催化学报 » 2012, Vol. 33 » Issue (1) :183-191 DOI: 10.1016/S1872-2067(10)60301-4

纳米 ZSM-5 沸石对芳烃苄基化反应的催化性能

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- 摘要
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摘要 采用晶种有机硅烷化法合成了纳米团簇 ZSM-5 沸石, 考察了其在芳烃苄基化反应中的催化活性. X 射线衍射、N2 吸附和扫描电镜等结果表明, 该沸石为由约 20 nm 的小晶粒聚集成的团簇体, 并形成晶间中孔, 其具有的外表面积是普通 ZSM-5 沸石的 5 倍之多. NH3-程序升温脱附和吡啶吸附的红外光谱结果显示, 沸石的纳米化可使其表面酸中心, 尤其是强酸中心的数目增多, 大大提高了大分子的酸性位可接近性. 在芳烃苄基化反应中, 纳米 ZSM-5 沸石克服了反应分子空间位阻对催化活性的影响, 表现出优异的催化活性, 363 K 时, 甲苯苄基化反应的速率常数是普通 ZSM-5 沸石的约 13 倍.

关键词: 纳米 ZSM-5 沸石 有机硅烷化 芳烃苄基化 酸性 可接近性

Abstract: ZSM-5 zeolite in the form of crystalline nanoclusters was synthesized from seeds functionalized by organosilane and its catalytic performance in the benzylation of aromatic hydrocarbon by benzyl chloride was investigated. The zeolite samples were characterized by X-ray diffraction, N_2 adsorption-desorption, and scanning electron microscopy. The results indicated that they were composed of aggregates of nanocrystals about 20 nm. They had high external surface area that was five times that of conventional ZSM-5. The amounts of acid sites were increased, which was indicated by acidity characterization by the Fourier transform infrared spectroscopy of adsorbed pyridine and temperature-programmed desorption of NH_3. Moreover, accessibility to the acid sites was improved by reducing the size of crystals to nanoscale. In the benzylation of aromatics by benzyl chloride, nanoscale ZSM-5 showed a much higher activity than conventional ZSM-5 because the diffusional limitation for the large reactant was decreased in the nanoscale ZSM-5 zeolite. The benzyl chloride conversion in the benzylation of toluene was 98% at 363 K for 10 h, which was 3.6 times higher than that over conventional ZSM-5. The apparent rate constant over the nanoscale ZSM-5 zeolite was ca. 13 times larger than that over conventional ZSM-5.

Keywords: nanoscale ZSM-5 zeolite, organofunctionalization, aromatic benzylation, acidity, accessibility

收稿日期: 2011-08-02; 出版日期: 2011-12-29

引用本文:

苗海霞, 薛招腾, 马静红等 .纳米 ZSM-5 沸石对芳烃苄基化反应的催化性能[J] 催化学报, 2012,V33(1): 183-191

MIAO Hai-Xia, XUE Zhao-Teng, MA Jing-Hong etc .Nanoscale ZSM-5 Zeolite for the Benzylation of Aromatic Hydrocarbons[J] Chinese Journal of Catalysis, 2012,V33(1): 183-191

链接本文:

http://www.chxb.cn/CN/10.1016/S1872-2067(10)60301-4 或 http://www.chxb.cn/CN/Y2012/V33/I1/183

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