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在交联聚苯乙烯微球表面同步合成与固载吡啶基卟啉及固载化钴卟啉的催化氧化性能

田鹏, 高保娇, 陈英军

中北大学化学工程系, 山西太原 030051

TIAN Peng, GAO Baojiao*, CHEN Yingjun

Department of Chemical Engineering, North University of China, Taiyuan 030051, Shanxi, China

- 摘要
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摘要 先通过季铵化反应将吡啶甲醛 (PyAL) 基团键合于交联聚苯乙烯微球 (CPS) 表面, 制得改性微球 PyAL-CPS, 再通过 Adler 反应, 成功地实现了吡啶基卟啉 (PyP) 在 CPS 微球表面的同步合成与固载, 制得功能微球 PyP-CPS, 它再与碘甲烷发生季铵化反应制成 N-甲基吡啶基卟啉 (MPyP) 季铵盐, 从而制得固载有阳离子卟啉的微球 MPyP-CPS; 最后通过与钴盐的配合反应, 制备了固载有阳离子钴卟啉的固体催化剂 CoMPyP-CPS. 考察了影响吡啶基卟啉在 CPS 微球表面同步合成与固载的因素, 固载化阳离子钴卟啉催化剂 CoMPyP-CPS 催化分子氧氧化乙苯的反应性能, 也考察了 CoMPyP-CPS(C) 与磷钨杂多阴离子 (PW) 的复合催化剂 CPW 的催化性能. 结果表明, 以改性微球 PyAL-CPS 与溶液中的吡咯及 4-吡啶甲醛为共反应物, 通过固-液 Adler 反应, 可以顺利地实现吡啶基卟啉在微球 CPS 表面的同步合成与固载, 所制备的固体催化剂 CoMPyP-CPS 具有较好的催化分子氧氧化乙苯反应活性, 而 CPW 催化剂的催化活性更高.

关键词: 交联聚苯乙烯 吡啶基卟啉 固载化 钴卟啉 乙苯 氧化

Abstract: Pyridylaldehyde (PyAL) groups were bound onto the surface of crosslinked polystyrene (CPS) microspheres via quaternization between 4-pyridylaldehyde and chloromethyl groups of chloromethylated crosslinked polystyrene (CMCPS) microspheres, obtaining the modified microspheres PyAL-CPS. Subsequently, synchronous synthesis and immobilization of pyridylporphyrin on CPS microspheres were successfully realized using 4-pyridylaldehyde, pyrrole and the modified microspheres PyAL-CPS as the co-reactants via the Adler reaction between solid-liquid phases, resulting in functional microspheres PyP-CPS, on which pyridylporphyrin (PyP) was immobilized. The microspheres PyP-CPS were then allowed to react with methyl iodide, and PyP was transformed to N-methyl pyridyl porphyrin (MPyP) iodide, leading to the formation of MPyP-CPS microspheres, on which cationic porphyrin was immobilized. Finally, the solid catalyst CoMPyP-CPS, on which cationic cobalt porphyrin was immobilized, was prepared through the coordination reaction between MPyP-CPS microspheres and cobalt salt. The effects of the main factors on the synchronous synthesis and immobilization of pyridylporphyrin on CPS microspheres were examined, and the catalytic performance of the solid catalyst CoMPyP-CPS in the oxidation of ethylbenzene by molecular oxygen was investigated mainly. Besides, the catalytic character of the composite catalyst CPW, which was prepared by association of CoMPyP-CPS (C) and phospho-tungstic(PW)heteropoly acid, was also investigated. The results show that the synchronous synthesis and immobilization of pyridylporphyrin on CPS microspheres can be favourably carried out using PyAL-CPS microspheres, 4-pyridylaldehyde, and pyrrole in the solution as co-reactants via the Adler reaction between solid-liquid phases. The immobilized cationic cobaltporphyrin catalyst CoMPyP-CPS has excellent catalytic performance in the oxidation of ethylbenzene by molecular oxygen, and the composite catalyst CPW possesses much higher catalytic activity.

Keywords: crosslinked polystyrene, pyridylporphyrin, immobilization, cobalt porphyrin, ethylbenzene, oxidation

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