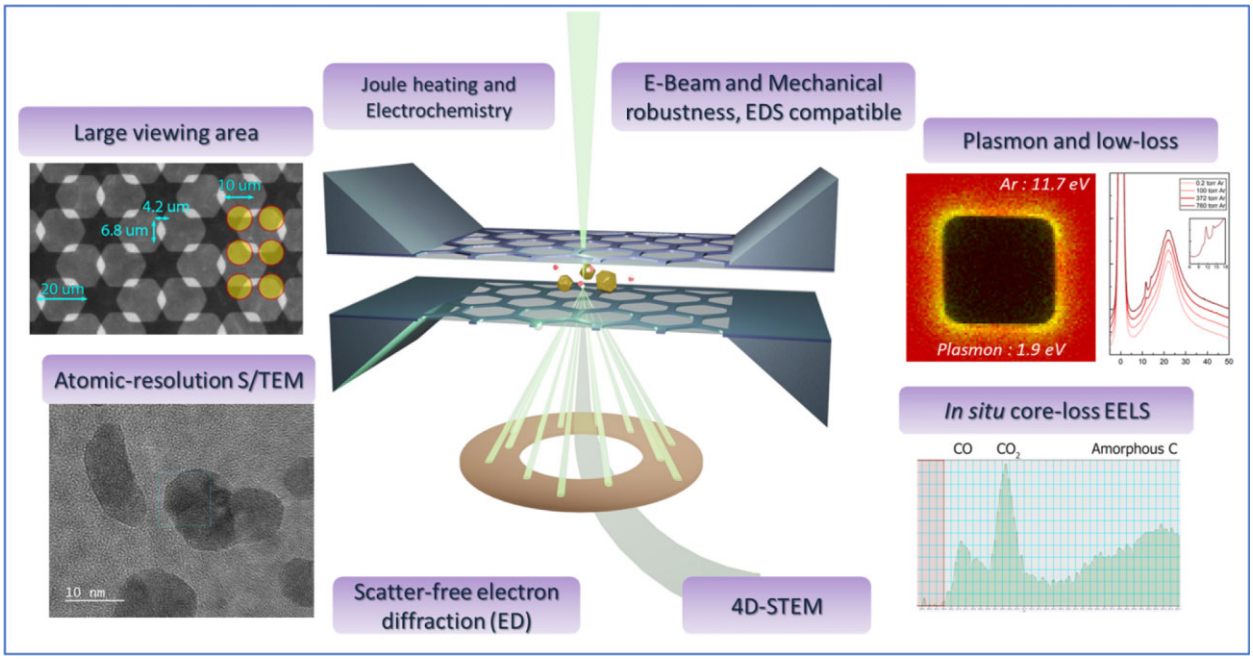

Fig. 1. The UT Window analytical fluidic-cell set-up: Schematic illustration of ultrathin (UT) SiNx and associated analytical methods. The honeycomb support structure maintains stability and provides robust support even with ultrathin (~ 5–10 nm) SiNx window. The reduced chromatic aberration enables resolution down to atomic-scale and facilitates spectral discrimination of fluidic species, in situ.

References

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3. Ribet, et al., *Materials Today* (2021): 10.1016/j.mattod.2021.05.006.
4. K. Hujsak *et al*, *Micron* (2018), DOI: 10.1016/j.micron.2018.03.001.
5. A US Provisional Patent Application (No. 63413097) regarding this work is filed. This material is partly based upon work supported by the National Science Foundation under Grant No. DMR-1929356 and AFOSR (FA9550-22-1-0300). This work made use of the EPIC facility of Northwestern University's NU*ANCE* Center, which has received support from the SHyNE Resource (NSF ECCS-2025633), the International Institute of Nanotechnology (IIN), and Northwestern's MRSEC program (NSF DMR-1720139). The author is grateful to the NU collaborative AI/ML team (Profs. Wei Chen, Dan Apley, Ankit Agarwal), the mega-library materials discovery group (Profs. Chad Mirkin and Chris Wolverton), and many students as well as NU*ANCE* staff (X. Hu, K. Koo, T. Abbott, R. Bleher, P. Smeets and R. dos Reis) for their invaluable contribution in advancing this area.

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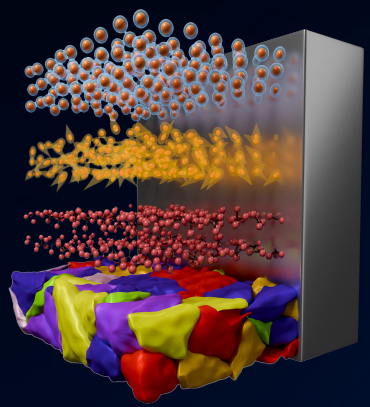

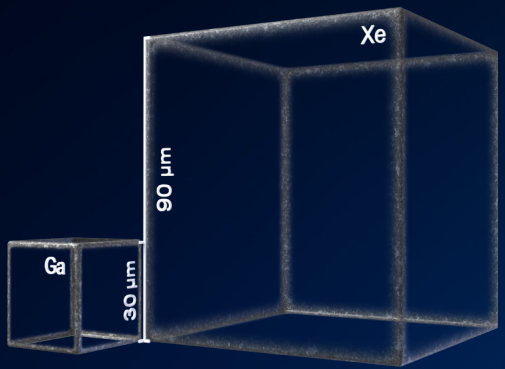

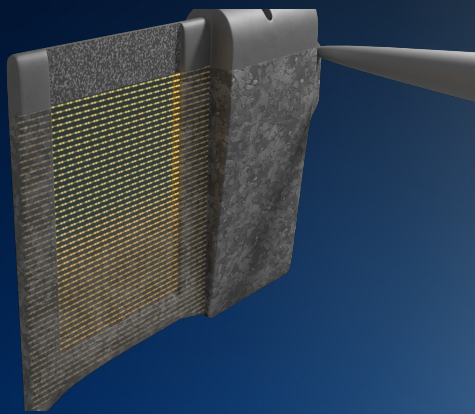

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