

216h Nox Reduction by Urea under Lean Conditions over Cu/Alumina Catalyst

Erol Seker, Erdogan Gulari, Nail Yasyerli, Christine Lambert, and Robert Hammerle

The elimination of NO_x in a fuel rich exhaust gas stream over a three-way catalyst is a well-understood and well-established process. However, NO_x removal from the oxygen rich exhausts of diesel engines is still a challenging problem. NO_x removal from the lean exhaust stream by the selective catalytic reduction with a hydrocarbon has been an active research area since it was demonstrated that hydrocarbons could be used as reductants. The uses of hydrocarbons as reducing agents to reduce NO_x from the exhaust of the lean burn engine are practical. However, they have low to moderate reducing efficiency and also produce undesirable by-products, such as N₂O. It seems that urea, as an ammonia source, is the best choice for the reduction of NO_x from the automobile exhaust because urea is not toxic and can be easily transported on board of an automobile as a high concentration aqueous solution. We present the effects of the preparation methods and copper loading on the activities of alumina supported copper catalysts for the reduction of NO_x by a urea solution under an oxidizing condition with and without SO₂. We prepared alumina supported catalysts using two sol-gel procedures and the impregnation method. Final catalyst formulation in both of the sol-gel procedures was achieved in a single pot whereas copper nitrate salt was impregnated onto a commercial γ -alumina and the sol-gel made alumina to prepare impregnation catalysts. Copper loadings were changed from 1 wt.% to 5 wt.%. In all experiments, 0.1 g of a catalyst was tested for NO_x reduction activity under a total gas flow rate of 176 cc/min containing 300 ppm NO (plus ~10 ppm NO₂ impurity in the NO cylinder), 7% O₂, ~300 ppm urea, 35 or 260 ppm SO₂ (when used) and He as balance. Among all the catalysts, 1 wt.% Cu on alumina prepared with the sol-gel procedure I (designated as 1%Cu-SG-I) showed the highest activity and the broadest temperature window of activity, such as ~98% conversion achieved between 400 o and 500 oC. However, other catalysts lost significant activities at temperatures above 450 oC. We also found out that the addition of 35 or 260 ppm SO₂ to the feed stream did not reduce the activity and N₂ selectivity of 1%Cu-SG-I catalyst as long as there was a continuous supply of the urea solution. Among the impregnation catalysts, 1 wt.% Cu on commercial alumina catalyst showed the lowest activity at all temperatures. In separate urea decomposition and ammonia oxidation studies, we found out that most of urea decomposed on the alumina support and all the sol-gel made catalysts were highly selectivity to N₂ (higher than 80%) in the oxidation of ammonia at all temperatures.