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研究论文

NOx Reduction on Fully Formulated Lean NOx Trap Catalysts Subjected to Simulated Road Aging: Insights from Steady-State Experiments

Jin WANG, Yaying JI, Uschi GRAHAM, Caio CESAR SPINDOLA DE OLIVEIRA, Mark CROCKER*

Center for Applied Energy Research, University of Kentucky, Lexington, KY 40511, USA

Jin WANG, Yaying JI, Uschi GRAHAM, Caio CESAR SPINDOLA DE OLIVEIRA, Mark CROCKER*

Center for Applied Energy Research, University of Kentucky, Lexington, KY 40511, USA

摘要 Fully formulated lean NO_x trap (LNT) catalysts of the type $Pt/Rh/BaO/Al_2O_3$ were prepared with and without incorporation of CeO_2-ZrO_2 in the washcoat, and their NO_x reduction behavior was evaluated in steady-state, continuous flow experiments. In the fresh state, CeO_2-ZrO_2 addition was found to exert little effect on NO_x reduction activity using H_2, CO, and NH_3 as the reductants. However, after simulated road aging, NO_x reduction activity was significantly impaired for the CeO_2-ZrO_2-free catalyst, whereas the performance of the CeO_2-ZrO_2-containing analog was affected to only a minor degree. These differences are explained on the basis of high-resolution transmission electron microscopy measurements showing that Pt supported on CeO_2-ZrO_2 remained highly dispersed after aging, whereas Pt supported on BaO/Al_2O_3 underwent significant sintering. In addition, the Pt/CeO_2-ZrO_2 component did not accumulate sulfur during aging, unlike $Pt/BaO/Al_2O_3$ for which significant sulfation of the Ba phase occurred. For both catalysts, selectivity to NH_3 in NO and NO_2 reduction by H_2 increased after catalyst aging, indicative of a change in the relative surface coverages of N and H ad-atoms on the precious metal sites.

关键词： lean NOx trap NOx storage-reduction catalyst steady state NOx reduction ceria-zirconia catalyst aging

Abstract： Fully formulated lean NO_x trap (LNT) catalysts of the type $Pt/Rh/BaO/Al_2O_3$ were prepared with and without incorporation of CeO_2-ZrO_2 in the washcoat, and their NO_x reduction behavior was evaluated in steady-state, continuous flow experiments. In the fresh state, CeO_2-ZrO_2 addition was found to exert little effect on NO_x reduction activity using H_2, CO, and NH_3 as the reductants. However, after simulated road aging, NO_x reduction activity was significantly impaired for the CeO_2-ZrO_2-free catalyst, whereas the performance of the CeO_2-ZrO_2-containing analog was affected to only a minor degree. These differences are explained on the basis of high-resolution transmission electron microscopy measurements showing that Pt supported on CeO_2-ZrO_2 remained highly dispersed after aging, whereas Pt supported on BaO/Al_2O_3 underwent significant sintering. In addition, the Pt/CeO_2-ZrO_2 component did not accumulate sulfur during aging, unlike $Pt/BaO/Al_2O_3$ for which significant sulfation of the Ba phase occurred. For both catalysts, selectivity to NH_3 in NO and NO_2 reduction by H_2 increased after catalyst aging, indicative of a change in the relative surface coverages of N and H ad-atoms on the precious metal sites.

Keywords： lean NOx trap, NOx storage-reduction catalyst, steady state, NOx reduction, ceria-zirconia, catalyst aging

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