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**Understanding Interfacial Proton and Hydride Transfer**

Proton and hydride transfers are complementary and critical elementary steps in energy conversion and catalysis. These reactions can be driven electrochemically via either outer-sphere electron transfer to a soluble molecular active site or can proceed via inner-sphere charge transfer directly at surface active sites on an electrode. While the former has been the subject of extensive investigations at the molecular level, relatively little is known about the latter, termed interfacial PCET (I-PCET) and hydride transfer (I-HyT). For I-PCET, we have constructed molecularly well-defined hosts of I-PCET on carbon electrode surfaces and used these systems to uncover the intrinsic pH dependent kinetics of this reaction. We find that a simple mechanistic model with contributions from hydronium, water, and hydroxide captures kinetic trends across 14 pH units and 3 order of magnitude in rate. For I-HyT, we employ chemo-selective molecular hydride acceptors to reveal that metal surfaces can catalyze the direct reduction of H2 to hydrides. Whereas molecular hydridicity is defined by the molecular structure of the donor, we find that surface hydridicity can be tuned by over 40 kcal/mol by varying the applied potential. These studies highlight the key distinctions between outer-sphere and interfacial PCET and hydride transfer and provide a foundation for designing new reactions and (electro)catalysts.

Bio

Yogesh (Yogi) Surendranath is Professor of Chemistry & Chemical Engineering at the Massachusetts Institute of Technology. He holds dual bachelor's degrees in chemistry and physics from the University of Virginia and a PhD in inorganic chemistry from MIT, obtained under the direction of Professor Daniel Nocera. After receiving his PhD, Professor Surendranath undertook postdoctoral studies as a Miller Research Fellow at UC Berkeley, under the direction of Professor Paul Alivisatos. In 2013, he launched his independent research program at MIT. The Surendranath group aims to address frontier challenges in energy conversion and sustainability by controlling interfacial reactivity at the molecular level.

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