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### Low-Temperature CO Oxidation Over CuO-TiO<sub>2</sub> Nanocatalysts

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Among the diverse catalytic processes, the heterogeneous catalytic CO oxidation is an important reaction for removal of small amounts of poisoning CO in fuel cell applications and environmental remediation. Therefore, there is a great need to develop highly active and stable nanocatalysts for catalytic CO oxidation at low temperature. Plasmonic nanocatalysts supported on reducible metal oxide such as CeO<sub>2</sub> and TiO<sub>2</sub> have been known for their superior catalytic activity at very low temperature but they are expensive and could suffer from particle agglomeration and sintering at high operating temperature (Veith, Lupini et al. 2009). Transition metals supported on reducible metal oxides are good substituents catalysts because of their low cost and wide-use along with activities per unit surface area similar to those of noble metal catalysts. They been shown to possess high oxygen release capacity at high range of temperature and have been shown as good candidate materials for oxygen storage and to provide oxygen for combustion and oxidation reaction at high temperature. (Royer and Duprez 2011; Hedayati, Azad et al. 2012; Song, Liu et al. 2013). In particular, supported CuO nanostructures have received a great deal of attention as non-expensive and non-plasmonic catalysts for oxidation reaction. (Caputo, Lisi et al. 2007; Hornes, Hungria et al. 2009; Royer and Duprez 2011; In, Vaughn et al. 2012; Komarneni, Shan et al. 2012; Chen, Xu et al. 2015; Fang, Xing et al. 2015; Kim and Liu 2015) In this study, we have developed a highly stable and active CuO-TiO<sub>2</sub> nanocatalyst that can catalyze the CO oxidation at low temperature window between 80–200°C. The CuO-TiO<sub>2</sub> nanocatalysts were prepared by the hydrothermal synthesis of TiO<sub>2</sub> nanotubes followed by the deposition precipitation of CuO nanoparticles in alkaline conditions. We first prepared the TiO<sub>2</sub> nanotube support by the hydrothermal treatment of TiO<sub>2</sub> spherical particles in strong alkaline solution at 140°C. We then synthesized a series of CuO-TiO<sub>2</sub> catalysts by deposition precipitation at constant pH, with sodium carbonate as the alkali precipitating agent and different loading ratios of Cu to TiO<sub>2</sub> between 2% and 30 wt.%. We studied the morphological and structural properties of prepared nanocatalysts using standard physical techniques including SEM, EDX, TEM, TGA, XRD and XPS in order to understand the structure-property relationship and to optimize their catalytic activity. We carried out multiple catalytic CO oxidation cycles in a continuous flow fixed-bed reactor at low temperature range (25–300°C) and studied the catalytic activity of

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the different CuO-TiO<sub>2</sub> nanocatalysts and their stability under stream. We also studied the effect of shape of the TiO<sub>2</sub> support and the effect of the mole ratio of CuO loading on the CO conversion rates. The catalytic activity of the single counterparts of CuO and TiO<sub>2</sub> were measured for comparison. The experimental results revealed that the CuO nanoparticles supported on TiO<sub>2</sub> nanostructures exhibited higher activity and enhanced CO conversion rates at lower temperature, compared to un-supported CuO nanoparticles. The increased activity at lower activation temperature is probably due to the increased degree of dispersion of the active CuO phase on the TiO<sub>2</sub> support as concluded from the EDX mapping study. Moreover, the results showed that the correlation between the catalytic activity of CuO-TiO<sub>2</sub> nanocatalysts and both the shape and crystalline phase of the TiO<sub>2</sub> support. The CuO supported on TiO<sub>2</sub> nanotubes demonstrated enhanced CO conversion rates at lower temperature compared to that supported on TiO<sub>2</sub> nanospheres. In all samples the CuO-TiO<sub>2</sub> nanocatalysts calcined at 400°C exhibited the anatase phase of the TiO<sub>2</sub> nanotubes support and demonstrated higher activity. The results also showed that increasing the Cu to Ti ratio could lower the activation temperature needed for CO to CO<sub>2</sub> conversion probably due to the enhanced synergetic effect of the two mixed metal oxides. In addition, the XPS study of the CuO-TiO<sub>2</sub> composite oxide structure indicated high degree of oxygen deficiency in CuO-TiO<sub>2</sub> nanocatalysts with higher Cu to TiO<sub>2</sub> loading and this could result in CO oxidation rates. The prepared CuO-TiO<sub>2</sub> nanocatalyst demonstrated a high stability for CO oxidation for test periods of up to 5 h under stream at 200°C. The prepared CuO-TiO<sub>2</sub> nanocatalysts could have potential applications in hydrogen purification in fuel cell systems and for CO removal in carbon dioxide lasers and in air quality industries.

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