Electronic Modulation of Cu-Pd Catalysts for Improved Ethanol Selectivity in CO_2 Electroreduction

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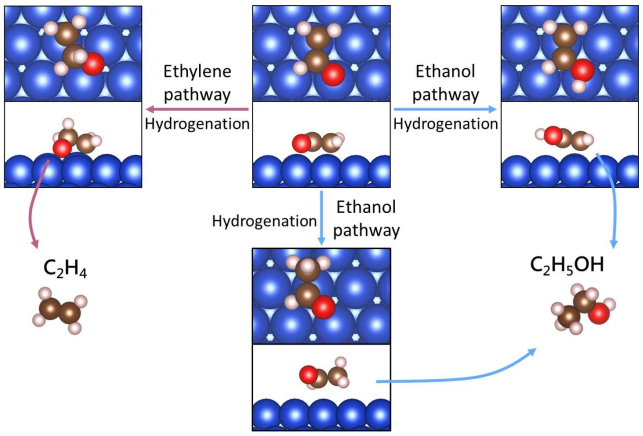
Copper-based catalysts have demonstrated significant potential in electrochemical CO_2 reduction reactions (CO_2RR) for producing valuable C_2 products, including ethano[1]. However, achieving high selectivity toward ethanol remains a challenge due to the competitive formation of ethylene and other byproducts[2]. In this study, we employ theoretical simulations and electronic structure analyses to investigate the selectivity-determining intermediate (*CH_2CHO) on Cu and Cu-Pd alloy catalysts. Density functional theory (DFT) calculations reveal that Pd incorporation into the Cu lattice modulates the electronic states of the intermediate, thereby enhancing ethanol selectivity while suppressing ethylene formatio. Projected density of states (PDOS) and integrated PDOS (IPDOS) analyses indicate that the electronic structure of Cu-Pd alloys stabilizes the *CH_2CHO intermediate in a configuration favorable for protonation at the oxygen site, promoting ethanol formation over ethylene[3].

To compare our theoretical insights, we synthesized $Cu_{1-x}Pd_x$ catalysts using electrochemical pulse deposition. Structural characterizations confirm the formation of a homogeneous Cu-Pd alloy, which effectively modifies the electronic structure and reaction pathway of the CO_2RR process[4]. Electrochemical measurements reveal that the $Cu_{96}Pd_4$ catalyst achieves a Faradaic efficiency of 57% for ethanol production, with a minimal ethylene selectivity of 2.2% at an ethanol partial current density of 228 mA cm^{-2} [5]. These findings highlight the critical role of electronic structure tuning in optimizing CO_2RR selectivity and provide a mechanistic basis for the rational design of Cu-based catalysts for ethanol production.



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