

论文

Ce_{1-x}Tb_xO_{2-δ}复合氧化物氧缺位的拉曼光谱研究

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摘要:

采用溶胶-凝胶法制备了Ce_{1-x}Tb_xO_{2-δ}复合氧化物, 利用不同Raman激发波长(514和785 nm), 结合X射线衍射(XRD)、氢气-程序升温还原(H₂-TPR)和氧气-程序升温脱附(O₂-TPD)表征, 考察了Ce_{1-x}Tb_xO_{2-δ}复合氧化物在O₂, He和H₂气氛下氧缺位的原位变化情况和CeO₂的F_{2g}特征Raman峰位的偏移. 实验结果表明, 随着Tb掺杂量的提高, 由于晶胞收缩使得CeO₂的F_{2g}特征Raman振动峰发生蓝移. 514 nm Raman激发波长反映了催化剂的表面信息, 而785 nm激发波长反映了整体信息. 正是由于表面和整体变化的不一致, 造成原位Raman实验过程中氧缺位浓度变化趋势的不同. 在He和H₂气氛下, 由于温度升高时伴随着Ce_{1-x}Tb_xO_{2-δ}中O₂气的脱出, 使复合氧化物的微观结构发生改变, 以致Ce_{0.9}Tb_{0.1}O_{2-δ}中的氧缺位浓度(A₅₈₇/A₄₆₅)在785 nm激发波长下出现先升高后下降的现象.

关键词: 原位拉曼光谱; Ce_{1-x}Tb_xO_{2-δ}复合氧化物; 氧缺位

Raman Spectroscopic Study of Oxygen Vacancies in Ce_{1-x}Tb_xO_{2-δ} Mixed Oxides

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Abstract:

A series of Ce_{1-x}Tb_xO_{2-δ} mixed oxides were prepared by a sol-gel method. In situ Raman spectroscopy combined with X-ray diffraction(XRD), H₂-temperature programmed reduction(H₂-TPR) and O₂-temperature programmed desorption(O₂-TPD) were used to analyze oxygen vacancies in the mixed oxides under different atmospheres(O₂, He, and H₂) with 514 and 785 nm excitation laser lines and the blue shift of the F_{2g} vibration mode of CeO₂. The results show that increasing Tb content in the sample result in the blue shift of F_{2g} vibration mode of CeO₂ because of dominant effect of the shrinking of the crystal. Furthermore, the difference in the changing trend of oxygen vacancies is due to the different Raman excitation laser lines. 514 nm excitation laser line can provide the surface information, while 785 nm excitation laser line can get the whole information of the sample. The A₅₈₇/A₄₆₅ ratio reflecting the oxygen vacancies shows an inflexion under He and H₂ atmospheres could be explained by the change in microstructure of the sample along with the release of O₂ during the heating process.

Keywords: *In situ* Raman spectroscopy; Ce_{1-x}Tb_xO_{2-δ} mixed oxide; Oxygen vacancy

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