

CeO₂ with different dimensional nanostructures for low temperature catalytic oxidation of toluene

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Abstract: The zero-dimensional (0D) hollow-sphere, one-dimensional (1D) hollow-tube and 2-dimensional (2D) sheet CeO₂ nanostructures were synthesized. Their catalytic properties for toluene oxidation were evaluated, and the effects of nanostructure dimension on the activities of the CeO₂ were investigated. The activities of the catalysts decreased in the order of 2D > 0D > 1D-CeO₂. The higher amount of surface adsorbed oxygen species is, the higher is the activity for toluene oxidation, identifying the key role of oxygen vacancy in this reaction via the activation of molecular oxygen over nanoceria.

Keywords: CeO₂, nanostructure, toluene.

1. Introduction

Ceria nanomaterials have attracted much attention in recent years, engineering the shape at nanometer level can radically alter their catalytic performances. Despite many of reported catalysts are polycrystalline with rather ill-defined morphologies, it is well known that the precise control of structure can modify the redox of Ce⁴⁺/Ce³⁺ ions, changing the oxygen vacancy behavior of ceria, which strongly affects their catalytic performance, in several reactions like CO, soot and volatile organic compounds (VOCs) oxidation and so on.¹ Remarkable progress has been made in the investigation of morphology-dependent properties of the catalysis behavior for CeO₂ nanostructured materials. However, the reported works have been mostly restricted to 0D polyhedral nanocrystals such as cubes, spheres, and octahedra, or 1D nanostructures, e.g. nanorods, nanowires, and nanotubes.²

In recent years, 2D nanostructured materials, namely nanosheets, have received a tremendous amount of attention for such sheets possess the features of relatively disperse active sites and a high ratio of surface active sites, and it is crucial to develop facile and robust synthesis method. Herein, we prepared ceria nanosheets by precipitation method and then evaluated the toluene oxidation activity with difference of nano-structural dimension of ceria.

2. Experimental

In this study, toluene combustions were carried out in a continuous flow quartz tube ($\Phi = 10.0$ mm) micro-reactor with 100 mg catalyst (40-60 mesh). The volumetric composition of the reactant mixture was 1000 ppm toluene in air with 100 ml/min. Toluene was generated and injected using a bubbler. At the beginning of each test, the catalyst was pretreated in the real reactant mixture for 2 h in order to overcome the over-estimation of toluene catalytic combustion conversion caused by adsorption of toluene. The performances at steady states (typically after 0.5 h) were used for discussion. The concentrations of the reactants and products were monitored on line by a gas chromatograph by double flame ionization detectors (FID), where the CO₂ were converted into CH₄ with a methanizer nickel reactor. All results showed there were only CO₂ in the downstream gas products, indicating the toluene molecules were completely decomposed over these catalysts. All the evaluated samples were calcined at 500 °C for 3 h in air.

3. Results and discussion

Figure 1 clearly shows that the 0D hollow-sphere, 1D hollow-tube and 2D sheet CeO₂ nanostructures were successfully fabricated. Then, we evaluate the catalytic performance of the as-synthesized CeO₂ nanostructures, they were employed to degrade toluene in air, as displayed in Figure 2. The difference of the catalytic performance for toluene oxidation on the three catalysts is really obviously, it can be seen that the

activity of 2D sheet CeO₂ nanostructures significantly better than that of nanosphere and nanotube. A more detailed analysis will be shown in the meeting.

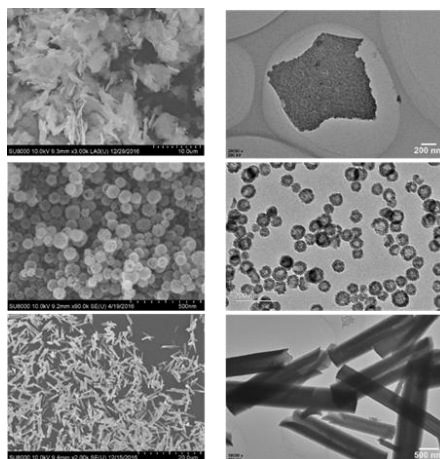


Figure 1. SEM and TEM image of nanosheet, nanosphere and nanotube CeO₂.

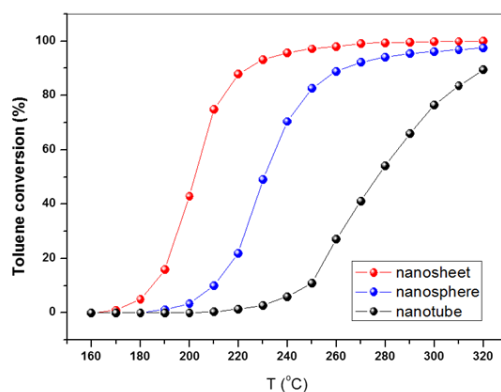


Figure 2. Catalytic activity for toluene oxidation versus reaction temperatures.

4. Conclusions

The 0D hollow-sphere, 1D hollow-tube and 2D sheet CeO₂ nanostructures were prepared for toluene oxidation, the activities of the catalysts decreased in the order of 2D > 0D > 1D-CeO₂. The amount of oxygen vacancy in these materials were highly related with their toluene oxidation behavior.

References

1. Trovarelli A, Llorca J, ACS Catal, 7 (2017) 4716.
2. Montini T, Melchionna M, Monai M, et al. Chem. Rev. 116 (2016) 5987.