

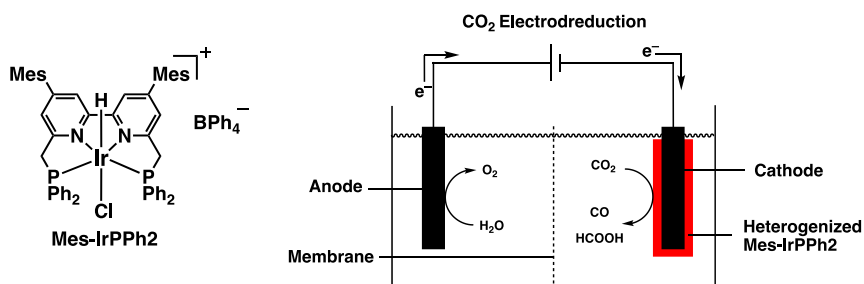
Electrochemical reduction of CO₂ to formic acid at a low overpotential using a heterogenized molecular iridium complex

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The conversion of carbon dioxide (CO₂) into valuable carbon resources through electrochemical and photocatalytic reactions has attracted attention as a promising method to counteract the current environmental and global energy crisis. Although numerous kinds of research on the development of catalysts for CO₂ electrochemical reduction (CO₂ER) have been reported, their performances are yet to be deployed for large-scale applications because of low durability and reusability. Heterogenization of molecular catalysts on electrode surfaces has emerged as a promising approach in terms of smaller catalyst loading, improving electron transfer efficiency, and impeding any deactivation of catalysts by steric confinement effects.¹

Previously in our laboratory, molecular iridium (Ir) complexes bearing PNNP-type tetradentate ligands have been investigated to show superior performance in converting CO₂ to formic acid (HCOOH) with high reactivity and selectivity under photoirradiation in the presence of 1,3-dimethyl-2-phenyl-2,3-dihydro-1*H*-benzo[*d*]imidazole (BIH) as a sacrificial electron donor.² Herein, the Ir complexes were successfully exploited to catalyze CO₂ER in an aqueous solution through heterogenization of the catalyst onto the surface of the carbon electrode. Under optimized conditions, CO₂ER was efficiently facilitated at a very low overpotential (ca. 100 mV) to furnish mainly HCOOH with a current density of 8.4 mA cm⁻² at -0.28 V vs RHE. Employing this cathode catalyst, both high product selectivity (94%) for HCOOH and Faradaic efficiency (> 90%) were achieved in an aqueous solution, where CO₂ER competes in selectivity with H₂ evolution.



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