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Electric double layer engineering for better electrocatalysis

Electrochemical energy conversion has been recognized as a promising strategy for pursuing a sustainable energy society. In this new energy scenario, electrocatalysis becomes a key technology enabling efficient energy conversion between chemicals and electricity. In the past few decades, significant efforts have been made to improve catalytic performance solely by modification of catalytic materials (*e.g.,* doping, alloying, morphological engineering, etc.), which has successfully led to remarkable advances in initial catalytic activity. Beyond materials engineering, solid-liquid interface engineering, *i.e.,* electrode-electrolyte interfaces (*e.g.,* electric fields, solvents, reactants, pH, cationic species, etc.), has recently been considered for better catalytic performance, but the understanding of the underlying fundamental mechanism is highly missing. Here I will present our recent findings that the nature and activity of ionic species in electrolytes play critical roles in electrocatalysis [1,2]. On the one hand, we demonstrated that the cation-coupled electron transfer (CCET) step, along with two common electron transfer mechanisms in aqueous electrolytes (electron transfer (ET) and proton-coupled electron transfer (PCET)), is available under certain conditions. The CCET mechanism, whose kinetics is affected by cation identity and concentration, governs overall electrocatalysis. On the other hand, we would like to introduce that electrocatalytic stability can also be affected by the chemical properties of the electrode-electrolyte interface. Electrochemical CO2 reduction and Pt dissolution were used here as model reactions to explain the cationic effect.

References

[1] S.-J. Shin *et al.,* On the importance of the electric double layer structure in aqueous electrocatalysis, **Nat. Commun**. 13 (**2022**) 174.

[2] S.-J. Shin *et al.,* A unifying mechanism for cation effect modulating C1 and C2 productions from CO2 electroreduction, **Nat. Commun**. 13 (**2022**) 5482.

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