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One-pot solvothermal synthesis of mixed Cu-Ce-O_x nanocatalysts and their catalytic activity for low temperature CO oxidation

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ABSTRACT

One-pot solvothermal method has been developed for the synthesis of high-surface area Cu-Ce-O_x binary nanocatalysts with various Cu contents in the presence of oleylamine as capping agent. The obtained binary nanocatalysts were characterized by different techniques including XRD, H_2-TPR, TEM, BET and XPS. The influence of Cu contents on their catalytic performance for the CO oxidation was also studied. Our results revealed that the Cu-Ce-O_x binary nanocatalysts show the better catalytic activity compared to those of the conventional Cu-Ce-O_x, bare nano-CeO_2 and nano-CuO catalysts. This is ascribed to mutual interaction and synergistic effect between copper oxide species and cerium oxide. Among the obtained nanocatalysts, the Cu-Ce-O_x nanocatalyst with 5.6 wt% Cu (noted as 5.6-Cu-Ce-O_x) exhibits the best catalytic activity; the 50% CO conversion can be reached at 55 °C. H_2-TPR profiles show two reduction peaks at low and high temperatures for these catalysts, which could be attributed to the reduction of highly dispersed CuO on the CeO_2 surface and the bulk-like CuO species, respectively. Using the one-pot solvothermal synthesis developed in this study, 6–8 wt% Cu content is needed for the high catalytic activity for the CO oxidation at low temperature.

1. Introduction

Precious-metal based catalysts such as Au/TiO_2, Au/CeO_2 and Pt/SnO_2 have high activity for low temperature carbon monoxide (CO) oxidation; however, due to the high cost of precious-metals, the development of efficient catalysts based on non-precious metals for catalytic oxidations is highly desired [1,2]. Among them, Cu-Ce-O_x catalysts attract special interest owing to their low cost and remarkable activities [3] that can even be compared to the precious-metals based catalysts for various reactions, such as water gas shift (WGS) [4,5], preferential CO oxidation and the oxidations of methanol and methane [6,7]. The role of ceria is not only related to its so-called oxygen storage capacity (OSC): take up oxygen under oxidizing conditions and release it under reducing ones; but also to improve dispersion of the base metals [1–7]. In addition, the presence of oxygen mobility in the CeO_2 lattice is important for the catalysts, since the oxygen is available for CO oxidation. Sedmak et al. [8,9] showed that the lattice oxygen in CeO_2 was highly involved in the catalytic reaction; because the Cu-Ce-O_x catalyst can catalyze CO oxidation even without oxygen in the reaction gas. Furthermore, Liu and Flytzani-Stephanopoulos [3,10] reported that the Cu-Ce-O_x nanocatalysts exhibited catalytic activity superior to the conventional Cu-based catalysts for CO oxidation; and these catalysts were as active as Pt based catalysts. This is due to the fact that the high surface area of nanosized ceria provides exposed active Cu sites and high oxygen vacancies resulting in better catalytic activity. In our previous work [11], we reported that the porous copper-metal oxide solids assembled from pre-synthesized metallic cupper and metal oxide nanoparticles (TiO_2 or CeO_2) in which two distinct nanoparticles (metal and metal oxide) are mutually dispersed forming porous hybrid materials [11]. These materials showed better catalytic activity for CO oxidation than those of commercial noble metal catalysts (Pt/Al_2O_3) and the conventional copper based catalysts. This also implies that nano-oxide support such as CeO_2 and TiO_2 may not simply act as a support, but also may play a direct role in the catalytic process. The homogenous distribution of copper and ceria nanoparticles in the material as well as its nanostructure have been suggested to greatly influence on the catalytic activity of CO oxidation. Furthermore, the synergetic interaction between metal and metal oxide NPs at the nano-scale interface plays a crucial role in the catalytic activity owing to facile redox interplay between copper and cerium redox couples (Cu^{2+}/Cu^{+} and Ce^{4+}/Ce^{3+}) [7,11,12].

For the type of mixed metal oxide nanomaterials, a number of preparation methods are described in the literature, including co-precipitation [3,4], precipitation–deposition [13], impregnation [14], solution combustion [15], inert gas condensation [16], reverse microemulsion [15–17], flame spray pyrolysis [6] and surfactant-templated method [17,18]. These preparations produce powders formed by nanosized crystals with BET surface areas between 20

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