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Electrocatalytic CO₂ Reduction on CuZnAl-based Oxide Catalysts: Tuning of the H₂/CO Ratio

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Electrochemical Reduction of CO₂ (ER-CO₂) is a very attractive alternative to tackle Global Warming¹. Cu-based materials have shown increased yields of hydrocarbon and oxygenate products, while its selectivity towards CO is low². Inspired by the thermocatalytic process, a traditional co-precipitation method was employed to synthesize CuZnAl-based oxide catalysts with a mesoporous structure. This CuZnAl catalyst was tested for the first time for the ER-CO₂ under ambient conditions. The chemicalphysical properties of the catalysts were studied by several characterization techniques (e.g. XRD, XPS, BET, SEM, TEM) and electrochemical impedance spectroscopy at different applied-potentials to understand the role of the modification of the catalyst components during operation in the final selectivity and activity. Results revealed that by adding amphoteric metal oxides like ZnO and Al₂O₃ to the CuObased catalyst contributed to promote CO formation over H₂. XPS measurements on the fresh samples revealed that the ternary CuZnAl catalyst presented a lower percentage (5%) of $Cu^0 + Cu^{1+}$ mixture on the surface than the other catalysts, being mainly constituted by Cu⁺². This material reached a Faradaic efficiency towards syngas of almost 95% at -0.89 V vs RHE. Nevertheless, the highest production rate of syngas was obtained at the most negative applied potential (~ 17 µmol h⁻¹ cm⁻² at -1.14 V vs RHE).). A tunable H₂/CO ratio was achieved by applying different potentials, reaching lower values by increasing applied negative potential (CO current density increased). In fact, a syngas with a H_2 /CO ratio of ~ 2 was obtained at -0.89 V vs RHE, which is a suitable raw material for further methanol synthesis³. The enhanced performance for syngas production of the developed CuZnAl catalyst is demonstrated to be attributed to its surface properties (i.e. alkalinity and the oxidation state on the surface, its lowest diffusional mass transfer resistance, its highest total pore volume and the lowest Cu crystals size among the prepared catalysts.



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