216c Steam Effect on No_X Reduction over Pt-Bao/Al_2O_3 Catalyst

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Pt-BaO/Al_2O_3 is a lean NO_x trap (LNT) model catalyst that provides a very effective method for NO_x removal in lean-burn gasoline engine exhaust. For a typical cyclic operation, NO is oxidized to NO_2 on the Pt sites, and the NO_2 is then being trapped by the BaO to form barium nitrate during the lean cycle. During the rich cycle, the nitrate decomposes and the released NO_x is reduced on the Pt sites into N_2. Many studies have been carried out on catalytic reduction of NO by H_2, CO, and by a mixture of H_2/CO over supported Pt group metal catalysts. With H_2 as reductant, NH_3 or HCN is the major reaction product. Steam effects in three-way catalysts have also been widely investigated, and recently the formation of NH_3 was reported for the reduction of NO by CO in the presence of water vapor on Al_2O_3-supported Pt catalyst. Although NH_3 is currently not an emission component regulated by EPA, it is still interesting to study in more detail the catalytic formation of NH_3 and its influence on the NO_x reduction for automotive applications, especially in view of potential use of H_2 in future H_2 internal combustion engines (ICE).

In this work, we investigated the decomposition characteristics of Pt-$Ba(NO_3)_2/Al_2O_3$ by TGA-IR, and the lean/rich (L/R) cycle reaction characteristics of a Pt-BaO/Al_2O_3 LNT model catalyst, focusing on the formation of NH_3 and exploring ways to minimize the NH_3 formation when CO or hydrocarbons are used as reductants. TGA-IR results showed that the original main decomposition products NO_2 and NO obtained under N_2 atmosphere transfer to NH_3 and H_2O in presence of H_2 atmosphere. The influences of BaO loading, the presence of Pt, and the TGA heating rate on the decomposition temperature and product distribution are discussed. L/R cycle experiments showed that under rich condition the major reduction product of NO_x in the presence of steam is NH_3. The formation of NH_3 is attributed the reaction between NO_x and H_2, with H_2 stemming from water gas shift (CO as reductant) or steam reforming (HC as reductant). Catalysts operated in L-R cycle mode are more effective to reduce NO_x into NH_3 than catalysts operated only under rich conditions. Additional experiments showed that the combination of Pt-BaO/Al_2O_3 and Co^{2+} exchanged zeolite could effectively inhibit the NH_3 formation in L-R cycle (L/R=60/20 sec). However, if operation under rich condition is prolonged to 60 seconds (L/R=60/60), the major product under rich condition is still NH_3.