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A rapid microwave-assisted solution combustion synthesis of CuO promoted CeO_2–M_xO_y (M = Zr, La, Pr and Sm) catalysts for CO oxidation

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ABSTRACT

A series of copper oxide promoted CeO_2–M_xO_y (M_xO_y = ZrO_2, La_2O_3, Pr_2O_3 and Sm_2O_3) mixed oxides were synthesized by microwave-assisted solution combustion method using urea as the fuel and the respective metal nitrates as the precursors. The physico-chemical properties of the synthesized materials were analysed by BET surface area, X-ray diffraction (XRD), Raman spectroscopy, temperature programmed reduction/oxidation (TPR/TPO), X-ray photoelectron spectroscopy (XPS) and oxygen storage capacity (OSC) methods. XRD measurements confirmed the formation of solid solutions between ceria and the doped metal oxides in the presence of copper promoter. Raman measurements suggested defective structure of the mixed oxide solid solutions resulting in the formation of oxygen vacancies. The TPR/TPO studies revealed that the reduction behaviour of ceria depends on the type of metal dopant. XPS studies confirmed the presence of cerium in both Ce^{3+} and Ce^{4+} oxidation states in all mixed oxides. All the doped mixed oxides exhibited better CO oxidation activity compared to the undoped copper–ceria catalysts. Among various samples, ZrO_2 doped copper–ceria showed a high activity ($T_{1/2} \sim 378$ K) followed by samarium, praseodymium and lanthanum oxide doped samples, respectively. Significance of the combustion synthesis method has been addressed that include simplicity, flexibility and the control of different favourable factors.

1. Introduction

Carbon monoxide treatment by catalytic oxidation, such as water-gas-shift reaction (WGSR), preferential oxidation of CO in H_2 rich streams (PROX) and its simple oxidation either in the presence of O_2 or NO have received a great deal attention recently by the scientific community. The presence of CO in the atmosphere (100 ppm or above) causes severe environmental and human health problems [1–4]. The other important fields where the CO oxidation reaction addressed are the pollution control devices for vehicle exhaust purification, indoor air cleaning, solid oxide fuel cells, carbon dioxide lasers and so on [5,6]. In many cases, the noble metal-based catalysts have proven to be the highly effective candidates towards the CO oxidation [7–9]. However, various disadvantages of noble metals, for example, high cost, limited availability and sensitivity to catalyst poisoning have stimulated an extensive search for alternative catalysts [10,11].

In recent years, copper–cerium based catalysts have received much attention, because they are inexpensive, environmentally benign and can be prepared easily [12,13]. The synergism between copper and ceria shows a remarkable influence on various oxidation reactions, and exhibits comparable activity with that of noble metal catalysts [14–17]. Particularly, ceria is an important rare earth oxide extensively employed in heterogeneous catalysis due to its high oxygen storage/release capacity (OSC) and easily inter-convertible Ce^{4+}/Ce^{3+} redox couple [3,18–20]. However, pure ceria is not an ideal candidate for increasing practical applications owing to its limited OSC and poor thermal stability. Modification of ceria with other metals is a key issue to overcome the aforesaid drawbacks. Hence, several research efforts are going on towards doping of ceria with other isovalent/aliovalent cations. The isovalent cations frequently employed include Zr^{4+}, Hf^{4+}, Ti^{4+}, etc., and the aliovalent cations are La^{3+}, Sm^{3+}, $Mn^{2+/3+}$, $Fe^{2+/3+}$, Eu^{3+}, Tb^{3+}, Pr^{3+}, etc. In many cases, the redox properties and chemical reactivity of doped ceria have been enhanced compared to pure ceria [21–23]. Therefore, the synergism between copper and ceria as well as enhanced physico-chemical properties of doped ceria have inspired the researchers to design copper promoted ceria-based mixed oxides [24–27]. Papavasiliou et al. [28,29] have investigated the effect of various dopants on copper–ceria for methanol steam reforming and WGSR. It was found that dopants such as Zn and Sm show a positive effect, whereas Zr, La, Gd and Y exhibit negative effect on the activity of copper–ceria catalysts.

Numerous research efforts have been undertaken to prepare Cu promoted CeO_2-based mixed oxides, of which solution-based methods are of particular interest in the laboratory and industry.

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