**Developing characterization strategies for photocatalytic oxynitrides: a SrTaOxNy case study**

Zahra Pourmand Tehrani,†‡ Christof W. Schneider,‡ Daniele Pergolesi,‡ Thomas Lippert†‡*§*

*†Laboratory of Inorganic Chemistry, Department of Chemistry and Applied Biosciences, ETH Zürich, Vladimir Prelog Weg 1, CH-8093 Zürich, Switzerland*

*‡ Laboratory for Multiscale Materials Experiments, Paul Scherrer Institute, 5232 Villigen PSI, Switzerland*

*§ International Institute for Carbon-Neutral Energy Research (I2CNER), Kyushu University, 744 Motooka 819-0395 Fukuoka, Japan*

Oxynitrides are a class of semiconductors absorbing light in the visible range (380-700 nm). The size of the bandgap and the energy position of the band edges make oxynitrides promising candidates for visible-light-driven water splitting, with their performances enhanced by co-catalysts 1. Probing the physiochemical evolution of a thin film ex-situ and under operation conditions provides insights into the physical and chemical processes occurring in the material.

Thin films possess relatively wide, well-defined and atomically flat surfaces, representing ideal model systems for the investigation of the semiconductor surface. Using this approach, we are exploring the origin for the enhanced charge extraction in a model oxynitride system, SrTaOxNy, with nickel-based (NiOx) co-catalysts. NiOx is among the most efficient metal-free co-catalysts for the oxygen evolution reaction (OER), suppressing the recombination of photo-generated electrons and holes3. Here, the OER is the bottleneck of the overall water splitting process and an improved understanding of OER mechanisms for oxynitride thin films is required.

Using ex-situ characterization techniques such as Neutron Reflectometry and Transmission Electron Microscopy, it is possible to identify material loss and modification in SrTaOxNy as a result of photo-electrochemical reactions. We identified the [011] lattice plane as a more favorable orientation compared to [001], with a significant performance enhancement upon NiOx deposition. Operando shallow angle X-ray absorption spectroscopy measurements are performed on SrTaOxNy with and without co-catalyst during photo-electrochemical solar water splitting with a custom designed photoelectrochemical cell. Using the outlined approach, we aim to answer the open question regarding the driving force behind the observed detrimental surface modifications, the synergistic effects the co-catalyst has, and the role each cation plays. Addressing all these questions has large implications on the design and potential discovery of novel high-performance materials.

**References**

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