Event-averaged time-resolved APXPS with chemical perturbations: studying gas/solid processes with a microsecond time resolution

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Recently at MAX IV, we have developed an event-averaging time-resolved Ambient Pressure X-ray Photoelectron Spectroscopy (APXPS) based on chemical perturbations for observing dynamic processes with micro- to millisecond time resolution. The method makes use of the rapid change in the gas pressure/composition as a perturbation that drives the system away from its equilibrium [1,2]. In the experiment, a sharp gradient in chemical potential is created by modulating the gas composition over the catalyst via mass flow controllers or a fast valve. Such gas pulse has internal pressure in the mbar range and a rising edge of a few hundred microseconds. A gated detector based on a fast camera is synchronized with the valve operation to measure X-ray photoemission spectra with up to 40 μs time resolution. We will present several experiments characterizing the setup's performance, including the CO oxidation reaction over Pt (111) to demonstrate the capability of the setup to correlate the gas phase composition with that of the surface during the transient supply of CO gas into an O_2 stream [1]. These experiments demonstrate that under CO pressure modulation conditions, the system remains active (i.e. producing CO2) at temperatures below the CO lift-off temperature (that is inactive under the flow conditions).

1. A. Shavorskiy et al. ACS Applied Materials & Interfaces 2021 13 (40), 47629-47641.
2. J. Knudsen et al. Nat Commun 12, 6117 (2021).