Carbon-Encapsulated Magnetic Nanoparticles for Magnetocatalytic CO_2 Hydrogenation to CO

Junhui Hu, Walter Leitner, Alexis Bordet
MPI für Chemische Energiekonversion Mülheim an der Ruhr

Abstract

The reverse water–gas shift (rWGS) reaction, which converts CO_2 and H_2 into CO and H_2O, offers a sustainable route to synthesis gas for low-carbon fuel production, such as Fischer–Tropsch synthesis. However, this transformation is equilibrium-limited and requires mild pressures and high temperatures (typically above 600–700 °C) to limit the formation of methane. [1]The development of rWGS catalysts with high activity and CO-selectivity at low temperatures is highly desired, but remains challenging. In the past years, our group has shown that magnetocatalysis (i.e. the activation of magnetically susceptible catalysts via the application of an alternating current magnetic field ACMF) can enable challenging transformations under particularly mild conditions.[2,3] Herein, we hypothesize that the strong temperature gradients generated by the direct and localized ACMF-mediated heating of magnetic catalysts (i.e. hot catalyst in a cold environment) will result in substantial benefits for the conversion of CO_2 to CO, for example by allowing the quick condensation of the H_2O co-product, thereby shifting the position of the thermodynamic equilibrium. Therefore, carbon-coated iron carbide nanoparticles (ICNCs) were immobilized as magnetically-susceptible heating agents on a Cu/Al_2O_3 spinel catalyst to produce a ICNCs@Cu/Al_2O_3 multifunctional catalytic system. Under ACMF (80 mT, 350 kHz) and H_2/CO_2 (5 bar, 3/2 ratio), localized hot spots generated by ICNCs selectively activated the Cu/Al_2O_3 surface at high temperature (~ 300 °C) while maintaining a low bulk reaction temperature (~150 °C)and low pressure(~5bar). This resulted in performances (47 % CO_2 conversion, 90% CO selectivity) superior to what was observed under conventional heating at 150 °C (no reaction) or 300 °C (11.7 % conversion, 99% CO selectivity). Most interestingly, these performances lay substantially above the typical thermodynamic equilibrium given for these reaction conditions, presumably due to the condensation of H_2O. This multifunctional system shows significant potential for low-temperature rWGS reactions, paving the way for sustainable fuel and chemical synthesis. In addition, this is an important demonstration of the potential thermodynamic benefits that the localized activation by magnetocatalysis can bring.

References:

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