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研究论文

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催化氧化NO催化剂Mn/ZrO_2的制备与性能研究

Preparation and NO catalytic oxidation activity of Mn/ZrO_2 catalyst

关键词：Mn/ZrO_2催化剂 NO氧化 SCO 反应机理

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摘要：以二氧化锆为载体负载锰氧化物，制备了MnO_x/ZrO_2催化剂用于催化氧化法（SCO）脱除烟气NO，考察了制备条件和反应条件对催化剂SCO活性的影响.同时，采用N_2吸附、SEM、XRD及XPS等手段对催化剂理化性质进行了表征.结果表明，采用等体积浸渍法，以硝酸锰作为前驱体制备的Mn8/ZrO_2/450催化剂在300 ℃、空速15000 h^{-1}条件下，NO转化率可达84%，表现出较好的NO氧化活性.催化剂的主要活性组分为MnO_2，在催化剂载体表面上呈现出较好的分散度.对催化剂进行了NO等温吸附与程序升温脱附，结合程序升温表面反应（TPSR）实验，探讨了催化剂表面NO催化氧化机理.研究发现，NO在该催化剂表面主要以吸附态反应，在含氧气氛中吸附态NO的氧化速率高于脱附速率，表面NO_2的脱附峰温与最佳SCO活性温度相吻合.上述结果表明，NO转化过程速率受氧化产物NO_2的脱附步骤控制，表面反应为快速步骤.

Abstract: A series of ZrO_2-supported manganese oxides catalysts (MnO_x/ZrO_2) were prepared by loading manganese oxides on ZrO_2 supporter for the selective catalytic oxidation (SCO) of flue gas NO to NO_2. The effect of operational parameters on the catalytic conversion of NO was investigated. The structures and properties of catalysts were characterized by N_2 adsorption, scanning electron microscope (SEM), X-Ray Diffraction (XRD) and X-Ray photoelectron spectroscopy (XPS). The MnO_x/ZrO_2 catalyst containing 8% manganese prepared by volumetric impregnation shows the highest catalytic activity for NO conversion at 300 ℃ with space velocity of 15000 h^{-1}. The highly dispersed MnO_2 is proved to be the active species for SCO. The mechanism of catalytic conversion of NO is studied by NO isothermal adsorption and temperature programed desorption (TPD) accompanied by temperature programed surface reaction (TPSR). Results show that the oxidation of NO is induced by NO adsorption and the oxidation rate of NO adsorbed on the catalytic surface is far higher than that of NO_2 desorption. Also the peak temperature of NO_2 desorption is in accordance with the best SCO temperature. Thus the rate controlling step for NO conversion process is the desorption of NO_2 rather than the oxidation of surface NO.

Key words: Mn/ZrO_2 catalysts oxidation of NO SCO reaction mechanism