PdAg Alloy Nanoparticles Encapsulated in N-doped Microporous Hollow Carbon Spheres for Hydrogenation of CO_2 to Formate

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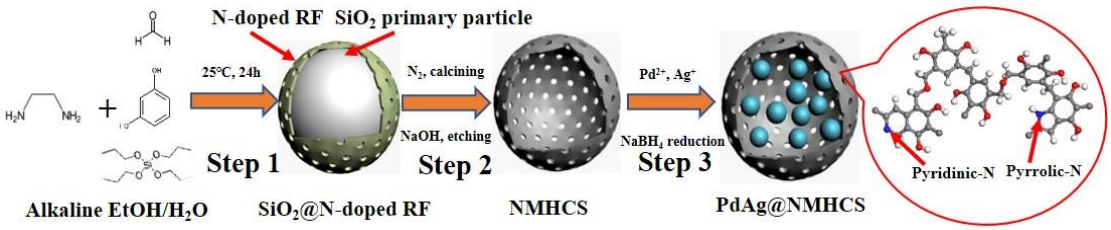
The greenhouse effect is now a global concern. Chemical conversion of CO_2 to beneficial compounds has attracted great interest. Formate has been considered a promising hydrogen storage material because its hydrogen storage capacity is high (53 g L^{-1}). Therefore, the CO_2 hydrogenation catalyst is used to generate formate, which ensures effective CO_2 utilization and achieves economically viable CO_2-mediated hydrogen cycles. Palladium-based heterogeneous catalysts are highly active for the synthesis of formate/formic acid from CO_2.[1,2] Microporous hollow carbon spheres (MHCS) is one of the most ideal carriers for immobilizing metal particles by providing excellent stability. In addition to the porous structure, doped nitrogen as an additional functional group can effectively improve catalytic performance because it can change the polarity and the electron distribution of the carbon matrix.[3]

Herein, we report a general synthesis approach to produce PdAg nanoparticles that are dispersed uniformly within N-doped MHCS (PdAg@NMHCS) as shown in the below Figure for efficient hydrogenation of CO_2 to formate. As a result, thanks to the formation of the ultrafine PdAg alloy nanoparticle size and uniform distribution in the NMHCS, the optimized catalyst shows excellent catalytic activity and good stability in the CO_2 hydrogenation to formate, affording a high turnover number (TON) value of 640 and 2750 at 100 °C after 2 and 24 h, respectively. According to kinetic analyses, NMHCS not only contributed to the adsorption of CO_2 due to the doping of nitrogen but also reduced the activation energy for CO_2 hydrogenation to formate.



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