

### Mechanistic investigation of electrochemical water oxidation

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Water splitting is regarded as a promising step towards environmentally sustainable energy schemes. The oxygen evolution reaction (OER) requires extremely high overpotential due to its slow reaction kinetics. We have been developing a new catalytic platform based on sub-10 nm-sized Mn oxide nanoparticles and detected key intermediate species, Mn(IV)=O, based on electrokinetic and in-situ spectroscopic analysis.

We further conducted electrochemical impedance spectroscopy (EIS) analysis to understand various electrochemical processes in film-type electrocatalysts. From EIS analysis, protons are involved in the electron transport process across the nanoparticle film, which directly proves the ongoing hypothesis of the oxo-hopping mechanism. Also, we focused on the entropic contribution for water oxidation. The activation enthalpy and entropy was measured from temperature dependent analysis. The high negative entropic contribution in heterogeneous water oxidation was confirmed. The entropy effect on OO bond formation by both the AB and RC mechanisms was studied. We anticipate that the unfavorable entropic effect in heterogeneous system could be improved by a hydrogen-bond network.