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负载型 V_2O_5/TiO_2 催化剂表面分散状态和性质对氨选择性催化还原 NO 性能的影响

唐富顺 1,2, 庄柯 1, 杨芳 1, 杨利利 1, 许波连 1, 邱金恒 1, 范以宁 1,*

1 南京大学化学化工学院, 介观化学教育部重点实验室, 江苏省纳米技术重点实验室, 江苏南京 210093; 2 桂林理工大学化学与生物工程学院, "应用电化学新技术"广西高校重点实验室, 广西桂林 541004

TANG Fushun1,2, ZHUANG Ke1, YANG Fang1, YANG Lili1, XU Bolian1, QIU Jinheng1, FAN Yining1,*

1Key Laboratory of Mesoscopic Chemistry of Ministry of Education, Jiangsu Provincial Key Laboratory of Nanotechnology, School of Chemistry and Chemical Engineering, Nanjing University, Nanjing 210093, Jiangsu, China; 2College of Chemistry and Bioengineering, Guilin University of Technology, Guilin 541004, Guangxi, China

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摘要 采用多种物理化学手段研究了不同负载量 V_2O_5/TiO_2 催化剂的 VOx 物种分散状态、表面酸性、可还原性及其选择性催化还原 (SCR) NO 性能. 结果表明, V_2O_5 在锐钛矿 TiO_2 表面的实测单层分散容量约为 1.14 mmol V/100 m² TiO_2, 与"嵌入模型"的估算值相符, 表明分散态的钒离子应键合在 TiO_2 表面的八面体空位上. 随着 V_2O_5 负载量的增加, V_2O_5/TiO_2 催化剂上 NO 转化频率 (TOF) 先急剧增加, 至 0.70 mmol V/100 m² TiO_2 (略超过分散容量的一半) 时达到极大 (约 8.3 × 10⁻³ s⁻¹), 然后又急剧下降; 同时, 孤立 VOx 物种可能倾向于分散在相邻的八面体空位上, 且通过 V–O–V 化学键相连形成聚合的 VOx 物种, V–O–V 键所占比例增加而 V–O–Ti 键所占比例减小, 催化剂表面单位钒离子的 Brönsted 酸中心量增加, 故催化剂的 TOF 急剧增加. 随着负载量进一步增加, 虽然催化剂表面单位钒离子的 Brönsted 酸中心量仍缓慢增加, 但 V–O–Ti 键所占比例减少, 导致钒离子的可还原性下降, 另外, 分散容量以上时晶相 V_2O_5 的形成也导致钒离子表面利用率下降, 从而导致催化剂的 TOF 下降. 桥式 Brönsted 酸位 (V–O(H)–V) 也是 SCR 反应活性中心之一, 不同负载量 V_2O_5/TiO_2 催化剂上 SCR 活性与表面 VOx 物种的分散状态、表面酸性和钒离子可还原性密切相关.

关键词： 钒氧物种　分散状态　选择性催化还原　B 酸位　可还原性　氮氧化物

Abstract： The effect of the dispersion state and surface properties of supported vanadia on the selective catalytic reduction (SCR) activity of NO over V_2O_5/TiO_2 catalysts was studied by various experimental techniques. The experimental monolayer dispersion capacity of V_2O_5 on anatase (6.86 VO_x/nm²) measured by XRD was almost the same as the concentration of surface vacant sites of anatase estimated by the incorporation model, and it was suggested that isolated vanadia species tend to be dispersed on adjacent octahedral vacant sites. An increase of the NO turnover frequency (TOF) at 300 °C to a maximum (8.3 × 10⁻³ s⁻¹) at a coverage near half a monolayer was related to the increase of the amount of weak acid sites (Brönsted acid site on each vanadium ion). The TOF decreased rapidly at high VO_x coverages because of a decrease of the reducibility of vanadia species and a decrease of the ratio of exposed vanadia species on the surface. The Brönsted acid sites on bridging V–O(H)–V and terminal V–OH of polymeric vanadia species were all active sites in the SCR reaction. The SCR activity of the V_2O_5/TiO_2 catalysts was related to the dispersion state, acidity, and reducibility of the vanadia species.

Keywords： vanadia species,　dispersion state,　selective catalytic reduction,　Brönsted acid site,　reducibility,　nitrogen oxide

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