

SO₂对NO催化氧化过程的影响V.NiO/γ-Al₂O₃上SO₂的作用机理

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摘要 采用程序升温脱附(TPD)及漫反射原位红外光谱(DRIFT)技术分析比较了SO₂存在前后, NO—O₂反应气体在NiO / γ—Al₂O₃催化剂上所形成吸附物种的变化情况, 发现SO₂能促使硝酸盐物种在低温分解并释放出NO₂, 而且耐热稳定的硝酸盐物种也比单纯NO-O₂吸附时多. 室温时催化剂表面上的SO₂以弱吸附物种为主, 特征红外吸收峰位于1324cm⁻¹附近, 温度升高后表面硫酸盐物种数量增多. 关联SO₂气氛中 NO₂的生成规律后得出, 类似于铅室反应中间体的多分子吸附物种[NO₂(SO₃)_x]是产生NO₂的活性物种, 由SO₂在载体或催化剂表面弱碱位吸附后吸引气相NO所产生, 解离O⁻起到稳定活性物种和补充弱碱位的作用. 同时该物种也是毒性物质SO₄²⁻的前驱体, 当KO氧化反应发生后催化剂的失活也开始了.

关键词 [二氧化硫](#) [氧化氮](#) [催化氧化](#) [氧化镍](#) [氧化铝](#) [烟气脱硫](#)

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Effect of SO₂ on NO Catalytic Oxidation V. Mechanism of SO₂ Promotion over NiO/γ-Al₂O₃

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Abstract The influence of SO₂ on the adsorption of NO-O₂ on the surface of NiO/γ-Al₂O₃ was investigated by use of temperature programmed desorption (TPD) and diffuse reflectance infrared Fourier transform spectroscopy (DRIFT). It was found that there are a large quantity of nitro and nitrites as well as monodentate nitrates formed after NO-O₂ adsorption at room temperature, while the bidentate and bridged nitrates are predominant and the desorption of NO₂ increases after NO-O₂-SO₂ adsorption. SO₂ is adsorbed in significant amounts as weakly bonded species, which is characterized by an intense and broad band near 1324 cm⁻¹. Small amounts of sulfates are present on the catalyst surface even at room temperature and grow with increasing adsorption temperature and treatment time. The decomposition of nitrates at low temperatures can account for the release of NO₂ during reaction in the presence of SO₂. A mechanism is proposed to explain the role of SO₂ in NO catalytic oxidation at low temperatures. Several types of multi-molecular intermediate species such as [NO₂(SO₃)_x], the composition of which is similar to the chamber crystals are suggested as active species for producing NO₂. These species are formed by combining NO in the structure of coordinated nitrates with SO₂ adsorbed on either weakly basic O⁻ sites or the basic OH- groups. Adsorbed O₂ species dissociated on neighboring active metal sites such as Ni²⁺ also participate in the formation of the multi-molecular intermediate and are helpful to the stability of the intermediate structures. The continuous adsorption of SO₂ on the freesites, especially on the Lewis acid sites near the intermediate species finally breaks down the intermediate structures. The decomposition of multi-molecular intermediates, which is responsible for the release of NO₂ > however, leads to the sulfation of the catalyst at the same time. As a result, the deactivation of the catalyst occurs.

Key words [SULFUR DIOXIDE](#) [NITROGEN OXIDE](#) [CATALYTIC OXIDATION](#) [NICKEL OXIDE](#) [ALUMINIUM OXIDE](#) [FLUE GAS DESULFURIZATION](#)

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