Contribution ID: **170** Type: **Contributed talk**

Time-Resolved Ambient Pressure Photoelectron Spectroscopy for Studying Mechanisms of Catalytic Reactions under Operando Conditions

Friday 30 August 2024 15:30 (15 minutes)

Recently, a new time-resolved Ambient Pressure X-ray Photoelectron Spectroscopy (APXPS) method based on chemical perturbations has been developed for studying dynamic processes with microsecond time resolution. The method uses the rapid change in the gas pressure/composition as a perturbation that drives the system away from equilibrium [1,2]. In the experiment, a sharp and strong gradient in chemical potential is created by modulating the gas composition over the catalyst via a fast valve. Such gas pulse has internal pressure in the mbar range and a rising edge of a few hundred microseconds. A time-sensitive delayline detector is synchronized with the valve operation to measure X-ray photoemission spectra with nano- to microsecond 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 O2 stream (Figure 1) [3]. 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, i.e. under the flow conditions. We will also demonstrate that the chemisorbed oxygen is observed during the catalytic cycle after the Pt(111) surface is saturated with the oxide. This points out a much higher activity of the O_chemisorbed towards CO oxidation than O_oxide, resolving the ongoing debate about the role of the Platnum surface oxide in the reaction.

Figure 1. Time-resolved APXPS measured during pulsing CO into a constant flow of O2 onto Pt(111) at 330 C. (Left) O 1s gas phase and surface time-resolved spectral maps. (Right) individual spectra measured at time delays indicated by the dashed arrows.

[1] J. Knudsen et al. Nat Commun 12, 6117 (2021). doi:10.1038/s41467-021-26372-y
[2] A. Shavorskiy et al. ACS APPLIED MATERIALS & INTERFACES 13, 47629 (2021). doi: 10.1021/acsami.1c13590
[3] C. Eads et al. in preparation

I plan to submit also conference proceedings

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Session Classification: Mikrosymposium 12/3: Time Resolved Techniques

Track Classification: 12. Time resolved techniques