Electrochemical Reduction of CO2 on IrxRu(1–x)O2(110) Surfaces

High overpotentials and low faradic efficiencies plague metal catalysts for direct conversion of CO2 to methanol and other liquid fuels. RuO2-based electrocatalysts have been observed to evolve methanol at low overpotentials, which has been attributed to an alternative reaction mechanism with oxygen-coordinated intermediates that can circumvent the limitations imposed by the scaling relations on metal catalysts. Here, we introduce an innovative concept of ligand effects in oxide catalysts. Both IrO2 and RuO2 binds OH* and other intermediates from the electrochemical reduction of CO2 (CO2RR) strongly, but the stable and miscible system IrxRu(1–x)O2exhibits anomalous weaker binding energy in the presence of CO* spectators, because of Ru–Ir ligand effects. The weakened adsorbate binding leads to a very low CO2RR onset potential (methanol evolution at −0.2 V RHE). An Ir atom at the bridge site with Ru neighbors binds intermediates such as OH* and OCHO* much weaker, because of synergistic ligand effects and adsorbate–adsorbate interactions.

Consequently, a RuO2 surface doped with Ir move close to the top of the predicted CO2RR volcano for oxides, which offers a significant improvement over state-of-the-art electrocatalysts for conversion of CO2 into methanol. Analysis of electronic structure parameters with adsorbate binding energies indicates the ligand effect depletes electrons from the Ir atom and shifts the t2g orbitals. The lack of electron donation from CO* spectators to Ir at the active site cause favorable adsorbate binding.

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