

Expanding the Toolbox of Carbon-Neutrality Research with Modulation-Excitation

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Abstract

The realization of a carbon-neutral future requires advancements in chemical technologies. From CO₂ emission reduction to its capture and utilization, the heart of such processes are materials with optimal performances. Establishing accurate descriptions of materials under their working conditions remains a challenge. Accordingly, modulation-excitation (ME) techniques have been pioneered and utilized to gain fundamental understanding of materials and their dynamics.

We have coupled ME with X-ray absorption spectroscopy (XAS)¹ and small-angle X-ray scattering (SAXS)² to unravel surface and interior dynamics, respectively. Both XAS and SAXS are bulk-sensitive techniques. The subtle surface and interior dynamics that are crucial for understanding catalytic processes, for example, typically induce too small changes relative to the bulk XAS spectra and SAXS patterns. We show that the utilization of ME-XAS and ME-SAXS is effective in capturing and extracting surface and interior dynamics with high signal-to-noise (S/N) ratios. ME-XAS was implemented to investigate methane oxidation on Pt/Al₂O₃ of varying particle sizes whereas ME-SAXS was used to detect the structural changes of Nafion films upon hydration.

In the last example, ME is coupled with diffuse reflectance infrared Fourier transform spectroscopy (DRIFTS) to investigate thermal CO₂ hydrogenation to methanol over Cu-based catalysts.³ Signals detected by *in-situ* DRIFTS include spectators in addition to active species participating in the catalytic cycle. ME-DRIFTS removed the spectator signals and enabled the identification of CO₂ hydrogenation mechanism, rate-limiting steps, and role of Ga in shifting the selectivity from CO to methanol.

References

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