



Abstract ID : 80

Friday 21 Oct, 9:40

In situ diffraction and PDF studies on the structure and crystallization of perovskite based catalysts for oxygen evolution reaction

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Electrochemical water splitting is an effective way to store intermediate electricity from renewable energy sources. Water electrolyzers generate hydrogen at the cathode (hydrogen evolution reaction, HER) while oxygen forms at the anode (oxygen evolution reaction, OER). OER involves a complex four electrons transfer and is therefore considered the bottleneck of the water splitting reaction.^[1] To date, iridium- and ruthenium-based materials are among the best-performing OER catalysts in alkaline conditions.^[2] However, the scarcity and high cost of these precious metals have encouraged tremendous research in efficient and low-cost alternatives for OER catalysts, among which perovskites have demonstrated excellent OER activities because of their tunable chemical and electronic properties and their stability under different reaction conditions. ^[3]

The conventional synthetic routes for phase pure perovskites need high calcination temperatures and they result in large crystallites and low active surface area. However, phase purity and accessible active surface area play a critical role in electro-catalytic performance. Our work is directed towards the low-temperature synthesis and investigation of the structure-property relationship of highly active lanthanum-based perovskites for alkaline OER. The synthesis route via solution combustion of nitrate precursor solutions in the presence of a fuel followed by calcination is optimized. As obtained perovskites show high catalytic performance for OER. In order to have a comprehensive understanding of the structure-property relationships of these catalysts, the crystallization pathway has been studied via in situ diffraction and total scattering experiments.

The synthesis leads first to the formation of spongy precursors, which are amorphous, and the solid-state reaction from the amorphous phase to the crystalline perovskite is quite fast as observed in the in situ x-ray diffraction data. To gather more information on the local atomic scale but also intermediate- and long-ranges, we performed temperature dependent total scattering experiments and subsequent pair distribution function (PDF) analysis. From the PDF refinements, we observe that the so-obtained spongy precursor after a dynamic self-combustion is a combination of distorted oxides and meta-stable higher-order perovskites. With the temperature increase, we could follow the decomposition of meta-stable phases to corresponding oxides and Ruddlesden Popper phases. Around 550°C we observe the formation of crystalline perovskite. Rietveld analysis and Fourier difference analysis reveal a deviation from the perfect crystal structure. The disorders are further studied by transmission electron microscopy, which confirms the presence of stacking faults. The combined effect of enhanced surface area, phase purity, and stacking faults are further correlated to the excellent OER performance of the catalysts.

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