First-Principles Study of Structure Property Relationships of Monolayer (Hydroxy)Oxide-Metal Bifunctional Electrocatalysts

In the present study, on the basis of detailed density functional theory (DFT) calculations, and using Ni hydroxy(oxide) films on Pt(111) and Au(111) electrodes as model systems, we describe a detailed structural and electrocatalytic analysis of hydrogen evolution (HER) at three-phase boundaries under alkaline electrochemical conditions. We demonstrate that the structure and oxidation state of the films can be systematically tuned by changing the applied electrode potential and/or the nature of substrates. Structural features determined from the theoretical calculations provide a wealth of information that is inaccessible by purely experimental means, and these structures, in turn, strongly suggest that a bifunctional reaction mechanism for alkaline HER will be operative at the interface between the films, the metal substrates, and the surrounding aqueous medium. This bifunctionality produces important changes in the calculated barriers of key elementary reaction steps, including water activation and dissociation, as compared to traditional monofunctional Pt surfaces.

The successful identification of the structures of thin metal films and three-phase boundary catalysts is not only an important step towards accurate identification and prediction of a variety of oxide/electrode interfacial structure-properties relationships, but also provides the foundation for rational design and control of “targeted active phases” at catalytic interfaces. The successful design of bifunctional electrocatalysts that exploit these structures, in turn, could ultimately lead to advances in the development of alkaline fuel cells.

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Organisations: Department of Physics, Purdue University
Contributors: Zeng, Z., Kubal, J., Greeley, J. P.
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