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研究论文

MnO_x 负载量对 $MnO_x/Ce_{0.7}Zr_{0.2}La_{0.1}O_2$-$Al_2O_3$ 催化剂上碳烟燃烧性能的影响

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摘要 采用等体积浸渍法制备了一系列不同 MnO_x 含量的 $MnO_x/Ce_{0.7}Zr_{0.2}La_{0.1}O_2$-$Al_2O_3$ ($Ce_{0.7}Zr_{0.2}La_{0.1}O_2$ /Al_2O_3 质量比 = 1) 催化剂, 并用 X 射线衍射、低温 N2 吸附-脱附、X 射线光电子能谱、O_2 程序升温脱附和 H_2 程序升温还原等手段对催化剂进行了表征, 考察了催化剂催化柴油车排放碳烟颗粒物燃烧的反应性能. 结果表明, 催化剂表面吸附的活性氧物种和 MnO_x 的低温区还原性能是决定催化剂活性的两大关键因素. 当 MnO_x 负载量为 5% 时, 催化反应所需的活性氧减少, 因而活性降低; 但 MnO_x 负载量增至 10% 时, 催化剂中 Mn 物种的可还原量提高, 从而增加其活性; 增至 20% 时, MnO_x 与表面吸附氧物种的可还原量间达平衡最佳值, 活性最佳, 碳烟起燃温度比无催化剂时降低了 179 ^oC; 负载量达 30% 后, 由于载体表面吸附氧物种数量的降低和还原峰温的上升使催化剂活性下降.

关键词: 锰氧化物 负载量 碳烟 催化燃烧 氧化铈 氧化锆 氧化镧 氧化铝

Abstract: A series of $MnO_x/Ce_{0.7}Zr_{0.2}La_{0.1}O_2$-$Al_2O_3$ supported catalysts with the $Ce_{0.7}Zr_{0.2}La_{0.1}O_2$:$Al_2O_3$ mass ratio of 1:1 and different MnO_x loadings were prepared by the incipient wetness method. The catalysts were characterized by X-ray diffraction, low temperature N_2 adsorption-desorption, X-ray photoelectron spectroscopy, O_2 temperature-programmed desorption, and H_2 temperature-programmed reduction. The catalytic performance of these catalysts for the combustion of diesel soot was investigated. It is found that surface-adsorbed active oxygen species and low-temperature reducibility of MnO_x are the determinants of catalytic activity. When the MnO_x loading is 5%, the catalyst activity decreases owing to the loss of active oxygen species, which are necessary for the catalytic combustion. When the MnO_x loading is increased to 10%, the catalyst activity is dramatically increased because of the enhanced reducible manganese species. Interestingly, the optimal values for reducible manganese species and surface-adsorbed oxygen species can be achieved in the catalyst with 20% MnO_x, and so the catalyst exhibits the best catalytic activity, giving a light-off temperature about 179 ^oC lower than that of the non-catalytic soot combustion. With a further addition of MnO_x species up to 30%, its catalytic activity is deteriorated mainly due to the decrease in surface-trapped oxygen species and upper shift of the reduction temperature.

Keywords: manganese oxide, loading amount, soot, catalytic combustion, ceria, zirconia, lanthana, alumina

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