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研究论文

CeO_2 助 Ni/MgO 催化剂用于化学气相沉积法制备多壁碳纳米管

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摘要 以柠檬酸燃烧法制备的 Ni/MgO, Ni/CeO2-MgO 和 Ni/CeO2 为催化剂, CH4 为碳源, 采用化学气相沉积法制备多壁碳纳米管(MWCNTs), 通过 N2 吸附、X 射线衍射、H2 程序升温还原和 X 射线光电子能谱对催化剂进行表征, 并运用热重和透射电镜表征了碳纳米管的质量和形貌. 结果表明, CeO2 的加入可有效地降低还原温度和增加易还原 Ni 物种的含量, 并使电子发生转移, 还原后的 Ni/CeO2-MgO 催化剂中, Ni 晶粒尺寸较小. 这表明 CeO2 的加入使得 Ni 物种的化学环境发生改变, 导致它和载体间的相互作用减弱, 从而促进 Ni 物种的还原, 且还原后, 高度分散在 CeO2-MgO 载体上, 从而催化剂的催化活性增加. 相比 Ni/MgO 催化剂, Ni/CeO2-MgO 为催化剂上生长的 CNTs 质量更高. 另外, 由 CeO2 助 Ni/MgO 催化剂制备出基本没有无定形碳、结晶度好的碳纳米管.

关键词: 镍 氧化镁 二氧化铈 化学气相沉积法 碳纳米管 甲烷裂解

Abstract: Ni/MgO, Ni/CeO_2-MgO, and Ni/CeO_2 catalysts were prepared by the citric acid combustion method. Their catalytic properties for the synthesis of multi-walled carbon nanotubes (MWCNTs) by chemical vapor deposition using CH_4 as carbon source were evaluated. The catalysts were characterized with N_2 adsorption, X-ray diffraction (XRD), H_2 temperature-programmed reduction (H_2-TPR), and X-ray photoelectron spectroscopy (XPS). The quality and structure of the CNTs were characterized by thermogravimetry (TG) and transmission electron microscopy (TEM). H_2-TPR showed that CeO_2 helped decrease the reduction temperature and increase the content of an easily reducible Ni species. XPS showed an energy shift of the Ni catalyst that was modified by CeO_2. XRD showed that the reduced nickel maintained their small particle sizes in the Ni/CeO_2-MgO catalysts. These findings suggested that the addition of CeO_2 changed the chemical environment of the nickel species, which resulted in a weaker interaction between the nickel and the support and increased the reducibility of the Ni species. Moreover, the reduced nickel was highly dispersed on the CeO_2-MgO support. The change enhanced the catalytic activity for the growth of carbon nanotubes (CNTs). Characterization by TG and TEM showed that the quality of the CNTs grown on Ni/CeO_2-MgO was better than those grown on Ni/MgO catalyst.

Keywords: nickel, magnesium oxide, cerium oxide, chemical vapor deposition method, carbon nanotubes, methane decomposition

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