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In-Situ tuning of Catalyst Surface Chemistry for Understanding Proton-Transfer in Bipolar Membranes

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Bipolar membranes (BPMs) provide a platform to isolate and study heterolytic water dissociation (WD) and the reverse H^+/OH^- recombination reactions (H^+/OH^- RC) within a confined ionic heterojunction formed between acidic and alkaline ionomer membranes. Previous work has demonstrated that incorporating nanoparticle catalysts within the heterojunction can reduce the overpotentials (*i.e.,* change in transmembrane bias) to drive WD and H^+/OH^- RC. Various mechanistic hypotheses have been proposed to explain the observed rate enhancement, typically invoking local electric fields and catalyst surface chemistry. However, the fundamental role of the catalyst within the BPM remains unclear, and alternative modalities for interrogating H^+-transfer kinetics in BPMs are thus necessary.

Here, we report a platform for controlling the WD and H^+/OH^- RC catalyst chemistry *in-operando* by polarizing the catalyst layer via an orthogonal circuit and measuring the full-cell (*e.g.,* BPM water electrolyzer or H_2 pump) response. We find that across various metallic and metal-oxide nanoparticle catalysts that the kinetics of H^+ transfer can be tuned substantially via catalyst polarization. The role of surface charging and chemistry are investigated by measuring the *i-E* and impedance response of the full cell as a function of catalyst polarization and temperature. These data provide newfound insights into the chemistry and physics of ion-transfer within the bipolar heterojunction, and more broadly, inform on ion-transfer reactions at a broad range of polarized (electro)chemical interfaces.