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研究论文

双模板法制备具有介孔孔壁的三维有序大孔二氧化铈及其改善的低温还原性能

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- 摘要
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摘要 以聚甲基丙烯酸甲酯 (PMMA) 为硬模板, 三嵌段共聚物 F127、十六烷基三甲基溴化铵 (CTAB) 或聚乙二醇 (PEG) 为软模板剂 (表面活性剂), 柠檬酸为络合剂, 硝酸铈为金属前驱体, 采用双模板法成功地合成出具有介孔孔壁的三维有序大孔 (3DOM) 结构的立方相 CeO_2 样品 CeO_2-F127, CeO_2-CTAB 和 CeO_2-PEG, 并利用多种分析技术表征了它们的物化性质. 结果表明, 三个样品均具有 3DOM 结构和蠕虫状介孔孔壁, 表面活性剂的种类对样品的孔结构和比表面积影响较大. 在制备过程中引入表面活性剂可提高 CeO_2 样品的比表面积, 所得 CeO_2-F127, CeO_2-CTAB 和 CeO_2-PEG 样品的比表面积分别为 60.5, 60.2 和 51.8 m^2/g. 具有 3DOM 结构的 CeO_2 样品的低温还原性显著好于无孔的体相 CeO_2, 它们的低温还原性按照 CeO_2-PEG < CeO_2-CTAB < CeO_2-F127 的顺序提高, 与其表面氧空位密度大小的顺序相吻合. CeO_2 因具有蠕虫状介孔孔壁的 3DOM 结构, 改善了其物化性质, 使此类材料在催化方面有着更广泛的应用前景.

关键词: 双模板法 三维有序大孔 二氧化铈 低温还原性能 蠕虫状介孔孔壁 表面活性剂

Abstract: Three-dimensionally ordered macroporous (3DOM) ceria with mesoporous walls and cubic crystal structures were prepared with polymethyl methacrylate (PMMA) as a hard template and triblock copolymer Pluronic F127 ($EO_{106}PO_{70}EO_{106}$), cetyltrimethylammonium bromide (CTAB), or poly(ethylene glycol) (PEG) as a soft template. Citric acid was used as a complexing agent and cerium nitrate was used as a metal precursor. The 3DOM CeO_2 samples were characterized by numerous analytical techniques. The as-fabricated CeO_2 samples had a 3DOM architecture with polycrystalline wormhole-like mesoporous walls. The nature of the soft template had an important effect on the pore structure and the surface area of the final product. The surface areas of the F127-, CTAB-, and PEG-derived 3DOM CeO_2 samples (denoted CeO_2-F127, CeO_2-CTAB, and CeO_2-PEG, respectively) were ca. 60.5, 60.2, and 51.8 m^2/g, respectively. The low-temperature reducibility of the 3DOM-structured CeO_2 samples was much better than that of the bulk counterpart and the low-temperature reducibility of the three 3DOM ceria samples increased according to: CeO_2-PEG < CeO_2-CTAB < CeO_2-F127, which coincided with the surface oxygen vacancy density sequence. The improved physicochemical properties associated with the formation of the 3DOM skeleton with wormhole-like mesoporous walls may be useful for applications such as CeO_2 materials in heterogeneous catalysis.

Keywords: dual-templating preparation method, three-dimensionally ordered macropore, ceria, low-temperature reducibility, wormhole-like mesoporous wall, surfactant

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