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分子印迹聚合物负载纳米金催化剂的制备及其底物识别性能

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摘要 以 4-硝基苯甲醇与氯金酸的络合物为模板, 利用聚合物空腔内胺基捕获 NaBH4 还原的纳米粒子, 设计和制备了一种具有底物识别性能的分子印迹聚合物负载纳米 Au 催化剂 (Au/MIP). 运用红外光谱、紫外-可见光谱和扫描电镜等方法对催化剂进行了表征. 同时以水为溶剂, 过氧化氢为氧化剂, 考察了催化剂在取代苯甲醇氧化反应中的催化性能. 结果表明, 以 Au/MIP 为催化剂时, 4-硝基苯甲醇转化率为 75.6%, 而以非印迹聚合物负载的纳米 Au (Au/NIP) 为催化剂时, 4-硝基苯甲醇转化率仅为 41.5%. 以其它取代苯甲醇为底物时, Au/MIP 与 Au/NIP 的催化活性差别不大. 这说明 Au/MIP 催化剂活性与反应底物结构有关, 脱除模板剂后它具有与 4-硝基苯甲醇相匹配的空腔结构和识别位点, 对反应底物表现出专一的识别性, 因而提高了催化剂活性.

关键词: 分子印迹聚合物 金纳米粒子 取代苯甲醇 催化氧化 底物识别性

Abstract: A molecularly imprinted polymer-supported gold nanoparticle (Au/MIP) catalyst, which has the characteristics of specific substrate recognition, was prepared by the template complex of 4-nitrobenzyl alcohol (4-NBA) and hydrogen tetrachloroaurate(III), where Au nanaoparticles were formed by the reduction with $NaBH_4$ solution and captured by amino groups ($-NH_2$) in the cavities of the MIP. The obtained samples were characterized with FT-IR spectroscopy, UV-Vis spectroscopy, and scanning electron microscopy. The catalytic activity and substrate recognition of the Au/MIP were investigated by the oxidation of substituted benzyl alcohol using H_2O_2 as the oxidant in water. It was found that the conversion of 4-NBA was up to 75.6% over Au/MIP when using the template molecule of 4-NBA as the substrate. However, the conversion of 4-NBA was only 41.5% over non-imprinted polymer-supported gold nanoparticle (Au/NIP) because no template molecule of 4-NBA was used in the preparation of catalyst. Furthermore, no significant difference of the catalytic activity between the catalysts Au/MIP and Au/NIP was observed when other substituted benzyl alcohols were used as the substrate. These results indicated that the catalytic activity of Au/MIP was related to the structure of substrates. The Au/MIP after removal of the template had molecular recognition shape and sites in the cavities matching to the substrate of 4-NBA molecule. The special recognizable cave of the Au/MIP exhibited unique substrate recognition and therefore improved the catalytic activity.

Keywords: molecular imprinted polymer, gold nanoparticle, substituted benzyl alcohol, catalytic oxidation, substrate recognition

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