催化学报 » 2012, Vol. 33 » Issue (6) :1014-1019 DOI: 10.3724/SP.J.1088.2012.11242



# $(CeO_2-ZrO_2-Al_2O_3)-(La_2O_3-Al_2O_3)$ 复合氧化物负载的 Pd 密偶催化剂: 载体焙烧温度的影响

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摘要 采用同时共沉淀法制备了 $(CeO_2-ZrO_2-Al_2O_3)-(La_2O_3-Al_2O_3)$ 新型复合氧化物 (CZA-LA), 考察了焙烧温度对 CZA-LA 负载的 Pd 密偶催化剂的影响, 并采用 N2 吸附-脱附法和 X 射线衍射对其进行了表征. 结果表明, 随着焙烧温度的升高, 尽管 CZA-LA 样品的比表面积降低, 但即使在 1000 ° C 焙烧 5 h 后, 其比表面积仍能保持在 122 $m^2/g$ 左右; 另外, CZA-LA 样品的孔体积降低幅度不大, 当在 700 ° C 及更高温度下焙烧后基本上保持稳定. 以不同温度焙烧 5 h 后的样品为载体, 采用等体积浸渍法制备了一系列整体式 Pd/CZA-LA 密偶催化剂 (Pd 含量 2.0 g/L). 催化剂对消除模拟汽油车尾气中 C3H8 活性测试结果表明, 各新鲜催化剂的活性相差不大, 均具有较低的起燃温度 T50 和完全转化温度 $T_{90}$. 当催化剂经老化处理后, 以 1000 ° C 焙烧的载体负载的 Pd 催化剂活性最高, $T_{50}$ 和 $T_{90}$ 分别为 310 和 341 ° C, ΔT 仅为 31 ° C, 仅比新鲜催化剂的高 7 ° C. 可见, 该催化剂表现出优异的催化活性和抗老化性能, 可以满足欧IV及更高标准的用于汽油车尾气净化的密偶催化剂的性能要求.

关键词: 钯 密偶催化剂 同时共沉淀 丙烷 热稳定性 汽车尾气净化 焙烧温度

Abstract: A new $(CeO_2-ZrO_2-Al_2O_3)-(La_2O_3-Al_2O_3)$ complex oxide (CZA-LA) was prepared by the simultaneous co-precipitation method and then it was calcined at different temperatures. The samples calcined at different temperatures were used as support to prepare Pd/CZA-LA catalysts by impregnation and they were further fabricated as close-coupled catalysts in monolith form. The Pd loading of close-coupled catalysts was 2 g/L. The Pd/CZA-LA samples were characterized by $N_2$ adsorption-desorption and X-ray diffraction, and the corresponding catalyst was examined by temperature-programmed reduction of $H_2$ and $C_3H_8$ conversion in a simulated automobile exhaust. It was found that both textural properties of the CZA-LA and catalytic performance of the Pd/CZA-LA were remarkably affected by the calcination temperature of CZA-LA support. The new complex oxide samples showed good structural and textural properties and were more resistant to aging at high temperature. The surface area of CZA-LA support decreased with increasing calcination temperature; however, it was still 122 $m^2/g$ for S1000 (noted as the CZA-LA support calcined at 1000 ° C for 5 h) sample. The pore volume was stable for all of the samples except S600 sample. The catalytic evaluation results showed that the catalytic performance of all fresh catalysts was very good and similar to each other. The light-off temperature ($T_{50}$ = 310 ° C) and complete conversion temperature ($T_{90}$ = 341 ° C) of propane over the Pd/S1000-Aged (noted as the Pd/S1000 catalyst aged at 1000 ° C for 5 h) was the lowest among the tested aged catalysts, and the temperature difference, ΔT, between the Pd/S1000-Aged and Pd/S1000-Fresh was as less as 7 ° C, indicating that the Pd/S1000-Fresh catalyst showed good catalytic performance for exhaust purification and good resistance to aging. The catalyst may have potential application in the field of advanced Pd close-coupled catalysts.

Keywords: palladium, close-coupled catalyst, simultaneous co-precipitation, propane, thermal stability, automotive exhaust purification, calcination temperature

收稿日期: 2011-12-16; 出版日期: 2012-05-10

引用本文:
方瑞梅, 何胜楠, 崔亚娟等 .$(CeO_2-ZrO_2-Al_2O_3)-(La_2O_3-Al_2O_3)$ 复合氧化物负载的 Pd 密偶催化剂: 载体焙烧温度的影响[J] 催化学报, 2012,V33(6): 1014-1019

FANG Rui-Mei, HE Sheng-Nan, CUI Ya-Juan etc .$(CeO_2-ZrO_2-Al_2O_3)-(La_2O_3-Al_2O_3)$ Complex Oxide Supported Pd Close-Coupled Catalysts: Effects of Support Calcination Temperature[J] Chinese Journal of Catalysis, 2012,V33(6): 1014-1019

链接本文:
http://www.chxb.cn/CN/10.3724/SP.J.1088.2012.11242 或 http://www.chxb.cn/CN/Y2012/V33/I6/1014



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