Effect of Precursor on the Coarsening Behavior of Nano-Particulate Alumina-Supported Pt-Pd Catalysts for NO Oxidation

Obiefune K. Ezekoye[1], Andrew R. Drews[2], Robert W. McCabe[2], George W. Graham[1] and Xiaoqing Pan[1]*
[1]*University of Michigan, Ann Arbor, Michigan 48109-2136 (U.S.A.)*
[2]*Ford Motor Company, Dearborn, MI 48121-2053 (U.S.A.)*
*panx@umich.edu

Introduction

NO oxidation is a key enabler for lean-burn automotive applications, and Pt is generally regarded as one of the best NO oxidation catalysts. It has previously been shown that the loss of Pt surface area in alumina-supported Pt catalysts resulting from particle coarsening at high temperatures under lean conditions can be moderated by alloying Pt with Pd, and that alloying Pt with up to 50 mol% Pd has a relatively minor effect on the turn-over frequency for NO oxidation [1]. The catalysts in the previous study were prepared from acetylacetonate (Ac-Ac) precursors using methods designed to promote alloy formation [2,3]. In this study we compare the coarsening behavior of Pt-Pd catalysts fabricated from nitrate precursors with the previous samples synthesized from Ac-Ac precursors.

Materials and Methods

Bi-metallic catalysts were prepared from Pt and Pd nitrate mixtures using the same support (Grace MI-307 high-surface-area alumina) and precious metal loadings as before. In the present study, catalysts were prepared by incipient wetness impregnation of the support (while catalysts from the Ac-Ac precursors were prepared by adsorption from solution). After impregnation, the catalysts were dried and calcined at 300°C for 1h in air, and catalyst samples were finally aged at 500°C and 900°C in a quartz tube furnace under lean conditions, as previously described [1].

These samples were then prepared for TEM examination by dispersing a small amount of powder on a 300 mesh carbon-coated copper grid. HRTEM was performed with a JEOL 3011 high resolution microscope equipped with an integrated x-ray EDS detector and a point-to-point resolution of 0.17 nm. Z-contrast scanning transmission electron microscope (STEM) analysis and other AEM techniques were performed on a JEOL 2010F field emission microscope.

Results and Discussion

Results from TEM precious metal particle size analysis of two catalyst compositions, pure Pt (labeled 100-0) and the 50 mol% combination of Pt and Pd (labeled 50-50) are summarized in Figure 1. (Particle size distributions for fresh catalyst samples could not be adequately characterized with the equipment available for this study and will be discussed in a future report.) After aging, catalysts synthesized from nitrate precursors have a larger mean particle diameter than samples from the Ac-Ac precursors, regardless of whether they contain Pd or not. This difference is greatest for the Pt-only sample aged at 500°C. Addition of Pd reduced the mean particle size for both aging conditions. Regardless of the precursor used, large highly faceted particles were found exclusively in the Pt-only catalysts, most strikingly for those samples aged at 900°C. The presence of particles with aspect ratios much greater than 1 (e.g., Fig. 2c) suggests that growth occurs via a vapor phase transport mechanism under the conditions described here.

Significance

Previous work has shown that alloying Pt with Pd, which is promoted by using Ac-Ac precursors, suppresses particle coarsening during lean aging. The present work focuses on bi-metallic catalysts from nitrate precursors which are more in line with materials that suppliers could employ to produce catalysts in large quantities. This work confirms that addition of Pd, even in a less directed manner, can still suppress coarsening of alumina-supported Pt, and it also demonstrates that although the precursor choice affects the mean particles size, the difference is less significant after high-temperature aging.

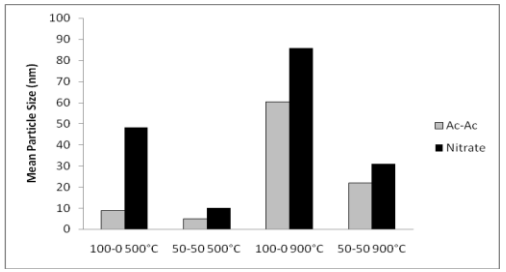
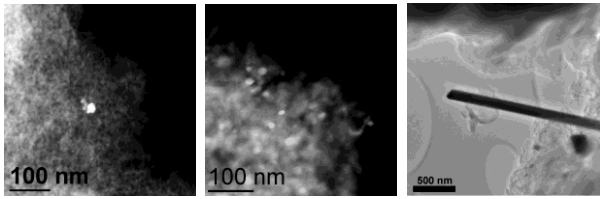


Figure 1. Mean precious metal particle size obtained from TEM observation.



(a) (b) (c)

Figure 2. Typical STEM images of 50-50 catalysts from (a) nitrate precursors and (b) Ac-Ac precursors, aged at 500°C. TEM image (c) of a Pt needle found in a 900°C aged Pt-only catalyst from nitrate precursor.

References

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