



Highly active and sintering-resistant heteroepitaxy of Au nanoparticles on ZnO nanowires for CO oxidation[☆]

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ABSTRACT

Gold was supported on commercial ZnO powders (P) and homemade ZnO nanowires (NWs) by a modified deposition–precipitation method. X-ray diffraction and transmission electron microscopy investigation indicated that the size of the Au nanoparticles (NPs) depended strongly on the calcination temperature. The Au NPs were highly dispersed (< 5 nm) on both supports with calcination temperatures < 400 °C. However, after calcination at 600 °C the Au NPs aggregated much more severely on ZnO P than on ZnO NWs. Gold NPs epitaxially grew into the {10–10} facets of the ZnO NWs after calcination at temperatures > 400 °C. Such unique anchoring mechanism accounts for the much better experimentally observed sintering resistance. X-ray photoelectron spectra showed that Au existed as both metallic Au⁰ and Au^{δ+} species in all the synthesized catalysts with or without calcination treatment; the ratios of Au^{δ+}/Au⁰, however, varied, depending on the treatment conditions. Catalytic tests showed that the activity for CO oxidation strongly depended on the size of the Au NPs. After calcination at 600 °C, the specific rate for CO oxidation at room temperature decreased about 30 times on Au/ZnO P but only about 4 times on Au/ZnO NW. Stability tests demonstrated that the Au/ZnO NW catalysts had better stability for CO oxidation.

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1. Introduction

Supported Au nanoparticles (NPs) have attracted intensive research interest in heterogeneous catalysis since Haruta et al. reported their remarkably high activity for low-temperature CO oxidation [1,2]. Supported Au catalysts have demonstrated high activity and/or selectivity in many important chemical reactions, including NO reduction, oxidation of hydrocarbons, water-gas-shift (WGS) reaction, removal of CO from hydrogen steams, etc. [3–9]. Previously, most of these reactions are usually catalyzed by platinum-group metals (PGM). Because of the greater availability of Au compared to PGM, supported Au catalysts could potentially replace expensive PGM catalysts in such reactions [3]. In addition, supported Au catalysts have advantages over other metals for many

reactions due to their unique catalytic properties. However, a major issue for supported Au catalysts is that Au NPs tend to agglomerate when being heated or used at elevated temperatures, which severely limit their practical applications. To reduce or even prevent such sintering effect, many attempts have been made including synthesis of alloy particles [10,11], fabrication of core-shell structures [12], encapsulation of metal particles [13], confinement of metal NPs in the pores of mesoporous supports [14–16] and enhancement of the interaction with a support material [17–20]. These strategies certainly improved the anti-sintering ability of Au NPs at reasonably high temperatures (e.g. < 500 °C). However, it rarely realized good sintering resistance to calcination at much high temperatures (e.g. > 600 °C) [21,22] and to achieve such a goal remains a grand challenge.

It is well known that for a supported catalyst the metal–support interaction (MSI) plays an essential role: MSI not only strongly affects the catalytic performance but also, to a great extent, determines the NP's stability as a durable catalyst [23]. Therefore, by carefully engineering the interfacial bonding between metal NPs and the support, one can enhance the stability/sintering-resistance of supported metal catalysts [24]. Heteroepitaxy, the oriented growth of one crystal/film on another with

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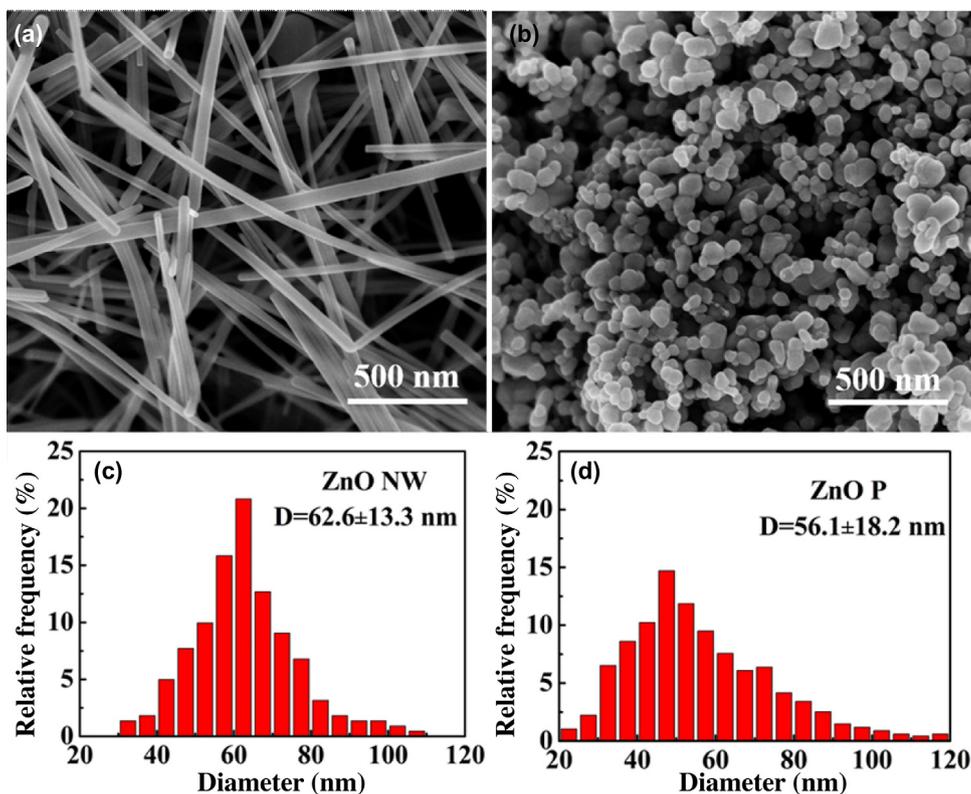


Fig. 1. SEM images and the diameter distributions of (a, c) ZnO NWs and (b, d) ZnO P.

fixed crystallographic orientation relationship, has been extensively studied [25] and widely applied in semiconductor industry [26]. The structural and dimensional resemblance between the crystal/film and the substrate leads to the formation of coherent or semi-coherent interfaces of which the energy is lower than that of the non-coherent configurations [27]. Such low-energy interfaces stabilize otherwise unstable composites or metastable polymorphs [27]. Therefore, epitaxial growth can be utilized to immobilize metal NPs on support materials, providing potential applications in the development of sintering-resistant supported metal catalysts [23]. It has been reported that epitaxial growth of metal NPs on support can improve the activity, selectivity and/or stability of the catalysts of interest [28–35]. For instance, it was reported that long-term stability of RuO₂ could be maintained during the Deacon reaction if RuO₂ has an epitaxial relationship with the surfaces of rutile TiO₂ and SnO₂ [28–31]. Similarly, Enterkin et al. found that growth of Pt NPs onto SrTiO₃ nanocuboids exhibited cube-on-cube epitaxy with a thermodynamically stable configuration and the morphology and selectivity can be tuned precisely in order to achieve the desired catalytic properties [32–34]. Li et al. [35] reported a well-defined cuboctahedral MgAl₂O₄ spinel support material that is capable of stabilizing Pt NPs on the relatively abundant support facets with an epitaxial relationship [36].

We have recently been using the epitaxial growth of metal NPs as a new approach to developing stable supported metal catalysts for various catalytic reactions [23,37–42] and have already demonstrated that this strategy can be extended to the fabrication of highly sintering-resistant Au NP catalysts [43]. In the present work, we extended the applicability to different catalytic reactions, provided more characterization data, and presented a systematic and detailed study of the newly developed supported Au catalysts for CO and methane oxidation as well as the steam reforming of methanol.

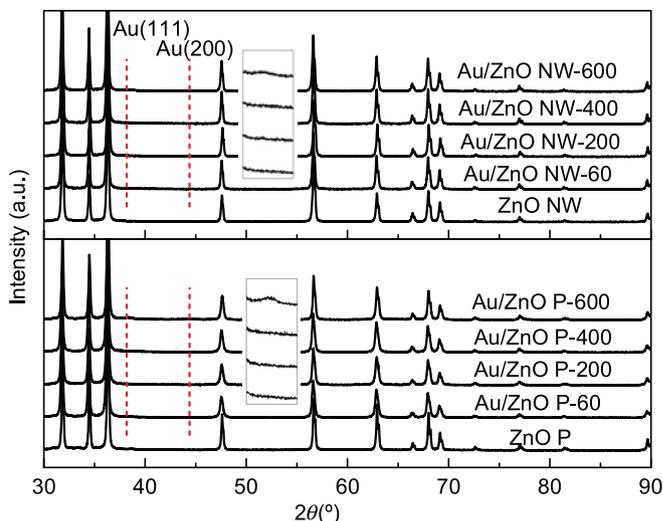


Fig. 2. XRD patterns of Au/ZnO NW and Au/ZnO P catalysts calcined at different temperatures. The insets show the enlargement of the Au (111) peak position.

2. Experimental

2.1. Catalyst preparation

ZnO nanowires (NWs) were fabricated by a modified vapor transport deposition method [44]. Firstly, a uniform mixture of ZnO and C was obtained by mixing ZnO powders (99%, Sigma-Aldrich) and carbon black (Carbot Corp) in the weight ratio of 1:1 with the addition of deionized water. Then the mixture was dried in the oven at 200 °C for 10 h to remove water. For the synthesis of ZnO

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