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Title: Dynamic Transformation of Functionalized Bismuth to Catalytically Active Surfaces for CO₂ Reduction to Formate at High Current Densities

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Abstract: A facile strategy to boost the activity and preserve the selectivity of CO2 reduction to formate using organic-functionalized metal catalysts, specifically 2-methyl-imidazole coordinated to Bi, Bi[2-MeIm], is reported, and the active surfaces are investigated during structural transformation under the applied cathodic potential necessary for CO2 reduction. Operando Raman spectroscopy unveils the structure evolution during the reaction and post-electrolysis analysis shows the formation of three phases of bismuth-based active surfaces. As a result, Bi[2-MeIm] achieves an excellent CO2 reduction performance with an average formate selectivity of approximate to 90% Faradaic efficiency and high activity reaching a current density of up to -1 A cm(-2) in a narrow window of cathodic potentials from -0.46 to -0.78 V versus RHE.

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