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UNDERSTANDING WATER–OXIDE INTERFACES TO HARNESS NEW PROCESSES AND TECHNOLOGIES

FOCUS ISSUE

Introduction

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Water interaction with metal oxides plays a fundamentally important role across environmental chemistry, geochemistry, catalysis, and in the control and performance of functional nanocrystalline oxides. Water is found virtually everywhere on Earth, from bulk fluid to trace molecules, and its interaction with natural and engineered materials often defines or at least influences a key part of their chemical and physical behavior. In turn, metal oxides play a dominant role as mineral particles across most natural environments, while comprising a compelling and versatile platform for the development of new functional materials with tailored properties that benefit society. Given the intrinsic correlation of their properties to water interactions, it is a common goal across materials and chemical sciences to develop a comprehensive understanding of water adsorption, water film formation, and water-mediated reactions on metal oxide surfaces. This is certainly true in heterogeneous catalysis and electrocatalysis, where water is omnipresent either as a reactant, a medium, an inhibitor or as a trace component in the feed. The effect of water on the morphological evolution during synthesis, particle assembly, and sintering is also important to the stability and functional properties of oxide nanostructures. Such processes are intimately related to how water molecules interact with their surfaces and affect local physical-chemical conditions.

Despite being studied for decades, the interaction of water with matter often still defies predictive-level understanding. With increasing water activity, the adsorption layer covering oxide surfaces evolves from a solid/vacuum interface to a solid/bulk liquid one. This transition entails a complex progression of water-surface interactions that become increasingly convoluted with water-self interactions, which is difficult to unravel experimentally or encompass in computational modeling. But this regime is critical as it holds the key for understanding of physical-chemical behavior of oxides under different water coverages. Fundamental surface science studies using advanced microscopic and spectroscopic methods to analyze single-crystalline substrates have greatly advanced our understanding of water/oxide interfaces. For these essentially 2D model systems, a robust molecular-level description of interfacial water structure and reactivity has routinely been achieved by combing high resolution measurements with molecular simulations. However, advancing beyond this well-studied and highly controlled case is necessary to move the overall complexity of the interfacial system toward more realistic operational environments. In particular, increasing the water activity and the types and characteristics of the metal oxide surfaces present, including irregular defective surfaces, defines an important emerging Frontier of research.

A companion Frontier focuses on understanding the additional complexity that arises when metal oxides spatially confine water between their surfaces at the nanoscale, such as within nanopores or between nanoparticles within aggregates in aqueous suspensions. Under confinement, the convolution of water-surface and water-self interactions is even less understood, despite its fundamental importance to bulk properties of porous metal oxides or phenomena such as particle-based crystal growth. For unsupported oxide nanostructures, water adsorption and thin water film formation can lead to chemical, structural and microstructural transformations such as dissolution/precipitation, morphological changes, crystallization, or oriented attachment and water-assisted self-assembly of nanoparticles. However, due to the confinement of mass transport at the oxide surface, these processes do not proceed exactly as they do in bulk liquid water. Knowledge about the chemical and physical stability of oxide micro- and nanostructures is of key interest for their processing, such as in the

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production of ceramics or for their use as materials components in catalysis.

The goal of this *JMR* Focus Issue on water–oxide interfaces is to provide readers up-to-date information on progress in the fundamental understanding at these important research frontiers. Topics include the impact of thin water films—and the confinement of related interfaces—on structure, stability, and transformation behavior of oxide materials, taken from different perspectives spanning materials and interfacial science, surface science, and catalysis. On behalf of the Guest Editor team, we would like to express our gratitude to the authors and reviewers of these high-quality manuscripts, all of whom dedicated their time and energy to the success of this issue.

On the cover

Conversion of carbon dioxide into stable carbonate minerals for safe storage is one example of how understanding water–oxide interfaces can help harness new processes and technologies. Carbon dioxide capture, utilization, and storage are essential to help manage climate impacts of continued utilization of carbon intense fossil fuels. In geologic sequestration of carbon dioxide, reaction of minerals with supercritical CO_2 can produce stable carbonate salts, but the reaction rate and product characteristics are closely tied to amount of water i.e., available to mediate the reaction. For example, this scanning electron micrograph shows an example of the complex growth behavior of zinc carbonate from zinc oxide reacted in wet supercritical CO_2. The spherical yet polycrystalline product shape results from a complex interplay of water-mediated zinc oxide dissolution, nucleation and growth of zinc carbonate particles, and associated particle–particle interfacial forces that cause aggregation in an ordered fashion. Improving fundamental understanding of this interplay will help enable a detailed mechanistic process understanding for carbonation necessary to facilitate CO_2 storage in the subsurface. Image credit: Dr. Xin Zhang, Pacific Northwest National Laboratory.

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