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Enhanced Electrochemical CO_2 Reduction to Hydrocarbons on Polybenzimidazole-Modified Copper Electrodes

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Electrochemical reduction of CO_2 to carbon-based fuels and feedstock is one of the most important strategies for mitigating climate change. Metallic copper (Cu) is unique among electrocatalysts, since it produces hydrocarbons and alcohols via electrochemical CO_2 reduction reaction (ECO_2RR). However, Cu electrocatalysts have some drawbacks, such as lack of selectivity, long-term stability, and the generation of hydrogen gas (H_2) through competitive hydrogen evolution reaction (HER). One strategy for improving Cu activity and selectivity towards ECO_2RR is its coating with organic additives. Herein, a facile procedure was used to coat Cu electrocatalysts with polybenzimidazole (PBI). This strategy reduced the HER and increased the activity and stability of the electrode for ECO_2RR. PBI stabilizes the atoms on surface defects through chemical coordination, thereby inhibiting copper's dynamic behavior (dissolution) during operation. The chemical modification of Cu with PBI reduced the HER from 42% to 18% of Faradaic Efficiency (FE) of H_2, while also improving the selectivity towards CH_4 and C_2H_4 on ECO2RR when compared to pristine Cu. We achieved ca. FE = 50% for CH_4 and 15% for C_2H_4 on CuPBI, compared to FE = 37% for CH4 and 8% for C_2H_4 on pristine Cu, at -1.2V vs. RHE. Furthermore, CuPBI is more stable than pure Cu in long-term experiments. After 18 h of ECO2RR, pure Cu retains ~30% of its maximum activity in producing C_2H_4, while CuPBI retains ~66%. Cu maintains ~40% of CH_4 production, while CuPBI maintains ~60% over the same period. These findings offer promising directions for improving the stability of active Cu sites for ECO_2RR, as well as developing more robust electrocatalysts required for effective CO_2 electrolysis in real-world settings.

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