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THE ROLE OF MICROENVIRONMENT ON THE ELECTROCATALYTIC ACTIVITY OF FE-N-C FOR CONVERSION OF CO₂ TO SYNGAS

Resumo: The electrochemical conversion of CO₂ into value-added products, such as hydrocarbons or synthetic fuels, has garnered significant scientific interest. In this context, the exploration of novel electrocatalysts for CO₂ reduction plays a pivotal role in advancing such sustainable energy technology. Single-atom catalysts have demonstrated immense potential for efficiently reducing CO₂ to syngas, benefiting from both their high density of active sites and their intrinsic catalytic activity. This study focuses on investigating the influence of various parameters on the microenvironment of Fe-N-C catalysts during the CO₂ reduction reaction (CO₂RR), in a membrane electrode assembly (MEA) electrolysis cell. Specifically, the impact of temperature, pressure, and catalyst surface modification with ionic liquid (IL) BMIM-PF₆ is evaluated. These variables allow for precise control of the local concentration of H⁺ and CO₂ on the Fe-N-C catalysts, which are the main species governing the extent of CO production. Interestingly, contrary to the typical behavior observed for Ag and Ni-N-C catalysts, increasing the pressure up to 2.5 bar leads to a decrease in Faradaic Efficiency for CO production (FE_{CO}). Additionally, even slight increases in the current density (from 10 to 20 mA cm⁻²) result in a four-fold enhancement in H₂ production. On the contrary, an increase in the cell temperature remarkably enhances the FE_{CO}, which is considered atypical for many CO₂RR catalysts. This observation suggests that additional thermal energy is required to facilitate the adsorption and desorption processes of CO₂ and CO, respectively. Furthermore, the incorporation of Fe-N-C with BMIM-PF₆, results in additional improvement in FE_{CO} as the back-pressure is increased up to 2.5 bar. This behavior suggests that BMIM-PF₆ serves not only as a co-catalyst but also functions as a CO₂ concentrator layer. In summary, results denote that the presence of ionic liquids (ILs) and the control of temperature are important factors for controlling the faradaic efficiency of CO₂ to CO conversion.

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