SELECTIVE CATALYTIC REDUCTION OF NO BY C_3H_6 OVER BIMETALLIC Rh-Ag/Al_2O_3 AND Rh-Ru/Al_2O_3 CATALYSTS

Areti KOTSIFA, Dimitris I. KONDARIDES and Xenophon E. VERYKIOS

Department of Chemical Engineering, University of Patras, GR-26504 Patras, Greece
E-mail: dimi@chemeng.upatras.gr

EXTENDED ABSTRACT

The selective catalytic reduction (SCR) of NO by propylene in the presence of excess oxygen has been examined over Al_2O_3-supported bimetallic Rh-M catalysts (M= Ag, Ru) of variable metal composition. Catalysts were prepared by the wet impregnation method using appropriate salts as metal precursors. The total metal loading was kept constant at 1 wt.% while the relative composition of the metallic phase (M/Rh+M) varied between 0 and 100%. The effect of the presence of silver or ruthenium on the chemisorptive properties of rhodium was investigated by equilibrium chemisorption of hydrogen at 25^oC and by FTIR spectroscopy, using CO as a probe molecule. The catalytic performance of the monometallic and bimetallic catalysts has been investigated in the temperature range of 150-550°C, using a feed composition consisting of 0.1% NO, 0.35% C_3H_6 and 5% O_2 (balance He) and a total flow rate of 200 cm^3/min (W/F=0.018 $g \cdot s \cdot cm^{-3}$).

Results show that the chemisorptive and catalytic properties of Rh are significantly influenced by the presence of Ag or Ru, in a manner which depends on the nature and composition of the bimetallic catalysts. Under the experimental conditions employed, the maximum conversion of NO to reduction products (N_2 and N_2O) increases from 56% over monometallic Rh to 75-80% over catalysts containing 5% Ag or 30% Ru, while further increase of the content of the second metal leads to a decrease of catalytic activity. FTIR spectra obtained following adsorption of CO on bimetallic catalysts show that the position of bands assigned to CO species adsorbed on rhodium shift toward higher vibrational frequencies in the presence of Ag or Ru, indicating weaker bonding of CO with the catalyst surface. This is further confirmed by the lower desorption temperatures observed over bimetallic catalysts in subsequent temperature desorption experiments. It is proposed that the weakening of bonds between CO molecules and rhodium is due to charge transfer or shift of electron clouds from the higher electronegativity metal atoms (Rh) towards those of lower electronegativity (Ag, Ru). As a result, the catalytically active rhodium phase becomes more electropositive and resistant to oxidation and, therefore, more active under reaction conditions.

It is concluded that the electronic type modifications induced on Rh by the presence of neighbouring Ag or Ru atoms alter its chemisorptive properties and, concomitantly, its catalytic behaviour for SCR of NO by propylene. It is shown that bimetallic catalyst formulations are promising candidates for the development of novel DeNO$_x$ catalysts efficiently operating under conditions found in the exhausts of lean-burn and diesel engines.

Key words: Rhodium catalyst, Rh-Ag catalyst, Rh-Ru catalyst, bimetallic clusters, nitrogen monoxide, propylene, selective catalytic reduction.