126f Surface Reaction Mechanism for Reduction of No by C3h6 under Lean Burn Conditions

Dinesh Mantri, Viral Mehta, and Preeti Aghalayam

Selective catalytic reduction (SCR) of NO by various hydrocarbons is an effective technique for reduction of pollutant emissions by automobiles, under excess oxygen conditions. The presence of oxygen in the exhaust of lean-burn gasoline engines makes the conventional catalytic converter impractical, opening the door to the novel SCR technology. In SCR, the effect of the presence of oxygen in the feed is to cause a drop in NO conversion at high temperatures. The temperature, at which the NO conversion is maximum, depends on both the catalyst and the reductant. Selectivities to N_2 and N_2O vary with the temperature of the reaction. In this presentation such experimentally observed features of the selective reduction of NO by C_3H_6 are analysed and explained using a detailed surface reactions model based on NO adsorption and dissociation.

Among the different hydrocarbons, C_3H_6 is found to be the best reducing agent for selective catalytic reduction of NO under lean burn condition. A detailed surface reaction mechanism is proposed for the NO-C_3H_6-O_2 reaction on Pt catalyst. Numerical simulation is used for the investigation and the performance of catalyst properties, the surface reaction mechanism is coupled with the Pseudo-Steady State Hypothesis (PSSH) and isothermal Continuously Stirred Tank Reactor (CSTR) model. The kinetic data required for the simulation is calculated using the Unity Bond Index-Quadratic Exponential Potential (UBI-QEP) method on Pt(111) or taken from the literature. The results obtained from the simulations compare well with experimental data from the literature. It is found that the conversion of NO shows a maximum value at ~290 ^oC. The C_3H_6 oxidation is observed to increase with temperature upto this peak temperature, and then remain constant with further increase in reactor temperature. The competition for catalyst surface sites between the surface species CO*, O* and N* (* refers to adsorbed species) is found to be critical. In particular, the increased tendency of oxygen to adsorb on the catalyst sites leads to the decrease in NO conversion with an increase in temperature, at temperatures higher than the peak one. This modeling study is also used to determine the effect of NO/C_3H_6 ratio on NO reduction, and to explain the reason behind the different selectivities observed as the temperature is changed. Finally, screening of catalysts in terms of high reduction of NO and low selectivities to undesired products is presented.