

CATALYTIC ABATEMENT OF POLLUTANTS FROM DIESEL EXHAUSTS

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Abstract—K and Co,K supported catalysts were studied to determine the mechanism of the soot combustion. The thermal stability of these catalysts and the resistance to water and sulfur are also addressed. The support plays an important role. La_2O_3 contributes through the formation of carbonate-type intermediates and CeO_2 supplies the oxygen necessary for the redox mechanism to take place. However, the addition of Co to the catalyst supported on La_2O_3 , to supply the redox function, leads to a loss of the thermal stability as a consequence of the formation of a perovskite structure. The soot-catalyst contacting phenomenon was also addressed. A synergic La-K effect was observed, by which mechanical mixtures of soot with K- La_2O_3 showed higher rates than those observed when K and La were deposited on soot. Preliminary experiments using barium as a catalytic trap for NO_x and the simultaneous removal of NO_x and soot are also presented.

Keywords-- Soot Combustion, Nitric oxide Removal, Cobalt – Potassium Catalysts.

I. INTRODUCTION

An active search has been under way in the last several years to find active catalysts for the abatement of diesel exhaust contaminants. The main pollutants emitted for this type of engines are diesel soot particles and nitrogen oxides. The combination of traps and oxidation catalysts appears to be the most plausible after-treatment technique to eliminate soot particles (Heck and Farrauto, 1995). The catalysts to be used in the combustion of diesel soot must be stable to high temperatures and the presence of other combustion products. On the other hand, the activity should be high enough at temperatures as low as 200°C for low charge diesel engines, unless an external heating power is supplied to the catalytic filter. In any case, the catalyst stability is a key factor in determining its applicability in a commercial scale.

Studies with a large number of formulations have been reported during the last few years, and the soot-catalyst contact appears to be one of the most important problems to be overcome (van Setteen et al., 2000). We have previously studied the catalytic behavior of catalysts containing cobalt and/or potassium supported

on La_2O_3 , CeO_2 , and MgO , mixed with soot under tight contact conditions (Miró et al, 1999, Querini et al, 1998, Querini et al, 1999). Based on these results and results presented in this work, the reaction mechanisms are proposed, being slightly different for each support. In this work, Barium has been included in the formulation of the more active catalysts, since it has been used as a NO_x trap (Mahzoul et al., 1999). The activity and stability of these catalysts is therefore also addressed.

II. EXPERIMENTAL

A. Soot and Catalyst preparation

The soot was obtained by burning commercial diesel fuel (Repsol -YPF, Argentina) as described elsewhere (Querini et al, 1998). Catalysts were prepared by wet impregnation, with different amounts of potassium (0, 4.5 and 7.5 wt.%), cobalt (0 and 12 wt.%), and/or barium (10, 16, 22 wt.%) by using KOH, $\text{Co}(\text{NO}_3)_2$ and $\text{Ba}(\text{CH}_3\text{COO})_2$ respectively, as starting materials. Cerium oxide and lanthanum oxide were used as support. The support was added to a suspension of the solution containing K, Co, and/or Ba, which was stirred at 90°C until a paste was formed. Then, the mixture was dried at 100°C and calcined at different temperatures (400 – 700°C).

To investigate about the soot-catalyst contacting effects, samples of soot directly impregnated with either (K) or (K, La) were prepared. The impregnation procedure was carried out from a slurry of soot and solutions of the salts (Lanthanum acetate, Potassium hydroxide) in methanol. Typically, 5 wt.% of the metals were loaded and the paste formed after stirring at 30°C was dried at 100°C.

B. Activity test

The catalytic activity for the combustion of soot was determined by temperature-programmed-oxidation (TPO), of carefully prepared mixtures of catalyst and soot (20:1). The catalysts were mixed with the soot in a mortar, during 3-5 min. A gaseous flow with 6% oxygen in nitrogen was used and the temperature was increased at a rate of 12°C/min, using 10 mg of the mechanical mixture. Experiments using $\text{NO} + \text{O}_2$ as feed were also carried out. Stability tests were also performed, treating the catalyst with 1000 ppm SO_2 at 400°C, and with air that was saturated with water at 25°C, also at 400°C.