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Effect of Thermal Ageing of Pd/Al₂O₃ Catalyst on the Selectivity of NH₃ Formation

YAO Li-peng¹, WANG Cheng-xiong^{1,2}, TAN Jian-wei³, REN De-zhi¹, ZHENG Ting-ting², ZHAO Yun-kun^{1,2*}

(1. State Key Laboratory of Advanced Technologies for Comprehensive Utilization of Platinum Metals, Kunming Institute of Precious Metals, Kunming 650106, China;

2. State-Local Joint Engineering Laboratory of Precious Metal Catalytic Technology and Application, Kunming Sino-platinum Metals Catalysts Co. Ltd., Kunming 650106, China;

3. School of Mechanical Engineering, Beijing Institute of Technology, Beijing 100081, China)

Abstract: The effect of thermal aging on the selectivity of NH₃ products in three-way catalytic reaction was studied. Steady-state experiments were carried out in dilute/rich circulating flow. X-ray diffraction (XRD), BET specific surface area, X-ray photoelectron spectroscopy (XPS), Raman spectroscopy (Raman) and transmission electron microscopy (TEM) were used to characterize the physicochemical properties of the catalysts. The results showed that high temperature aging led to a decrease in the dispersion of palladium, an increase in particle size and a decrease in the relative proportion of the Pd²⁺ active species. These microstructural changes contribute to the reduction of the three-way activity of catalysts, resulting in the increase in ammonia selectivity. Thermal ageing induces a significant increase in active palladium particles on the catalyst surface, and larger particles promotes NO to dissociate to form active nitrogen species, which in turn react with hydrogen to form NH₃.

Key words: Pd/Al₂O₃ catalyst; thermal ageing; NH₃ formation; three-way catalytic reactions(TWC); Pd particle

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热老化对 Pd/Al₂O₃ 选择性催化 NH₃ 反应性能的影响

姚丽鹏¹, 王成雄^{1,2}, 谭建伟³, 任德志¹, 郑婷婷², 赵云昆^{1,2*}

(1. 昆明贵金属研究所 稀贵金属综合利用国家重点实验室, 昆明 650106;

2. 昆明贵研催化剂有限责任公司 贵金属催化技术与应用国家地方联合工程实验室, 昆明 650106;

3. 北京理工大学 机械与车辆学院, 北京 100081)

摘 要: 研究了在三效催化反应过程中热老化对 Pd/Al₂O₃ 选择性催化 NH₃ 反应性能的影响。在稀/富循环流中进行稳态实验, 采用 X 射线衍射(XRD)、BET 比表面积、X 射线光电子能谱(XPS)、拉曼光谱(Raman)和透射电镜(TEM)对催化剂的物理化学性质进行表征。结果表明, 高温热老化导致活性金属钯的分散度降低、颗粒尺寸增大、Pd²⁺活性物种的相对比例下降, 这些微结构变化促使催化剂的三效活性降低、氨选择性增加。热老化诱导催化剂表面活性钯颗粒明显增大, 较大的钯颗粒有助于 NO 解离生成活性氮物种, 进而与氢反应生成 NH₃。

关键词: Pd/Al₂O₃ 催化剂; 热老化; NH₃ 生成; 三效催化反应(TWC); Pd 颗粒

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第一作者: 姚丽鹏, 男, 硕士研究生, 研究方向: 贵金属催化剂。E-mail: ylp@163.com

*通讯作者: 赵云昆, 男, 研究员, 研究方向: 汽车尾气后处理技术。E-mail: yk.zhao@spm-catalyst.com