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研究论文

交联聚苯乙烯微球固载的双齿席夫碱型氧钒 (Ⅳ) 配合物催化分子氧氧化苯甲醇

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- 摘要
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摘要 利用水杨醛 (SA) 和氨甲基 (MA) 交联聚苯乙烯 (CPS) 微球反应, 制得键合有双齿席夫碱配体 SAAM 的交联聚苯乙烯 SAAM-CPS 微球, 再与硫酸氧钒发生螯合配位反应, 制备了固载有席夫碱型氧钒 (Ⅳ) 配合物催化剂, 采用红外光谱、扫描电镜及热失重等手段对催化剂进行了表征, 并用于催化分子氧氧化苯甲醇反应, 考察了催化剂用量、反应温度和溶剂对反应性能的影响. 结果表明, 通过 SAAM-CPS 微球表面配体与硫酸氧钒之间的螯合配位反应, 可成功制得固载化的席夫碱型氧钒 (Ⅳ) 配合物催化剂; 它在分子氧氧化苯甲醇的反应中表现出很高的催化活性与选择性. 在常压 O_2 和 90 ^oC 的温和条件下, 可高效地将苯甲醇转化为单一产物苯甲醛. 另外, 溶剂极性越弱, 催化剂活性和苯甲醛收率越高, 且具有优良的循环使用性能.

关键词: 交联聚苯乙烯微球 氧钒 (Ⅳ) 配合物 非均相催化 苯甲醇 分子氧 苯甲醛

Abstract: The reaction between salicylaldehyde (SA) and aminomethyl (AM) group-modified crosslinked polystyrene (CPS) microspheres was allowed to be carried out, resulting in the SAAM-CPS microspheres, on which bidentate Schiff base-type oxovanadium(IV) complex was immobilized. Subsequently, the coordination reaction between SAAM-CPS microspheres and vanadyl sulfate was performed, obtaining Schiff base-type oxovanadium(IV) complex immobilized on the CPS-[VO(SAAM)$_2$] microspheres, namely achieving the heterogeneous oxovanadium (IV) complex catalyst. The CPS-[VO(SAAM)$_2$] microspheres were characterized by Fourier transform infrared, scanning electron microscope, and thermal gravimetric analysis. The complex catalyst was used in the oxidation of benzyl alcohol by dioxygen and the catalytic activity was examined. The experimental results show that, through the coordination reaction between SAAM-CPS microspheres and vanadyl sulfate, the immobilized Schiff base-type oxovanadium(IV) complex can be successfully prepared. In the oxidation of benzyl alcohol by dioxygen, the heterogeneous oxovanadium(IV) complex catalyst CPS-[VO(SAAM)$_2$] has very high catalytic activity and excellent selectivity. Under the mild conditions such as at ordinary pressure of dioxygen and at a lower temperature of 90 ^oC, benzyl alcohol can be transformed to benzaldehyde as a single product with yield of 80%. The solvent porlarity affects the catalytic activity of the catalyst greatly. The stronger the solvent polarity, the higher the catalyst activity is and the higher the benzaldehyde yield is. The CPS-[VO(SAAM)$_2$] catalyst has excellent reusability.

Keywords: crosslinked polystyrene microsphere, oxovanadium(IV) complex, heterogeneous catalysis, benzyl alcohol, dioxygen, benzaldehyde

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