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宽工作温度烟气脱硝催化剂制备及反应机理研究

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Preparation of catalyst with wide working-temperature and the reaction mechanism of flue gas denitration

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摘要 以溶胶-凝胶法制备介孔TiO_2载体，采用分步浸渍法制备了V_2O_5-WO_3/TiO_2催化剂，借助BET、NH_3-TPD、H_2-TPR、SEM、活性评价、In-situ FT-IR等手段，考察了催化剂的结构、酸性、还原性、脱硝活性及反应机理等。介孔TiO_2载体比表面积为158.6 m^2/g，制成催化剂后比表面积略有降低，约为136.7 m^2/g。针对模拟烟气在$\varphi_{NH_3}/\varphi_{NO}=0.8$的条件下测试催化剂的脱硝活性温度窗口为250~400 ℃，脱硝转化率达到80%。NH_3-TPD和H_2-TPR表征结果表明，催化剂在活性温度范围内具有典型的表面酸性位，载体TiO_2与V_2O_5之间存在的相互作用使得V_2O_5还原温度降低。利用In-situ FT-IR研究NH_3和NO在V_2O_5-WO_3/TiO_2催化剂表面吸附和氧化的反应过程发现，NH_3可同时吸附在L酸位和B酸位，NH_3在活性位上氧化脱氢形成NH_2物种是SCR脱硝反应的控制步骤。研究NO+O_2+NH_3反应时发现，吸附NH_3的催化剂引入NO和O_2后，共价吸附的NH_3首先消失。选择性催化还原反应发生在吸附态NH_3和气态或弱吸附态的NO之间，该反应遵从Eley-Rideal反应机理。

关键词： 选择性催化还原 脱硝 原位红外光谱 NH_3吸附 反应机理

Abstract: The V_2O_5-WO_3/TiO_2 catalyst using the mesoporous support of TiO_2 made by sol-gel method was prepared with two-step impregnation method and tested for the selective catalytic reduction (SCR) of NO by NH_3. The characterization of the catalyst with BET, NH_3-TPD, H_2-TPR, SEM, activity evaluation and in-situ FT-IR was made to have a deep understanding of the structure, acidity, redox property, catalytic performance, de-NO_x activity and the reaction mechanism. The mesoporous TiO_2 has a surface area of 158.6 m^2/g, and the prepared de-NO_x catalyst has a slightly decreased surface area of 136.7 m^2/g. The V_2O_5-WO_3/TiO_2 catalyst enables the NO conversions to reach to about 80% at 250~400 ℃ and $\varphi_{NH_3}/\varphi_{NO}$ = 0.8, showing the feature of wide working-temperature for the catalyst. The surface adsorption of reactants characterized by in-situ FT-IR shows that NH_3 is adsorbed on both the Lewis and Br nsted acidic sites to generate a few different transformation species. The transformation from NH_3 to NH_2 is the rate-determining step for de-NO_x reaction in NH_3-SCR. It is found that the NH_3-SCR reaction occurs between the adsorbed NH_3 and gaseous NO, which follows the Eley-Rideal reaction mechanism.

Key words: selective catalytic reduction de-NO_x in-situ FT-IR NH_3 adsorption reaction mechanism

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