

Cu 掺杂对介孔 VO_x-TiO₂ 催化苯羟基化制苯酚的影响

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摘要 将 Cu 作为第二金属, 制备了不同 Cu 掺杂量的和不同温度下焙烧的双金属改性的 Cu/VO_x-TiO₂ 复合催化剂, 并用于液相苯直接羟基化制苯酚反应中. 固定钒的含量为 4.3%, 合成了一系列不同 Cu 掺杂量 ($w = 0.29\% \sim 2.5\%$) 的催化剂, 并在不同的温度下 (350~650 °C) 进行了焙烧. 利用 X 射线衍射、N₂ 吸附-脱附、扫描电镜、透射电镜、H₂ 程序升温还原以及 X 射线光电子能谱对催化剂进行了表征. 结果表明, 加入 Cu 后催化剂仍保持有序的介孔结构, 并且有效地促进了 VO_x 物种在载体 TiO₂ 上的分散和 VO_x 物种的还原, 同时提高了催化剂的热稳定性, 其中 Cu 以 +2 价的形态存在于催化剂中. 另外, 考察了催化剂用量, 反应温度等对苯羟基化反应性能的影响.

关键词: 铜 钒物种 介孔二氧化钛 苯 羟基化 苯酚

Abstract: Liquid phase hydroxylation of benzene to phenol with hydrogen peroxide over VO_x-TiO₂ catalyst samples with Cu as a second metal was investigated. A series of Cu/VO_x-TiO₂ (vanadium loading was 4.3%) catalysts were prepared within the range of Cu loading (0.29% - 2.5%) and calcined at the temperatures of 350 - 650 °C. The catalyst samples were characterized by N₂ adsorption-desorption, scanning electron microscopy, H₂ temperature-programmed reduction, X-ray diffraction, transmission electron microscopy, and X-ray photoelectron spectroscopy. After the addition of Cu, the Cu/VO_x-TiO₂ catalyst had more ordered mesoporous structure as compared with the VO_x-TiO₂ catalyst, and the vanadium was monodispersed on the TiO₂ support. The presence of Cu²⁺ ions on the catalyst surface was shown by XPS measurements. These Cu²⁺ ions probably contributed to the dispersion of vanadium on the surface of the TiO₂ support, and the more facile reduction of VO_x. The Cu²⁺ ions also strengthened the thermostability of the Cu/VO_x-TiO₂ catalyst. The effects of some other variables (Cu loading, catalyst amount, and reaction temperature) on the catalytic performance were also investigated.

Keywords: copper, vanadium species, mesoporous titania, benzene, hydroxylation, phenol

收稿日期: 2012-09-27; 出版日期: 2012-12-24

引用本文:

徐丹, 贾丽华, 郭祥峰. Cu 掺杂对介孔 VO_x-TiO₂ 催化苯羟基化制苯酚的影响[J]. 催化学报, 2013, V34(2): 341-350

XU Dan, JIA Li-Hua, GUO Xiang-Feng. Cu-doped mesoporous VO_x-TiO₂ in catalytic hydroxylation of benzene to phenol[J]. Chinese Journal of Catalysis, 2013, V34(2): 341-350

链接本文:

http://www.chxb.cn/CN/10.1016/S1872-2067(11)60487-7 或 http://www.chxb.cn/CN/Y2013/V34/I2/341

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