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Multi-walled Carbon Nanotubes Supported Nickel Nanoparticles Doped with Magnesia and Copper for Adiponitrile Hydrogenation with High Activity and Chemoselectivity under Mild Conditions

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Abstract Multi-walled carbon nanotubes supported nickel nanoparticles doped with magnesia and copper catalysts were prepared by incipient wetness impregnation method and used in adiponitrile (ADN) hydrogenation to 6-aminohexanenitrile (ACN) and 1,6-hexanediamine (HMDA). The prepared catalysts were characterized by X-ray photoelectron spectroscopy (XPS), X-ray diffraction (XRD), transmission electron microscopy (TEM), high resolution transmission electron microscopy (HRTEM), high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM), scanning electron microscopy (SEM) and energy dispersive X-ray (EDX), temperature-programmed hydrogen reduction (H_2-TPR), H_2 chemisorption, temperature-programmed ammonia desorption (NH_3-TPD), temperature-programmed carbon dioxide desorption (CO_2-TPD) and N_2 adsorption-desorption. The results showed that the introduction of magnesia could lead to form NiO-MgO eutectic so as to restrain the reduction of nickel oxide, and it might increase the alkaline site which is conducive to the formation of primary amines in ADN hydrogenation so as to increase the selectivity to ACN and HMDA. Moreover, the formation of NiO-MgO eutectic can also inhibit the sintering of nickel in a certain extent, hence promote the nickel dispersion. And it was revealed that doping of copper can highly promote the catalytic activity by attributing to the strong synergetic effect between copper and nickel which can lead to better dispersion of nickel nanoparticles, larger metallic surface area, lower reduction activation energy of nickel oxide precursor and higher ratio of Ni^{0+} on the surface of the support. Multi-walled carbon nanotubes supported nickel nanoparticles doped with copper and magnesia presents the best catalytic performance of 96.27 % conversion of ADN and 91.22 % selectivity to ACN and HMDA under 2 MPa and lower temperature of 328 K.

Keywords Adiponitrile; Hydrogenation; 6-aminohexanenitrile; 1,6-hexanediamine

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