

Synthesis and Catalytic Application of One-Dimensional and Porous α - Fe_2O_3

Ziyi Zhong*, Judith Ho, Jaclyn Teo

¹Institute of Chemical and Engineering Sciences, 1 Pasek Road, Jurong Island, Singapore, 627833

*Zhong_ziyi@ices.a-star.edu.sg

Introduction

In the last decade there has been a dramatic increase in the number of publications related to gold (Au) catalysis, though, traditionally, Au-containing catalysts were almost marginal value. However, this view is only partially true when the gold particles size is large ($d \geq 10$ nm), in which intrinsic size effects and/or metal/support interactions are only weakly expressed [1-2]. When the Au particles are below 5nm in size, remarkable activity at low temperatures, e.g., below 100°C in CO oxidation, is reported [2]. The Au-based catalysts are active for many reactions, e.g., for oxidation of CO and volatile organic compounds (VOCs) at low temperatures. Among the catalyst supports, it is reported that α - Fe_2O_3 and TiO_2 are good ones for the supported Au catalysts for CO oxidation. However, commercial α - Fe_2O_3 usually has low surface area which limits its catalytic activity, and the stability of the very tiny Au particles still poses a big challenge. Herein we report a method that can synthesize porous 1-D α - Fe_2O_3 by a one-pot reaction, and then the porous nanostructure was deposited with very small Au particles using our invented method. The prepared catalysts exhibited very high catalytic activity for CO oxidation at low temperatures.

Materials and Methods

The 1D and porous α - Fe_2O_3 was prepared by a hydrothermal method at 100°C for 10 hours. $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ was used as precursor and a structure-directing agent was used to lead to a preferred growth along certain direction. The prepared product was dried and calcined at 300°C for 0.5h, and was further used as catalyst supports. The Au/ α - Fe_2O_3 catalyst was prepared using our developed method[3]. Firstly, 0.50g α - Fe_2O_3 was put in 10ml DI water, then 0.01M HAuCl_4 and lysine was added subsequently. The pH value of the suspension was adjusted to 5 -6 with 0.10M NaOH. The suspension was subjected to sonication for 20 seconds, and during the period, freshly prepared NaBH_4 was injected instantly. The suspension solution was washed with DI water for 4 times by centrifuge. Similarly Au supported catalyst on a commercial α - Fe_2O_3 was also prepared.

The oxidation of CO at low temperatures was chosen as a model reaction. The measurement of catalytic oxidation of CO was carried out in a fixed-bed microreactor. Prior to the test, the catalyst was pretreated in air at 200°C for 1h. Reactant gas containing 1% CO in air was passed through the catalyst bed at a GHSV of 25500h⁻¹. The outlet gases were analyzed with an on-line GC (Shimadzu).

Results and Discussion

The as-prepared iron oxide was α - $\text{FeO}(\text{OH})$. TEM results showed that the as-prepared iron oxide was 1D nanostructure with a diameter of ca.15-25nm, and a length of ca. 100-150nm. After the calcination at 300°C, it was converted to α - Fe_2O_3 with a surface area of

143m²/g; Also, some pores were created in the 1D structure. The pore size peaked at ca. 2-3nm. The Au loading was ca. 3.0 wt% and the Au particle size was ca. 2-6 nm. The prepared Au catalysts showed a very high catalytic activity. At 50°C 100% conversion of CO was achieved, and this conversion could be maintained even the GHSV was increased to 70,000h⁻¹. As a comparison, commercial α - Fe_2O_3 was also used as the catalyst support, and a much poorer catalytic activity was observed.

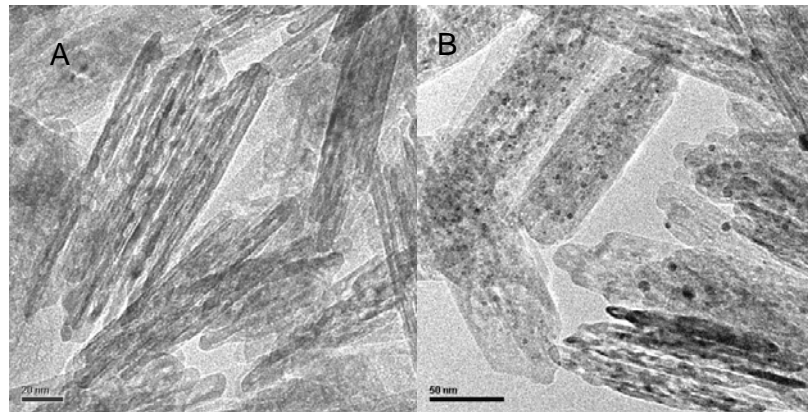


Fig.1. 1D and porous α - Fe_2O_3 before (A) and after (B) Au deposition. The small black dots in Fig.1B are Au particles.

Conclusions

We have successfully developed a method for synthesis of 1D and porous α - Fe_2O_3 . The supported Au catalyst on it exhibits very high catalytic activity of CO oxidation at low temperatures.

References

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