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研究论文

MgO/h-BN 复合载体对 Ba-Ru/MgO/h-BN 氨合成催化剂性能的影响

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摘要 采用浸渍法制备了系列不同质量比的 MgO/h-BN 复合载体负载的 Ru 基氨合成催化剂, 采用 X 射线衍射、N2 低温物理吸附、X 射线荧光、扫描电镜、透射电镜、程序升温分析等手段对催化剂进行了详细的表征, 并在固定床反应器上考察了它们在氨合成反应中的催化性能. 结果表明, MgO/h-BN 复合载体中 h-BN 含量对催化剂活性的影响较大, Ba-Ru[1:1] (摩尔比)/MgO/h-BN [8:2] (质量比), Ba-Ru[1:1]/MgO/h-BN[6:4]和 Ba-Ru[1:1]/MgO/h-BN[5:5]催化剂上氨合成活性均高于 Ba-Ru/MgO 催化剂. 在 425 ° C, 5.0 MPa, N2/H2 = 1/3 和 5000 h?1 条件下, Ba-Ru[1:1]/MgO/h-BN[8:2]表现出最优催化活性, 达 506.9 ml/(gcat•h). 这可归因于 MgO/h-BN 复合载体上存在较高数量的碱性位, 特别是弱碱性位和中等强度碱性位, 而这些碱性位可能是由 MgO 和 h-BN 之间的相互作用造成.

关键词： 氨合成 钌催化剂 氧化镁 六方氮化硼 碱性

Abstract： A series of MgO/hexagonal boron nitride (h-BN) composite with different mass ratio were synthesized by impregnation method and used as supports for ruthenium catalysts in ammonia synthesis reaction. The catalysts were characterized by X-ray diffraction, N_2 physical adsorption, X-ray fluorescent spectrometer, scanning electron microscope, transmission electron microscope, temperature programmed reduction of H_2, and temperature programmed desorption of CO_2 (CO_2-TPD). The activity measurements of ammonia synthesis were carried out in a fixed-bed flow reactor with a mixture of N_2 and H_2 atomsphere under steady-state conditions (5.0 MPa, 350 – 500 ° C, 5000 h^{-1}). The results showed that the rate of ammonia formation was strongly influenced by the h-BN content used in the catalysts preparation process. The Ba-Ru[1:1] (molar ratio of Ba to Ru = 1:1)/MgO/h-BN[8:2] (weight ratio of MgO to h-BN = 8:2), Ba-Ru[1:1]/MgO/h-BN[6:4] and Ba-Ru[1:1]/MgO/h-BN [5:5] catalysts exhibited higher activity than Ba-promoted Ru/MgO catalyst. At 425 ° C, 5.0 MPa, flow rate of 5000 h^{-1}, and a N_2/H_2 = 1/3 atmosphere, the optimum activity of 506.9 ml/(g_{cat} • h) was achieved when MgO/h-BN[8:2] was used as the catalytic support. The execllent activity was mainly attributed to the basicity of the MgO/h-BN combination-type support, especially “weak” and “medium” basic sites. Large numbers of basic sites were observed from CO_2-TPD characterization due to the interaction between MgO and the h-BN material

Keywords： ammonia synthesis, ruthenium catalyst, magnesia, hexagonal boron nitride, basicity

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