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C K-edge Selective Probing Ultrafast Surface Chemistry in Catalytic CO Oxidation on Ru (0001)

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Nearly all chemical processes involved in energy conversion utilize catalytic chemical transformations at interfaces between solids and liquids or gases. While most of the existing understanding is based on a static view of reactions at interfaces, the X-ray Free Electron Laser (FEL) opens up the dynamic regime where studies of the reaction mechanism to observe transformations on timescales down to femtoseconds becomes possible. In order to understand the reaction mechanism, it is essential to unveil and characterize the transient short-lived intermediates and transition states under ultrafast time scales. In the previous LCLS experiments, we demonstrated i). the transient precursor state that weakly interacts with the surface from CO desorption on Ru1 and ii). the transition state region in CO oxidation on Ru,2 both at the O K-edge. Recently we brought the selective probing of ultrafast chemistry in CO oxidation on Ru (0001) surface to the C K-edge, which should add complementary knowledge to its previous O K-edge counterpart. The experiments were performed at DiProl beamline at FERMI in Elettra, where the energy range of FEL is capable to reach the C K-edge with an installed high harmonic undulator. The endstation used in the current experiment were transferred from Stockholm to FERMI, which equips with X-ray absorption (XAS) and X-ray emission detectors and allowing the record of reaction products by mass spectrometry. During the pump-probe FEL experiment, 400 nm optical laser was used as a pump, while the FEL pulses were scanned across the carbon 1s to $2\pi^*$ resonance in the range of 286-296.5 eV. For XAS measurements, both linear horizontal and linear vertical polarized FEL pulses were applied in order to observe the molecular states with different geometries, e.g. π and σ states. Such measurements allowed the chemical- and site-specific observation of molecules in the catalytic reaction. Furthermore, by tuning the delays between laser and FEL, the electronic structure evolution down to the timescale < 1 ps was recorded, relevant to the transition state formed during the ultrafast catalytic processes. Information of the transient states obtained from the current work are under analysis, and the theoretical understanding will be added. Detailed results and conclusions will be present in the poster of this abstract during Science@FELs 2018.

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