Activation and Deactivation Mechanisms of Rhodium/Alumina Catalysts

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Introduction

It is believed that the strong interaction between rhodium and alumina substrate in a high temperature oxidizing atmosphere is the main source of deactivation of rhodium [1]. This has led to the search for non-alumina alternative rhodium catalyst supports in the last three decades [2]. Yet, due to their unique pore structure, stability, surface functionality and low cost, alumina and surface-modified alumina are still heavily used in catalyst industries. A number of studies have been reported on the deactivation of rhodium/alumina catalysts [3]. Both reversible and irreversible deactivation were observed. Several deactivation mechanisms have been proposed including the formation of rhodium aluminate and alumina-encapsulated rhodium particles, and diffusion of rhodium oxide in alumina. In spite of the progress, there is a lack of systematic and quantitative correlation between the deactivation and the regenerability of the catalysts. A clear deactivation/activation mechanism is required for a better catalyst design.

Materials and Methods

Powder catalyst samples were prepared, aged, and studied, using a number of characterization methods, including TGA/DTA, pulsed CO chemisorption, H_2/O_2 redox cycling and adsorption capacity, N_2 adsorption, XPS, XRD, and diffuse reflectance UV-Vis. Catalyst activity for NOx reduction was evaluated by a powder reactor.

Results and Discussion

In this study, we conducted a more quantitative mapping of the deactivation and activation processes of Rh/alumina catalysts. The results show that rhodium deactivation is mainly through a continuous penetration of ionic rhodium species into the alumina structures. No abrupt structural change was observed before the total collapse of the alumina pore system. The completeness and easiness of the regeneration of aged rhodium to an active metal form depends largely on how severely the catalyst was aged. The implication of these results for catalyst applications is discussed.

References

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