PREPARATION OF A COPRECIPITATED Au/Al_2O_3 CATALYST: EFFECTS OF THE PREPARATION CONDITIONS ON CATALYST PROPERTIES

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Introduction

Supported Au catalysts have been found to be very active for oxidation reactions including CO, H_2 and methane. They are also active in the selective oxidation of propene and other hydrocarbons, alcohols, and CO in an H_2-rich stream [1]. All of these reactions require the design and development of new catalysts for achieving high activity, selectivity, thermal stability and long life. Various methods are employed in the preparation of supported metal catalysts, the most commonly used methods being impregnation and precipitation. Although impregnation is one of the easiest methods of making a catalyst, the homogeneity of product, especially for high metal loading, and the reproducibility of the process are better in coprecipitation, and also, the metal crystals formed are smaller. The activity of supported gold based catalysts has been shown to depend strongly on both preparation method and the support. Gold catalysts prepared by impregnation do not show high activity in oxidation reactions [2]. In order to obtain a gold catalyst showing high activity in CO oxidation reaction, the catalysts have to be prepared via coprecipitation or deposition –precipitation [3]. Beside the preparation method also the synthesis conditions, such as precipitation pH, precipitation reagent and calcination temperature, have significant effects on the properties of gold catalysts[4].

In this study a coprecipitation method has been developed and modified for the preparation of Au/Al_2O_3 catalysts developed for CO oxidation in H_2 rich environment (for fuel cell applications). A parametric study has been conducted to investigate the effect of preparation variables on the structural and catalytic properties of the catalyst and to develop a suitable coprecipitation recipe for preparing various gold-alumina catalysts.

Results and Discussion

Au/Al_2O_3 catalysts with a target gold loading of 5.0 wt % were prepared by coprecipitation method from chloroauric acid and aluminum nitrate solutions with different precipitation agents. Chloroauric acid was first dissolved in distillated water and cooled to 5 to 10 ^{0}C. Then the pH of the $HAuCl_4$ aqueous solution was adjusted to a constant value by adding HNO_3 . Different precipitation agents (NaOH, KOH, LiOH and Na_2CO_3) and precipitation pH were employed to investigate the effects of these variables on catalyst properties.

Physical and chemical characterization is an integral part of catalyst design and is performed either as a quality control operation after preparation or with the aim of finding a link between catalyst performance and the catalyst structural and electronic properties, which is the most common case in catalysis studies. Different physical and chemical characterization methods have been used to characterize the catalysts. Physical properties such as specific surface area, pore volume and pore volume distribution were determined for each catalyst sample after each pretreatment and reduction procedure. The total surface areas (TSA) and the total pore volumes (TPV) were determined by nitrogen adsorption from N_2-He mixtures using a multipoint technique together with the BET equation and by the nitrogen adsorption-desorption method, respectively. Metal surface areas of coprecipitated catalyst samples were determined by irreversible CO adsorption by using a Hewlett Packard 5890 Series II gas chromatograph fitted with a TCD. TSA, TPV and MSA values of coprecipitated Au/Al_2O_3 catalysts were discussed in terms of preparation pH and precipitation reagent.

Temperature-programmed (TP) studies (TP-oxidation and TP-reduction) were performed for the thermal characterization of the catalysts prepared by using thermogravimetric analysis (TGA). SEM and XRD analysis were performed to determine the catalyst structure.

The effects of preparation parameters were discussed on the basis of catalyst physical and structural properties. It is found that both precipitation reagent and precipitation pH have significant effect on the physical and structural properties of coprecipitated Au/Al_2O_3 catalyst.

References

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