X-RAY PHOTOELECTRON SPECTROSCOPY OF ELECTROCHEMICAL INTERFACES FOR SOLAR FUEL PRODUCTION

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Solar fuels could resolve increasing demand for energy but fuel production lacks efficient catalyst materials. Limiting reaction steps include the Oxygen Evolution Reaction (OER) and the CO_2 Reduction Reaction (CO_2RR) which are crucial for hydrogen and hydrocarbon fuel production using only sunlight, water and carbon dioxide as inputs. Operando analysis of reaction intermediates at the solid-liquid interface provides fundamental understanding of catalytic reaction mechanisms and structure-activity/selectivity relationships, which can guide the design of superior electrocatalysts.

At present, X-ray Photoelectron Spectroscopy (XPS) probing of the solid-liquid interface is limited to electrochemical operation at rather low current densities. Recently, "tender" X-ray Ambient Pressure XPS and a dip-and-pull electrochemical cell depicted in Fig. 1 were utilized to study Ni–Fe electrocatalyst at different potentials [1]. The approach allowed operando measurements just above the onset of OER. A two-dimension model was used to describe the spatial distribution of electrochemical potential, current density and pH as a function of the position above the electrolyte meniscus and to provide guidance towards enabling the acquisition of operando XPS at high current density. The current density of 10 mA/cm^2 is the desired operation condition in photoelectrochemical devices. New electrochemical cell designs and early results allowing higher current densities will be presented.

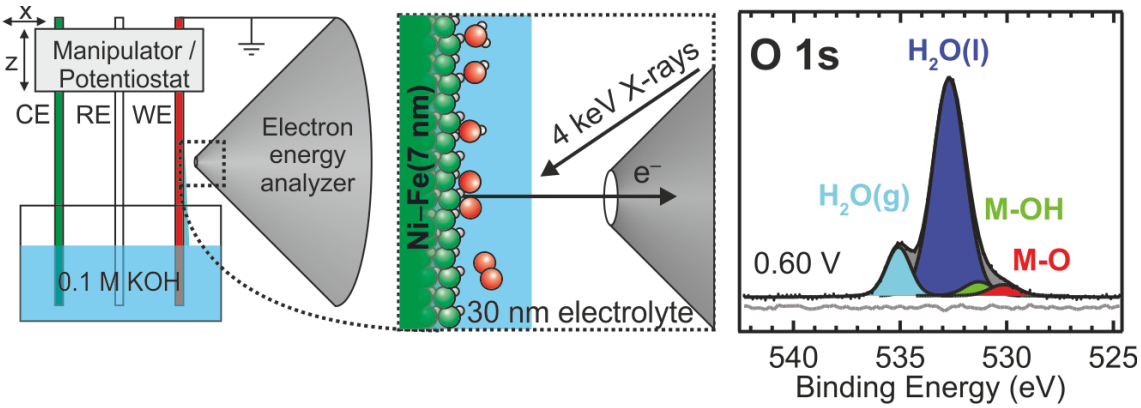

Fig. 1 – Schematic illustration of the dip-and-pull electrochemical cell at the Advanced Light Source (ALS) "tender" X-ray Ambient Pressure XPS beamline 9.3.1.

[1] Ali-Löytty, H. *et al.* Ambient-Pressure XPS Study of a Ni-Fe Electrocatalyst for the Oxygen Evolution Reaction. *J. Phys. Chem. C* (2016). doi:10.1021/acs.jpcc.5b10931