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Enhancing electrolysis performance of perovskite-type electrodes by polarization-driven exsolution of metallic catalyst particles: A synchrotron-based in-situ XRD and XPS study of near-surface chemistry

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Mixed ionically and electronically conducting perovskites are nowadays employed in electrochemical devices such as solid oxide fuel cells, gas separation membranes and membrane reactors. Their future application in solid oxide electrolysis cells (SOECs) is therefore almost obvious. For a successful introduction into SOECs, however, we need an in-depth understanding of their surface chemistry.

In this study, geometrically well-defined model-composite electrodes were investigated containing perovskite thin films such as La0.6Sr0.4FeO3-δ (LSF) and La0.8Sr0.2Cr0.9Ni0.1O3-δ (LSCrNi). On LSF, electrolysis of water was performed at 620 ℃ and the electrochemical activity as well as the surface chemistry were simultaneously studied by impedance spectroscopy and near-ambient pressure X-ray photoelectron spectroscopy (NAP-XPS), respectively. Upon cathodic polarization the evolution of metallic iron species was observed, which was accompanied by a strong increase of the electrode's water splitting activity. By means of in-situ surface XRD experiments the exsolved particles were identified to be α-Fe. After retracting the applied voltage the metallic iron was immediately re-oxidized and the high water splitting activity disappeared.
On LSCrNi electrodes it is shown that this boosting effect of exsolved transition metal particles can also be transferred to CO2 electrolysis. Again simultaneous impedance and NAP-XPS measurements were performed to correlate electrochemical activity with surface chemistry and presence of metallic Ni was confirmed during electrolysis of CO2 at 700 ℃. In reference measurements on Ni free electrodes much lower electrochemical reaction rates were found, therefore confirming the beneficial effect of the exsolved Ni particles. Depending on the applied voltage the products of CO2 splitting were either CO or graphitic carbon. Graphitic depositions, which were formed under too high cathodic polarization, led to a strong deterioration of the electrode performance. But retracting the voltage completely removed the carbon and the electrode was completely reactivated.

These results clearly identify perovskite electrodes decorated with exsolved metallic particles as catalytically superior in high temperature electrolysis of H2O and CO2. Hence, this novel type of electrolysis electrodes may open new pathways in the quest for optimized energy storage via SOECs.

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