

Mixed Oxy-Hydride Materials as PCET Reagents

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Proton-coupled electron transfer (PCET) reactions are crucial in chemical energy conversion, which involves converting electric energy into bond energy and vice versa. In these reactions, electrons generated during the oxygen evolution reaction are used to reduce small molecules such as protons, and CO₂. While PCET reactions at molecular complexes have been extensively studied for energy conversion, most literature on water splitting by materials has focused on electron transfer at the interface [1]. For example, TiO₂, widely used for photo-electro-catalytic water splitting, is typically discussed as an electron transfer reagent. However, Mayer and co-workers [2] showed that TiO₂ nanoparticles act as PCET reagents in interfacial reactions. Materials with hydridic H-atoms have been rarely studied, although M–H surface species are key intermediates in energy conversion reactions, such as the hydrogen evolution reaction (HER) or the hydrogenation of polar double bonds. This prompted us to investigate Perovskite-type oxides (ABO₃) partially substituted with hydride anions as PCET reagents. We synthesized BaTiO_{3-x}H_x through topotactical solid-state synthesis with varying amounts of hydride anions. The anionic sublattice consists of both O²⁻ and H⁻ anions surrounding Ti cations in a mixed **IV/III** oxidation state (Fig. 1) [3]. The material has been thoroughly characterized, and initial studies on its PCET properties are underway.

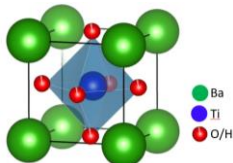


Figure 1. Crystal structure of BaTiO_{2.81}H_{0.19} showing cubic structure.

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- [2] J.N. Schrauben, et al., *Science*, 2012, **336**, 1298-1301, 10.1126/science.1220234.
- [3] C. Eklöf-Österberg., et. al., *Journal of Phys. Chem. C.* 2019, **123 (4)**, 2019-2030, 10.1021/acs.jpcc.8b08451.