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Control of Emissions of Nitrogen Oxides

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This volume contains papers given at a symposium sponsored by the Catalysis and Surface Science Secretariat at the 205th National Meeting of the American Chemical Society, held in Denver, Colorado, from 28th March to 2nd April 1993. Although the symposium covered a number of aspects of environmental gaseous pollution from mobile and stationary sources, the principal emphasis was on control of nitrogen oxide (NOx) emissions, and work was reported on platinum group metal based catalysts, notably containing platinum, palladium and rhodium.

Approximate molecular orbital computations of adhesion and nitric oxide (NO) reduction for rhodium, palladium and platinum monolayers on the (0001) faces of oxygen and aluminium of α-alumina, were reported by T. R. Ward, P. Alemany and R. Hoffmann (Cornell University) who concluded that the support significantly affects the Fermi levels of the composite system, which in turn affects the NO adsorption mode. They suggest that the platinum-oxygen interface is particularly suited for both dissociative adsorption and coupling of two adsorbed nitrosyls, yielding a reduced form of N_2O_2.

The state of platinum-rhodium catalysts during three-way catalysis continues to be important. B. E. Nieuwenhuys and J. Siera (Leiden University) and K.-I. Tanaka and H. Hirano (University of Tokyo) presented a joint paper on differences in behaviour of platinum, rhodium and platinum-rhodium surfaces towards NO reduction. Silica was chosen to minimise support effects; experiments were performed with synthetic gas mixtures. They concluded that the catalytic properties depend strongly on the gas phase composition and temperature, and on the composition of the platinum/rhodium particles. Under oxidising conditions and at temperatures above 600 K, rhodium segregates to the surface; but under reducing conditions, platinum-rhodium alloy particles are reformed. This significantly influences the reactions of NO with both carbon monoxide(CO) and hydrogen, since rhodium is generally a better catalyst than platinum for the CO/NO reaction, while platinum is better for the hydrogen/NO reaction. Thus the predominant mechanism for NO removal depends on catalyst surface composition.

Infrared studies performed by G. Srinivas, S. S. C. Chuang and S. Debnath (University of Akron) on the interaction of NO and CO on rhodium/silica and cerium-rhodium/silica clearly show the formation of rhodium-NCO and silicon-NCO as the predominant species during the CO/NO reaction at 723 K. However, isotopic studies under steady state conditions indicate that these species are "spectator" species rather than the principal reaction intermediates.

Palladium-only catalysts were compared with platinum, rhodium and platinum/rhodium systems for three-way control in closed loop controlled motor vehicles, by J. C. Summers and W. B. Williamson (Allied Signal). While palladium has significantly better hydrocarbon control, its NOx performance, especially under rich operating conditions, tends to be adversely affected by lead and sulphur. This can be overcome by developing palladium catalysts less sensitive to these poisons, or by combining separate palladium only and platinum/rhodium catalysts in multibrick systems, leading to converters containing varying amounts of all three metals, depending on car calibration and fuel quality.

While most papers discussed NO reduction, the effects of sulphated platinum/zirconia catalysts in NO and sulphur dioxide (SO_2) oxidations were reported by J. R. H. Ross and coworkers, (University of Twente). The sulphated catalyst had higher activity in both reactions, but increased selectivity for NO. A similar increase in NO selectivity occurred after ageing platinum/zirconia in a NO/SO_2/oxygen gas mixture, and sulphate could be detected on the aged catalyst. These effects are ascribed to changed surface acidity modifying adsorption strength.

Despite the papers being at least 18 months old, they are still a valuable addition for the design of catalytic systems to meet the emission limits set for the next decade. D.E.W.