

杂原子 Sn-β 分子筛的脱铝补位两步法制备、表征及其催化环己酮 Baeyer-Villiger 氧化性能

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摘要 采用酸处理 Al-β 脱铝以产生 T “空位”, 再高温焙烧插入 Sn, 即脱铝补位两步法制备了杂原子 Sn-β 分子筛. 考察了 β 沸石中 T “空位”数量、母体硅铝比和焙烧温度等因素对 Sn-β 分子筛形成与性能的影响, 并利用 X 射线衍射、红外光谱、紫外-可见光谱、扫描电镜、X 射线荧光光谱和电感耦合等离子体原子发射光谱等手段及环己酮 Baeyer-Villiger (B-V) 氧化反应, 对催化剂进行了表征和评价. 结果表明, 脱铝补位两步法可以制备 Sn-β 分子筛, 且 Sn 以四配位形式存在于分子筛骨架中, 在对环己酮 B-V 氧化反应中表现出较高的催化活性.

关键词: 锡 杂原子 β 沸石 环己酮 Baeyer-Villiger 氧化反应 ε-己内酯

Abstract: A Sn-β zeolite was prepared by a two-step postsynthesis method. The procedure consists of first creating vacant T-sites with associated silanol groups by dealumination of Al-β zeolite with nitric acid and then impregnating the resulting Si-β zeolite with an aqueous solution of SnCl₄ · 5H₂O, followed by calcination at varying temperatures. The effects of the amount of vacant T-sites, SiO₂/Al₂O₃ ratio of Al-β zeolite and calcination temperature on the formation of the Sn-β zeolites and their catalytic performances for Baeyer-Villiger oxidation of cyclohexanone were investigated. The structure of the catalysts was characterized by X-ray diffraction, [infrared spectroscopy](#), UV-Vis spectrophotometry, scanning electron microscopy, [X-ray fluorescence spectrometry](#) and inductively coupled plasma atomic emission [spectrometry](#). The results show that, the Sn-β zeolite has been synthesized using a two-step postsynthesis method, and the Sn⁴⁺ ions are incorporated in the framework of the zeolite in a tetrahedral environment. The synthesized Sn-β zeolite shows impressive catalytic performance in Baeyer-Villiger oxidation of cyclohexanone using hydrogen peroxide as an oxidant.

Keywords: tin, heteroatom zeolite β, cyclohexanone, Baeyer-Villiger oxidation, ε-caprolactone

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