Highly reproducible syntheses of active Au/TiO₂ catalysts for CO oxidation by deposition–precipitation or impregnation

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Abstract

Gold catalysts supported on TiO₂ were prepared by a deposition–precipitation (DP) method to investigate how highly reproducible performance of the gold catalysts in CO oxidation can be achieved. A protocol was established for synthesizing identically performing catalysts by different operators. The results show that for this synthesis route, the calcination step is not needed to form highly active Au/TiO₂ catalysts, but leads to decreased activity. Improved catalytic activity was observed when a high solution pH was adjusted during the precipitation. Surprisingly, wet impregnation followed by ammonia steam treatment and a washing step with water also leads to Au/TiO₂ with 2- to 4-nm individual gold particles highly dispersed on the TiO₂ surface. In addition, this catalyst is active for room temperature CO oxidation. The temperature for 50% conversion of CO is below 25°C, which is comparable to that of the gold catalyst prepared by the DP method. Therefore, contrary to reports in the literature, the impregnation method can be used in the preparation of high-activity gold catalysts.

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1. Introduction

A tremendous increase in research on gold catalysis was triggered by Haruta’s first reports on the unexpectedly high catalytic activity of supported gold nanoparticles in CO oxidation [1,2]. Several methods for the preparation of highly active gold catalysts have been developed. Besides classical deposition–precipitation (DP) proposed by Haruta [3], other methods, including co-precipitation [4], co-sputtering [5], chemical vapor deposition [6], grafting [7], and adsorption of gold colloids on metal oxides [8] have been investigated. Usually these preparation methods can produce small (<10 nm) Au particles that are strongly attached to the supports. Despite the numerous methods developed, however, the DP method still seems to be the most efficient method of preparing highly active gold catalyst. In particular, using TiO₂ as a support, small, highly dispersed gold nanoparticles on the support can be achieved. Notably absent in the list of synthetic methods is the classical impregnation, because this pathway typically results in the formation of large gold particles with correspondingly low activity.

It has been pointed out that the nature of the support materials, as well as the physical state of the support, can strongly influence the activity of the resulting gold catalysts [9,10], although recent reports have demonstrated that “naked” gold particles can exhibit activity in aerobic glucose oxidation, which is comparable with supported particles [11]. Various metal oxides, including TiO₂ [3], Al₂O₃ [12], ZrO₂ [13], MgO [14], Fe₂O₃ [15], and Co₃O₄ [16], have been investigated as gold supports, with the goal of creating high catalytic activity. In addition, ordered porous silicas, such as SBA-15 [17] and MCM-48 [18], have been successfully used for the synthesis of active gold-based catalysts. Besides the preparation methods and the types of the supports, various other synthetic conditions, including pH value in the DP method, pretreatment, and calcination temperature, significantly influence the properties of the gold catalysts [13,19].

Despite the numerous studies published on supported gold catalysts, there is still no clear picture of the origin of the catalytic activity. In fact, the results reported in literature concerning gold catalysis are often contradictory, due to difficul-