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Highly Active CeO_2 Nanorods Support for CO Oxidation

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The study of metal nanoparticles on oxide supports is of importance in heterogeneous catalysis because the size and nature of the interaction of a metal nanoparticle with an oxide support are critical in determining catalytic activity and selectivity [1-5]. Reduction and oxidation (redox) at elevated temperatures are essential steps for the catalytic reactions. Due to the superior oxygen transport capacity, ceria (CeO_2) with other oxides constitute an important class of catalysts and/or catalyst supports that can exchange oxygen rapidly under variable reducing or oxidizing conditions, mainly accommodated by a reversible valence change ($2Ce(IV)O_2 \leftrightarrow Ce_2(III)O_3 + 1/2O_2$). In this paper, we report a facile synthesis of CeO_2 nanorods with {200} surfaces by a hydrothermal reaction, and catalytic activity characterization of 1wt% Au/CeO_2 and 1wt% Ni/CeO_2, aiming to understand the support shape effect for low temperature CO oxidation.

CeO_2 nanorods were prepared using a hydrothermal method. Typically 0.1M $Ce(NO_3)_3{\cdot}6H_2O$ and 6M NaOH mixtures were heated to ~130 °C and held for 48 hrs in a sealed 200 mL Teflon-lined autoclave (~50 % fill). Then the autoclave was cooled to room temperature before the solid products were recovered by suction filtration. The materials were washed thoroughly with distilled water to remove any co-precipitated salts, then washed with ethanol to avoid hard agglomeration in the nanoparticles, and dried in air at 50 °C for 12 hrs. Transmission electron microscopy (TEM) characterization was performed using a JEOL2100 operated at 200 kV and equipped with an EDAX detector and annular dark-field detector. CO temperature programmed reduction (CO-TPR) study was examined using CO chemisorption on the Quantachrome iQ and Micrometrics 2920 to explore how much CO adsorbs as a function of temperature. The catalytic oxidation of CO was conducted by using a fixed bed plug flow reactor system. 1vol%CO/20vol%O_2/79vol%He with a 70 mL/min flow rate was supplied through mass flow controller and passed through the catalyst bed. The catalyst (~100 mg) was mixed with quartz wool (coarse, 9 μm) and filled in the quartz tube reactor. The reaction temperature was programmed between room temperature and 300^{o}C and monitored by thermocouple. The reactant CO and product CO_2 were analyzed by using an on-line gas chromatograph (SRI multiple gas analyzer GC, 8610C chassis) system.

Figure 1 (a) and (b) shows typical low magnification and high resolution TEM images of CeO2 nanorods prepared by a hydrothermal method, respectively. We have used CO temperature programmed reduction (TPR) to measure the CO consumption ($2CeO_2+CO \rightarrow Ce_2O_3+CO_2$) during the reduction of CeO_2. Figure 1 (c) shows the CO-TPR profiles of CeO_2 nanorods, 1wt% Au/CeO_2, and 1wt% Ni/CeO_2 under a 5%CO/95%Ar. The two reduction regions for the surface and bulk reductions, respectively, are characteristics of ceria-type redox catalysts, and reduction percentage, reduction temperature and the distribution between the surface and bulk areas are good indicators for the overall performance of the material as an oxidation-reduction catalyst. We will present the correlation study between the CeO_2 nanoparticle size/shape and reactivity as determine by TEM, CO-TPR, and flow reactor test on the catalyst activity of CO oxidation.

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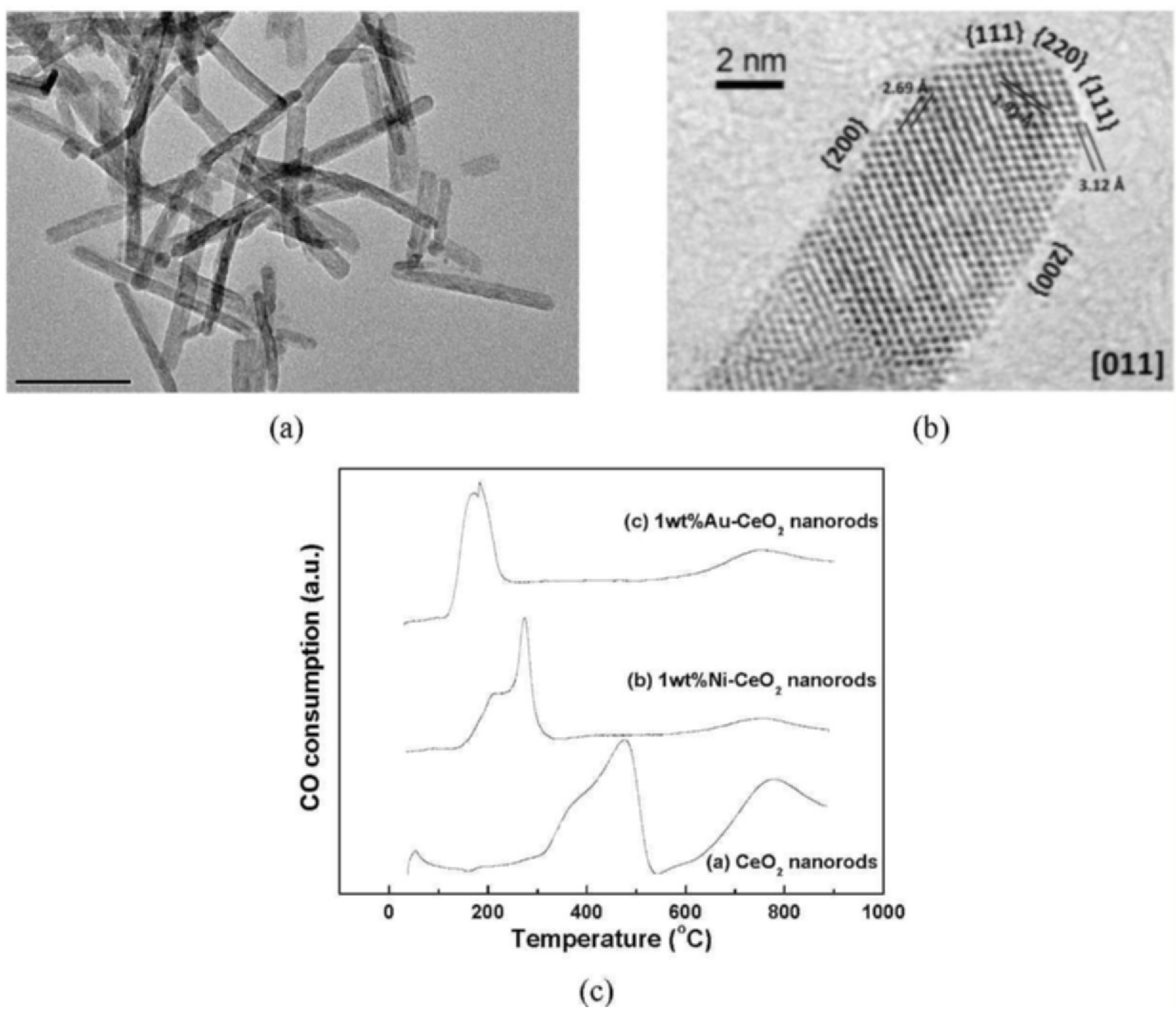

Figure 1 low magnification (a) and high resolution (b) TEM images of CeO_2 nanorods, and CO-TPR profiles of CeO_2 nanorods, 1wt% Au/CeO_2 and 1wt% Ni/CeO_2 under a 5%CO/95%Ar gas atmosphere.

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