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研究论文

模板沉淀法制备高比表面积MnO_x-CeO_2催化剂及其CO低温氧化活性

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摘要 以十六烷基三甲基溴化铵(CTAB)为模板剂, $Ce(NO_3)_3$和$Mn(NO_3)_2$为前驱体, 通过沉淀法制备了一系列晶粒小于5 nm的高比表面积MnO_x-CeO_2催化剂, 并考察了催化剂的CO氧化反应性能. 采用XRD、Raman光谱、TPR和N_2气吸附脱附等手段对催化剂的比表面积、晶粒大小和物相组成进行了表征. 当Mn摩尔分数≤34%时, 催化剂的比表面积在160~170 m^2/g之间; 当锰含量进一步提高后, 催化剂的比表面积呈下降趋势. 当Mn摩尔分数≤34%时, XRD只检测到CeO_2物相, 而Raman光谱则检测到α-Mn_2O_3的存在. 催化剂上表现出较好的CO氧化活性, 这主要归因于高比表面积. 随着锰含量的增加, 催化剂的轻化频率(TOF)下降, 表明高分散、小晶粒的氧化锰物种是催化剂的活性物种. H_2-TPR结果表明, 催化剂的CO氧化活性还与催化剂中高价锰物种有关. 焙烧温度升高使催化剂的晶粒增大、比表面积减小, 同时催化剂中锰的平均价态降低, 导致CO氧化活性下降.

关键词 MnO_x-CeO_2 十六烷基三甲基溴化铵(CTAB) 高比表面积 CO氧化

分类号 0643

Preparation of Highly-specific Surface Area MnO_x-CeO_2 Catalysts by a Template Precipitation Method and Its Performance for Low Temperature CO Oxidation

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Abstract Nanosized MnO_x-CeO_2 catalysts with a high-surface area were prepared by a template precipitation method, with $Ce(NO_3)_3$ and $Mn(NO_3)_2$ as the precursors and surfactant CTAB as the templating agent. Their catalytic activities for CO oxidation were examined. The catalysts were characterized by XRD, Raman spectroscopy, H_2-TPR and N_2 adsorption techniques. BET specific surface areas of the catalysts were between 160 and 170 m^2/g when the Mn mass fraction was lower than 34.3%. However, surface area decreased with further increasing Mn mass fraction. XRD results show that there was only CeO_2 phase when the Mn mass fraction was lower than 34.3%, while Raman spectra indicate the existance of α-Mn_2O_3. The high specific surface area was benefical to CO oxidation activity. TOF of the catalysts decreased with increasing Mn content, indicating that the highly dispersed Mn species with a small crystallite size were the active species. H_2-TPR results suggest that the activitiy was related to the high-valance Mn species. High calcination temperature caused growth of the catalyst crystallite size and decrease of surface areas, as well as decline of avarage valance of Mn species, which led to decline in the catalytic activity.

Key words MnO_x-CeO_2 CTAB Highly-specific surface area CO oxidation

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