

水热老化对不同方法制备的 Fe-ZSM-5 用于 NH₃ 选择性催化还原 NO_x 的影响

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摘要 采用液体离子交换、等体积浸渍和固相离子交换制备了一系列 Fe-ZSM-5 催化剂, 并将其用于 NH₃ 选择性催化还原 NO_x (NH₃-SCR) 反应. 运用 X 射线衍射、紫外-可见漫反射吸收光谱和原位漫反射傅里叶变换红外光谱对催化剂进行了表征. 结果表明, Fe-ZSM-5 催化剂表面 Fe 物种可分为孤立 Fe³⁺物种、低聚 Fe 氧化物团簇和 Fe₂O₃, 各催化剂上 NH₃-SCR 反应活性不同的根本原因是其表面 Fe 物种分布不同. 水热老化后, Fe-ZSM-5 催化剂上 400 °C 以下反应时 NO_x 转化率降低, 而高温活性略有提高, 各催化剂活性差异减小; 同时 NH₃ 氧化活性和 NO 氧化活性均明显降低. 这可归因于水热老化使 Fe-ZSM-5 催化剂表面的孤立 Fe³⁺物种相对浓度降低, 低聚 Fe 氧化物团簇和 Fe₂O₃ 相对浓度增加, 以及催化剂表面 Brønsted 酸性位显著减少所致.

关键词: 氮氧化物 选择性催化还原 铁 ZSM-5 分子筛 氨 水热老化

Abstract: A series of Fe/ZSM-5 catalysts were prepared by liquid ion exchange, incipient wetness impregnation, and solid-state ion exchange to investigate the selective catalytic reduction (SCR) of NO_x by NH₃ (NH₃-SCR). The effect of hydrothermal deactivation of Fe-ZSM-5 catalysts prepared by different methods as a function of Fe loading was investigated. Freshly made and hydrothermal aged Fe-ZSM-5 catalysts were studied through NH₃-SCR activity test and characterized using X-ray diffraction, UV-Vis diffuse reflectance spectroscopy, and in situ diffuse reflectance infrared Fourier transform spectroscopy. Iron species on the surface of Fe-ZSM-5 catalysts were assigned to isolated Fe³⁺ species, oligomeric Fe_xO_y clusters, and Fe₂O₃ particles based on the UV-Vis spectra. The iron species distributions in the Fe-ZSM-5 catalysts prepared by these methods were quite different, which resulted in difference in SCR activity. The NH₃-SCR activity of different Fe-ZSM-5 catalysts became very similar after aging, and the activity of NH₃ oxidation and NO oxidation decreased with the aged catalysts.

Characterization results indicated that the activity change of the aged Fe-ZSM-5 catalysts was due to the change of iron species distribution in Fe-ZSM-5 catalysts after hydrothermal aging. The relative concentration of isolated Fe³⁺ species was decreased, whereas the relative concentration of oligomeric Fe_xO_y clusters and Fe₂O₃ particles was increased in the aged catalysts. A considerable decrease in the Brønsted acidity of catalysts was observed for the aged Fe-ZSM-5 catalysts.

Keywords: nitrogen oxide, selective catalytic reduction, iron, ZSM-5 zeolite, ammonia, hydrothermal aging

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