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研究论文

钙改性的 Pd/CeO_2-ZrO_2-Al_2O_3 催化剂催化甲醇裂解反应

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摘要 采用共沉淀法制备了未改性的和 Ca 掺杂的 CeO_2-ZrO_2-Al_2O_3 样品, 进一步用浸渍法制备了 Pd/CeO_2-ZrO_2-Al_2O_3 (Pd/CZA) 和 Pd/CeO_2-ZrO_2-Al_2O_3-CaO (Pd/CZACa) 催化剂. 运用 X 射线衍射、N_2 吸附-脱附、储氧量测定、CO 化学吸附、NH3 程序升温脱附、CO_2 程序升温脱附、H2 程序升温还原和 X 射线光电子能谱对催化剂进行了表征, 并考察了其催化甲醇裂解反应活性. 结果表明, Ca 的添加使载体酸性位中毒, 弱碱中心数目增加, 从而影响了催化剂上吸附物种的吸附-脱附平衡过程; 同时, 使得金属和载体间相互作用增强, 增加了 Pd 周围的电子密度, 使 Pd 保持在部分氧化状态 $Pd^{\delta+}$ ($0 < \delta < 2$), 进而提高了甲醇催化裂解反应的活性, 使甲醇完全裂解温度降低了 34 ° C.

关键词: 钯 钙 甲醇裂解 氧化铈 氧化锆 氧化铝

Abstract: CeO_2-ZrO_2-Al_2O_3 and CeO_2-ZrO_2-Al_2O_3-CaO supports were prepared by the coprecipitation method and were loaded with Pd by impregnation to form Pd/CeO_2-ZrO_2-Al_2O_3 (Pd/CZA) and Pd/CeO_2-ZrO_2-Al_2O_3-CaO (Pd/CZACa) catalysts. The catalysts were characterized by X-ray diffraction, low temperature N_2 adsorption-desorption, oxygen storage capacity, CO chemisorption, NH_3 temperature-programmed desorption (NH_3-TPD), CO_2 temperature-programmed desorption (CO_2-TPD), H_2 temperature-programmed reduction (H_2-TPR), and X-ray photoelectron spectroscopy (XPS). The catalyst activity for methanol decomposition showed that the Ca modification improved the low-temperature activity of the catalyst, which lowered the complete conversion temperature on 34 °C. NH_3-TPD and CO_2-TPD showed that the addition of Ca poisoned the acid sites of the support or increased the number of weak basic sites and therefore changed the adsorption-desorption equilibria of the adsorbed species. It also enhanced the metal-support interaction and increased the electronic surroundings of Pd sites, which maintained Pd in a partly oxidized ($Pd^{\delta+}$) state and consequently increased the activity for methanol decomposition according to H_2-TPR and XPS measurements.

Keywords: palladium, calcium, methanol decomposition, ceria, zirconia, alumina

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