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论文

改性纳米ZSM-5催化剂上正辛烷转化反应的研究

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摘要:

以纳米晶粒HZSM-5(20~50 nm)沸石为活性组分, 用碱性介质水热处理、 负载混合稀土和ZnO(或GaO)组合改性方法对纳米HZSM 5分子筛进行改性, 并用TEM, XRF, IR及XRD等手段对催化剂进行表征. 以正辛烷的芳构化和异构化为模型反应, 研究了改性纳米ZSM 5催化剂总酸和酸强度分布、 L/B酸位比例对正辛烷异构化和芳构化反应性能的影响以及催化剂酸强度、 L/B酸位比例与催化剂稳定性和积炭的关系. 结果表明, 碱性介质水热处理和混合稀土改性后, 总酸量减少和酸强度降低导致纳米HZSM-5催化剂的芳构化活性减弱, 异构化活性增强, 稳定性明显提高. 在碱性介质水热处理和负载混合稀土改性的基础上, 再负载适量氧化锌(或氧化镓)改性的催化剂, 总酸量增加, 强酸中心数量减少, B酸略有减少, 而L酸明显增加, L/B酸位比值增加. L酸中心和B酸中心的协同作用和较合适的L/B(1.4~1.7)比值使改性的纳米ZSM-5催化剂保持了较强的和稳定的芳构化和异构化活性, 催化剂积炭失活速率降低. 芳烃和异构烷烃产率分别达到约50%和30%, 高辛烷值的烷基芳烃(C7~C9)和异构烷烃(C4~C6)的选择性分别达到84%和80%.

关键词: 纳米HZSM-5沸石; 正辛烷; 改性; 芳构化; 异构化

Conversion of n-Octane over Modified Nano-crystallite ZSM-5 Catalyst

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Abstract:

By combining alkaline medium hydrothermal treatment with loading mixed rare earth oxide, zinc oxide (or gallium oxide) on nanoscale ZSM-5(20—50 nm) as the active component, respectively, the modified nanoscale ZSM 5 catalysts were prepared, and characterized by TEM, XRF, IR, XRD and so on. The aromatization and isomerization of n-octane were used as the model reactions. The effects of the total acid amount, the acid strength distribution and the molar ratio of L to B acid sites on performance of aromatization and isomerization of the modified nanoscale HZSM-5 catalysts were investigated, and the relationship between acid strength and molar ratio of L to B acid sites and coking was also studied. The results indicate that the decrease of the total acid amount and the reduction of acid strength of the nanoscale HZSM-5 catalysts modified by combining alkaline medium hydrothermal treatment with loading mixed rare earth oxide resulted in wea kening aromatization activity, strengthening isomerization activity and improving stability obviously. On the basis of the alkaline medium hydrothermal treatment and loading mixed rare earth oxide, the acid properties of the nanoscale HZSM-5 catalyst were regulated again after loading proper amount of zinc oxide(or gallium oxide), as a result, the total acid amount of catalysts increased, stronger acid sites of the catalysts decreased, the Br nsted acid sites of the catalyst decreased lightly, the Lewis acid sites of the catalyst increased obviously and the molar ratio of L to B acid sites of the catalyst increased. The proper molar ratio of L to B(1.4—1.7) and the synergistic action between Lewis acid sites and Bronsted acid sites made the catalysts have a stronger and more stable aromatization and isomerization activity, the rate of coking of the catalyst declined. The yields of aromatics and iso paraffins in the products over the combining modification nano scale HZSM-5 catalysts reached about 50% and 30%, respectively, the selectivity of the alkyl-aromatics(mainly C7—C9) and iso-paraffins(mainly C4—C6) with a high Octane Number attained 84% and 80%, respectively.

Keywords: Nanoscale HZSM-5 zeolite; n-Octane; Modification; Aromatization; Isomerization

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