Superior CO_2 Electrochemical Reduction Performance of Cu@AuCu Catalysts

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Abstract

The electrochemical CO_2 reduction reaction (CO_2RR) represents a viable alternative to help close the anthropogenic carbon cycle and convert intermittent electricity from renewable energy sources to chemical energy in the form of value-added chemicals. Development of economic catalysts possessing both high faradaic efficiency (FE) and mass activity toward CO_2RR is critical in accelerating CO_2 utilization technology. In this study, a composite structured Au-Cu catalyst where an alloyed AuCu shell caps on a Cu core (Cu@AuCu) is developed and evaluated for CO_2-to-CO electrochemical conversion. X-ray absorption spectroscopy (XAS) was performed for deeper structural insights of the Au-Cu catalysts, showing that the $\chi_{Au\text{-}Cu}$ value, the hetero-atomic intermix (Cu) around Au atoms dramatically dropped down from 52% to 33%, as the bulk composition varied from Au_1-Cu_9 to Au_3-Cu_7. Overall, Au atoms distribute more uniformly in the shell region at a lower Au content and are able to bond with more Cu atoms around. However, the coordination probability of Au-Au bonds increases at a higher Au content since homogeneity Au atoms prefer to gather, thus, leading to the reduced intermix of Cu around Au atoms. Additionally, the Cu@AuCu catalyst shows a compositional-dependent volcano-plot for selective CO production, which is balanced by the free energy for COOH* formation and CO* desorption. The optimized Au_2-Cu_8 catalyst achieves a FE_{CO} as high as 94% and a superior CO mass activity of 439 mA/mg_{Au} at -0.8 V, particularly, only at an Au content of 17%. Insights gained in this study will not only shed new lights on the structure-property relationship of Au-Cu catalysts, but also benefit the design principles for high-performance CO_2RR catalysts at high efficiency of noble metal utilization.

Keywords - electrochemical CO_2 reduction reaction, core/shell, CuAu, X-ray absorption spectroscopy, faradaic efficiency

References

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