

NO and C oxidation with Pt recovered from spent catalytic converters

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Introduction

Automotive catalysts today make almost 50% of the global demand for the Platinum Group Metals (PGM) due to emission limits enforced by international legislation. The catalysts contain either platinum or platinum/palladium combinations for oxidize carbon monoxide (CO) and unburnt hydrocarbons (HC) to carbon dioxide (CO₂) and water (H₂O). The recovery of these precious metals present in spent automobile catalytic converters [1] has been an important topic not only for the economic aspect but also for recycling rare natural resources. However, the PGM content is low (2 g per unit)[2], and the unit are enclosed in steel sheet and widely scattered.

The main purpose of this work is to develop a selective recovery process of platinum from a spent catalytic convert using a strong basic ion exchange resin and then to carry out catalytic activity comparison between catalysts based on recovered platinum from spent catalytic converters (Al₂O₃ as a support) and commercial Pt/Al₂O₃ catalysts.

Materials and Methods

For Pt recovering, 250g of a spent catalytic convert was treated with 1 liter of aqua regia (HCl/HNO₃ = 3:1) as leaching solution [3]. The obtained solution was then processed by means of the Column ion-exchange procedure[4] to recover the platinum. During the process, absorption atomic analyses were carried out to measure the platinum concentration in the solution.

Two 2wt% Pt/Al₂O₃ catalyst powder samples were synthesized via incipient wetness impregnation IWI [5]. The first sample was prepared by using the recovered platinum, whereas the second one was prepared by using an aqueous solution of hydrogen hexachloroplatinate (IV) provided by Sigma-Aldrich.

The activity of the prepared catalysts towards the carbon oxidation was analyzed by means of Temperature-Programmed Combustion (TPC), carried out in a fixed-bed micro-reactor under standard operating conditions [6] (air flow rate of 50 Nml·min⁻¹, carbon/catalyst 1:9 mass basis, W/F = 0.05 g·s·cm⁻³, GHSV of 9,000 h⁻¹, heating rate of 5 °C/min). The outlet gas was monitored by a CO/CO₂ NDIR analyzer (ABB).

The same catalysts were also tested for the oxidation of NO towards NO₂ with the same apparatus [6] according to the following conditions, gas mixture: 93 ppmv NO; 15 vol.% O₂, N₂ = balance; flow rate of 350 Nml·min⁻¹, W/F = 0.02 g·s·cm⁻³, GHSV of 40,000 h⁻¹, heating rate of 5 °C/min). The outlet gas was monitored by both a NO/NO₂ chemiluminescence analyzer (Eco Physics) and a N₂O NDIR analyzer (ABB).

Results and Discussion

After the leaching with aqua regia, a solution containing an elevated concentration of platinum (640 ppm) and several undesirable compounds (Al, Fe, Si, among others) was obtained. The column ion-exchange procedure with a strong anionic resin (Amberlite IRA - 400) was used to separate the platinum complex [PtCl₆]²⁻, recovering 71% of the present platinum, with a pureness of 91% (9% of Ca).

The SEM analysis carried out with the BSE detector showed a ceramic matrix of Al₂O₃ with platinum cluster having an average diameter of 1 µm finely dispersed onto the surface for the 2 wt% Pt_(recovered)/Al₂O₃ (see Fig. 1A). An higher platinum cluster average diameter was observed for the 2 wt% Pt/Al₂O₃ commercial sample (not reported).

The catalytic activity tests (Fig. 1B) concerning the carbon combustion showed that the 2wt% Pt_(recovered)/Al₂O₃ exhibited a slightly higher peak combustion temperature (T_p=561°C) compared with the commercial 2 wt% Pt/Al₂O₃ (T_p=555°C) but with a lower CO₂ formation (not reported). Shifting our attention towards the NO oxidation to NO₂, both catalysts showed a similar performance, reaching the highest conversion (~61%) at 310°C with no production of nitrous oxide.

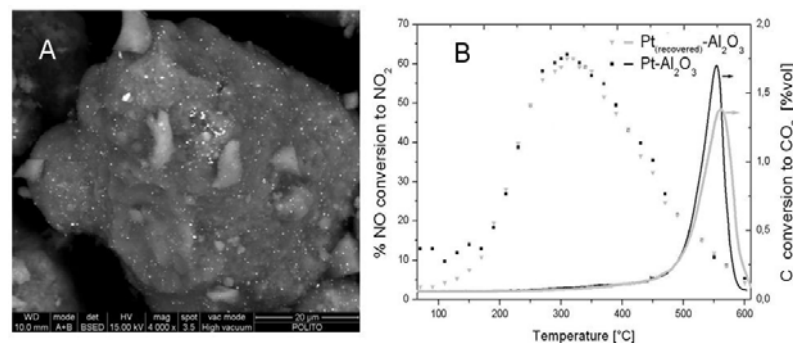


Figure 1. A) SEM view of 2 wt% Pt_(recovered)/Al₂O₃ with a magnification of 4000X; B) activity results regarding the C conversion to CO₂ and NO conversion to NO₂.

References

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