研究论文

脱铝MCM-49分子筛的结构、酸性及苯与丙烯液相烷基化催化性能研究

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摘要 采用硝酸回流和水蒸气两种处理方法对用动态水热法合成的纳米MCM-49分子筛进行脱铝改性. 用XR D、氦气吸附-脱附、NH₃-TPD、FTIR和NMR等技术进行了表征,并考察了脱铝前后MCM-49分子筛在苯与丙烯液相烷基化反应中的催化性能. ²⁷AI核磁共振谱表明,硝酸回流和水蒸气处理能有效地脱除MCM-49分子筛的部分骨架铝和非骨架铝. NH₃-TPD和FTIR表征结果表明,脱铝降低了MCM-49分子筛的Brönsted酸和Lewis酸的酸量. 硝酸脱铝未改变分子筛的酸强度分布,而在600 ℃水蒸气脱铝则造成酸强度的降低. 实验条件下, MCM-49分子筛的丙烯转化率为99.5%,异丙苯的选择性为73.6%. 脱铝改性降低了烷基化反应活性和异丙苯的选择性,提高了收率. 与脱铝前MCM-49分子筛相比,常温下经硝酸处理5 h的脱铝MCM-49分子筛在保持催化活性相当的情况下,异丙苯收率提高了5.3%.

关键词 脱铝MCM-49分子筛 烷基化 催化性能

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Structure, Acid Properties and Catalysis Performance of D ealuminated MCM-49 Zeolites for the Alkylation of Benzen e with Propylene

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Abstract Nano MCM-49 zeolite was synthesized and dealuminated by acid reflux and hydrother mal treatment, respectively. The resulting solids were characterized via XRD, N₂ adsorption-de sorption, NH₃-TPD, FTIR and ²⁷Al NMR. The amounts of total acid sites on dealuminated MCM-49 samples were decreased. For the acid reflux samples, the acid strength is not modified. For the hydrothermally dealuminated zeolite at 600 °C, the acid strength is weaker as observed by NH₃-TPD studies. ²⁷Al NMR reveals that the acid dealumination at room temperature for 5 h results in a decrease of the amounts of aluminum atoms in framework of zeolite. After dealumination under acid reflux for 10 h, the spectrum becomes broader, and the shape of the peak corresponding to the tetrahedral aluminum atoms was modified. After hydrothermal dealumin

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nation at 600 $^{\circ}$ C for 3 h the broadening of the peaks increased and a new peak at δ 30 appeared, due to extraframework aluminum in a tetra- or pentacoordinate. Nano MCM-49 zeolite shows an excellent catalytic activity and selectivity in the liquid-phase alkylation of benzene with propylene. The conversion of propylene reaches 99.5%, and the selectivity to IPB is 73.6%. The propylene conversion can be decreased by dealumination. Comparision of the product distribution between MCM-49 and dealuminated MCM-49 suggests that dealumination could improve the selectivity to monoalkylation. For D-25-5 zeolite, the conversion of propylene is parallel to that of MCM-49 zeolite, while the yield to IPB increases 5.3%.

Key words Dealuminated MCM-49 zeolite Alkylation Catalysis performance

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