https://doi.org/10.1038/s41929-026-01509-8

# A new era in operando

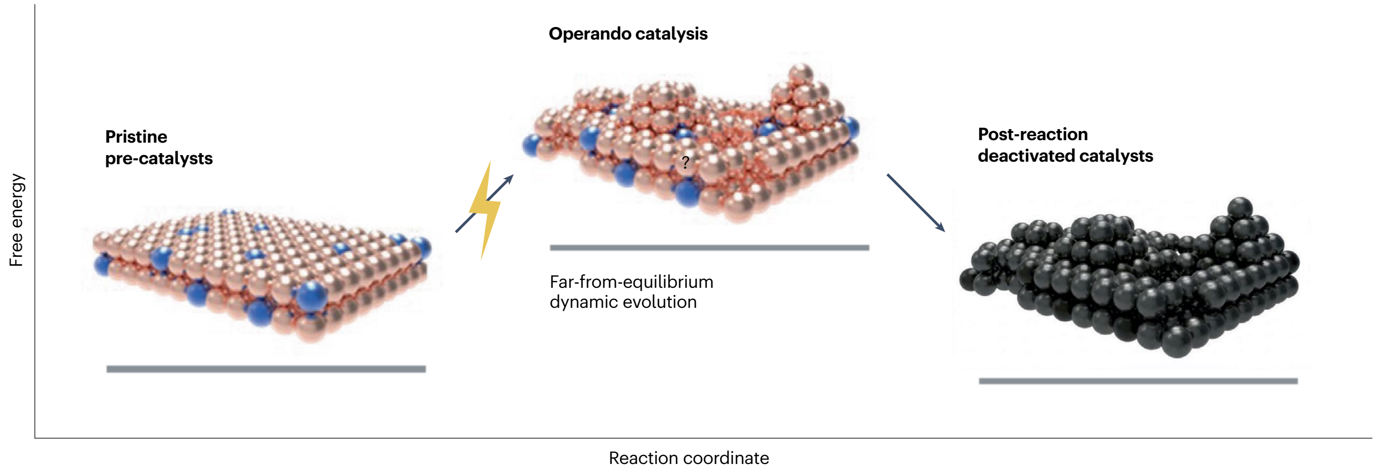
**The 2025 Materials Research Society (MRS) Fall Meeting highlighted the latest developments at the frontiers of materials research. This report focuses on multimodal operando/in situ methods employed to probe dynamic evolution of catalysts under far-from-equilibrium operating conditions – a key challenge in advancing catalyst development.**

Catalysis is central to building a sustainable and environmentally responsible future. The rational design of heterogeneous catalysts with high activity and long-term durability is essential to accelerate the global transition toward a carbon-neutral energy landscape, which in turn requires a fundamental understanding of reaction mechanisms and active site structures. However, catalysts undergo profound morphological and chemical transformation under far-from-equilibrium operating conditions (for example, strongly reducing/oxidizing electrochemical potentials), leading to substantial discrepancies between pristine pre-catalysts and the true active phases that emerge during operation. This inherently dynamic nature of catalyst structure and activity requires multimodal operando characterization approaches capable of directly monitoring catalysts during performance cycles to bridge the knowledge gap of their working principles in practical operating conditions. The 2025 edition of the MRS Fall Meeting, as usual, returned to Boston's Hynes Convention Center where it captured a rapidly growing and interdisciplinary community in developing operando methodologies to push the frontiers of fundamental understanding and applications of catalysts. Across three representative heterogeneous catalysis symposia including but not limited to electrocatalysts, thermal catalysts, and photocatalysts, numerous presentations highlighted recent advances in developments and applications of operando analytic techniques including optical microscopy, electron microscopy, vibrational spectroscopy and synchrotron-based X-ray methods, as well as their integration with cutting-edge machine learning and data mining methods, paving the way for broad implementation of operando methods in chemistry and energy material communities.

## Operando electrocatalysis

Minhua Shao (Hong Kong University of Science and Technology) investigated dynamic molecular behaviours on catalysts surfaces using in situ surface-enhanced infrared absorption spectroscopy (SEIRAS). Anthony Shoji Hall (University of Pennsylvania) discussed the critical role of interfacial water structures, probed by in situ shell-isolated nanoparticle-enhanced Raman spectroscopy (SHINERS), in promoting electrochemical C–C coupling, where disordered water structures effectively enhance the selectivity of CO reduction toward multi-carbon products[1]. Peidong Yang (UC Berkeley) and Miquel Salmeron (Lawrence Berkeley National Laboratory) reported the potential-dependent adsorbate detachment mechanism from Ag nanoparticles, tracked by in situ infrared nanospectroscopy (nano-FTIR) and surface-enhanced Raman spectroscopy (SERS), and subsequent formation of an electrocatalytic microenvironment that enables efficient $CO_2$-to-CO conversion by creating a confined localized field near the nanoparticle surface[2]. Haotian Wang (Rice University) presented the bicarbonate salt formation mechanism during the $CO_2$ reduction reaction ($CO_2RR$) in membrane electrode assembly (MEA) electrolyzers. In situ optical microscopy and Raman spectroscopy reveal that carbonate ions within liquid droplets transform into bicarbonate ions driven by $CO_2$ flow as the droplet migrates from the catalyst/membrane interface towards the backside of the gas diffusion electrode[3]. These in situ insights informed their research to demonstrate suppression of salt formation by flowing $CO_2$ gas into an acidic bubbler, leading to >1,000-hour stability in MEA operation.

Other studies highlighted the promise of in situ/operando electron microscopy and X-ray spectroscopy to probe structural evolution in nanomaterials under operating conditions. Beatriz Roldán Cuenya and See Wee Chee (Fritz Haber Institute) discussed potential-dependent kinetics of the $Cu_2O$-to-Cu reduction during nitrate



CREDIT: ADAPTED FROM I. KIM ET AL. NAT. CATAL. 8, 697–713 (2025); SPRINGER NATURE LTD.

reduction reaction by tracking time-resolved structural changes in $Cu_2O$ nanocubes and quantified oxidation states of these changing catalysts, monitored by multimodal operando electrochemical liquid-cell transmission electron microscopy (EC-TEM) and electrochemical liquid-cell transmission X-ray microscopy (EC-TXM), respectively[4]. Tyler Mefford (UC Santa Barbara) also employed EC-TXM to obtain space-resolved maps of Mn oxidation states of $\alpha$-$K_xMnO_2$ nanorod under oxygen reduction reaction and oxygen evolution reaction conditions in alkaline media. Haimei Zheng (Lawrence Berkeley National Laboratory) presented their advanced EC-TEM method achieving atomic resolution to directly monitor the dynamics of Cu catalysts during the $CO_2RR$, revealing liquid-like amorphous interphase with mediating surface reconstruction[5]. Yao Yang (Cornell University) developed an operando electrochemical liquid-cell scanning TEM (EC-STEM) method that has the potential to probe catalyst (de)activation across a wide temperature range from −50 to 300 °C. Electrochemical four-dimensional STEM (4D-STEM), assisted by machine learning, shows the potential to reveal the complex structure of beam-sensitive energy materials during operating conditions[6]. Joe Patterson (UC Irvine) discussed an influence of fresh electrolyte supply through a flow system on electrochemical reaction kinetics. Vasiliki Tileli (EPFL) presented advanced electrochemical liquid-phase SEM (EC-SEM) employing free-standing graphene as both liquid-cell membrane and electrode material to improve the spatial resolution and expand the stability of the potential window[7]. Hugo Pérez (DENSsolutions) exhibited a gas-cell TEM holder that enables concurrent heating and biasing control, allowing for operando investigation of solid oxide fuel cells operating under 600–800 °C.

## Operando thermocatalysis

Wenjie Zang (University of Rochester) discussed distinct migration behaviours of Pt single atoms on $TiO_2$ substrates under reducing environment, moving from substrate surfaces to substrate–substrate interfaces under 760 torr of 5% $H_2$/Ar atmosphere at 450 °C, tracked by in situ gas-cell TEM, which causes attenuated CO adsorption on the Pt single atom sites[8]. Alexandre Foucher (Oak Ridge National Laboratory) reported a multimodal gas-cell TEM approach integrated with electron energy-loss spectroscopy (EELS) and 4D-STEM to investigate zeolite-based nanocatalysts, revealing migration of Al during calcination and changes in crystallinity of Cu and Zn under $H_2$ or $O_2$ environments. Eric Stach (University of Pennsylvania) introduced multiple examples studying structural evolution in diverse nanocatalysts under various reaction conditions through in situ gas-cell TEM. Additionally, he showcased an ultra-high-pressure gas-cell TEM holder ensuring a STEM image resolution of 1–2 nm from 0 to 27.5 bar, bridging the pressure gap between laboratory studies (1–2 bar) and industrial conditions (10–100 bar) (ref. 9). Vinayak Dravid (Northwestern University) presented an advanced gas-cell TEM approach integrated with ultrathin $SiN_x$ window to effectively reduce electron scattering through the window, resulting in improved imaging resolution and spectral signal.

Miaofang Chi (Oak Ridge National Laboratory) employed in situ environmental TEM (ETEM) to investigate sintering mechanisms of $Pt/Al_2O_3$ under controlled gas environments such as vacuum, $H_2$, $O_2$, and $H_2$–$O_2$ mixture, unravelling that the mixed environment significantly accelerated the sintering rate with a previously unobserved caterpillar-like translational migration of Pt nanoparticles[10]. Wei-Chang David Yang (NIST) discussed how structural changes and local chemical environments affect $CO_2$ capture and conversion in amine-functionalized mesoporous silica at the nanoscale, probed by ETEM coupled with EELS and Raman spectroscopy. Frances Ross (MIT) introduced a secondary imaging mode in ETEM to obtain three-dimensional (3D) surface-sensitive topology, bridging the information gap between two-dimensional STEM projection data and real-time structural evolution in 3D space under gaseous environments[11]. Stig Helveg (Technical University of Denmark) reported an ETEM approach exhibiting ~50 pm of resolution at pressures up to 1 mbar, which is achieved by adopting a specialized TEM pumping system, minimized e-beam penetration depths through gas layers, a high-brightness field emission gun, dual aberration correction, and a direct electron detection camera. Judith Yang (Brookhaven National Laboratory) presented a machine learning workflow integrated with ETEM experiments, enabling image denoising and automatic feature extraction, which accelerates interpretation of vast and complex time-resolved ETEM datasets[12].

Other presentations illustrated the promise of operando X-ray diffraction analysis for fundamental understanding of catalytic active site reconstruction. Michael Toney (University of Colorado Boulder) used multimodal X-ray scattering and imaging to track the formation of nanoscale Zn clusters in aqueous Zn batteries. David Simonne (MIT) reported a multiscale operando diffraction approach combining operando surface X-ray diffraction (SXRD), Bragg coherent diffraction imaging (BCDI), and crystal truncation rod (CTR) analysis to elucidate the structural evolution of Pt nanoparticles supported on $\alpha$-$Al_2O_3$ during $NH_3$ oxidation.

## Operando photocatalysis

Jennifer Dionne (Stanford University) discussed photocatalytic $NH_3$ production mechanisms from the AuRu bimetallic nanocatalysts. In situ diffuse reflective infrared Fourier transform spectroscopic (DRIFTS) unravelled that light illumination accelerates the hydrogenation of $N_2$, enhancing $NH_3^*$ coverage while suppressing partially hydrogenated $NH_x^*$ species, overcoming the potential limiting factor in thermal catalytic reactions in which $NH_3$ is decomposed at higher temperatures[13]. Additionally, optically coupled gas-cell TEM revealed that their structure underwent dynamic atomic rearrangement under the presence of both gas and light illumination, including surface faceting and core-shell structure formation. Harry Atwater (Caltech) reported operando mapping of local pH in Au/p-GaN electrodes during photoelectrochemical water splitting and $CO_2RR$ by employing fluorescent confocal microscopy combined with pH sensitive fluorescent dyes.

## Operando-freezing approach for beam-sensitive materials

There were multiple presentations exhibiting cryogenic techniques for beam-sensitive materials analysis with minimum beam-induced damage. Yi Cui (Stanford University) introduced the decade long development of multimodal EM characterizations for advancing next-generation batteries, specifically highlighting cryogenic electron microscopy (cryo-EM) and X-ray photoelectron spectroscopy (cryo-XPS)[14]. While these techniques are highly applicable to beam-sensitive catalysts, their applications have mostly been limited to post-reaction materials. At this meeting, operando-freezing approaches were reported where energy materials under operating conditions are rapidly plunged into cryogenic temperatures to kinetically trap their active structures. They rapidly froze a TEM grid containing mixed-halide perovskites under light illumination, preserving transient photoexcited ion distributions within the materials, which revealed the photo-induced iodine migration from grain boundaries to centres and subsequent development of anisotropic strain. Yuzhang Li (UCLA) developed an electrified cryo-EM (eCryo-EM) technique that enables electrical biasing to a TEM grid, which is rapidly frozen to explore non-equilibrium states during battery operation[15]. Collective snapshots of the operating materials at controlled time intervals quantified early-stage solid electrolyte interphase growth kinetics. Such operando-freezing approaches hold significant promise for elucidating atomic-scale dynamic processes of beam-sensitive catalysts under operating conditions, and bridge the knowledge gap between ex situ atomic-scale cryo-TEM and in situ/operando nm-scale time-resolved liquid-cell TEM.

## The future is operando

The development of operando characterization techniques will be crucial for the advancement of catalysis science across diverse fields including electro-, thermo-, and photocatalysis. We anticipate that, with continuous technical advances such as those presented at the MRS Fall Meeting, multimodal operando methods will become indispensable for understanding dynamic catalytic mechanisms for the broad chemistry and energy materials communities.

**Sungin Kim[1], Haichuan Zhang[1], Qian Chen[2], Jungwon Park[3], Dongsheng Li[4] & Yao Yang[1]** ✉

[1]Department of Chemistry and Chemical Biology, Baker Lab, Cornell University, Ithaca, NY, USA. [2]Department of Materials Science and Engineering, The Grainger College of Engineering, University of Illinois Urbana-Champaign, Urbana, IL, USA. [3]School of Chemical and Biological Engineering, Institute of Chemical Processes, Seoul National University, Seoul, Republic of Korea. [4]Physical Sciences Division, Pacific Northwest National Laboratory, Richland, WA, USA.

✉e-mail: yaoyang@cornell.edu

## References

1. Zhang, H., Raciti, D. & Hall, A. S. *Nat. Chem.* **17**, 1161–1168 (2025).
2. Shan, Y. et al. *Nat. Catal.* **7**, 422–431 (2024).
3. Hao, S. et al. *Nat. Energy* **10**, 266–277 (2025).
4. Yoon, A. et al. *Nat. Mater.* **24**, 762–769 (2025).
5. Zhang, Q. et al. *Nature* **630**, 643–647 (2024).
6. Kim, S. et al. *J. Am. Chem. Soc.* **147**, 23654–23671 (2025).
7. Toleukhanova, S. et al. *Adv. Mater.* **36**, 2311133 (2024).
8. Zang, W., Lee, J., Christopher, P. & Pan, X. *Microsc. Microanal.* **30**, ozae044.788 (2024).
9. Hollyer, G. G., Zakharov, D. N., Alsem, D. H., Parkin, C. & Stach, E. A. *MRS Commun.* **15**, 898–905 (2025).
10. Smith, J. et al. *ACS Mater. Lett.* **6**, 3301–3311 (2024).
11. Lee, S. et al. *ACS Nano* **18**, 21258–21267 (2024).
12. Li, M., House, S. D. & Yang, J. C. *Nat. Mater.* **24**, 664–665 (2025).
13. Yuan, L. et al. *Nat. Energy* **11**, 98–108 (2026).
14. Cui, Y., Zhang, Z., Sinclair, R., Chiu, W. & Cui, Y. *Nat. Rev. Phys.* **8**, 40–54 (2026).
15. Wang, C. et al. *Sci. Adv.* **11**, eadv3191 (2025).

## Competing interests

The authors declare no competing interests.