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Role of SiO₂ in synthesis of SiO₂-supported CeO₂ composites

Ning Fang^{a,*}, Yongyi Ding^a, Chengbao Liu^{b,c,d}, Zhigang Chen^{a,b,c,d,*}

^a School of Material Science and Engineering, Jiangsu University, Zhenjiang 212013, China

^b School of Chemistry, Biology and Materials Engineering, Suzhou University of Science and Technology, Suzhou 215009, China

^c Jiangsu Key Laboratory for Environment Functional Materials, Suzhou 215009, China

^d Jiangsu Collaborative Innovation Center of Technology and Material for Water Treatment, Suzhou 215009, China

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ABSTRACT

Pure and SiO₂-supported CeO₂ samples were prepared by Ce(NO₃)₃ decomposition, precipitation, and sol-gel methods in an attempt to study the role of SiO₂ in the synthesis of these materials. During synthesis process, SiO₂ support uniformly adsorbed cerium ions in aqueous solution, preventing nucleation and crystal growth of CeO₂ during the subsequent water evaporation and calcination steps. Uniform adsorption and inhibition were enhanced by NH₄⁺ and, to a larger extent, C₅H₇O₅COO⁻. Despite the dispersion of cerium ions on SiO₂ reduced the temperature at which CeO₂ was formed, crystal size and crystallinity of CeO₂ in composites were significantly lower than that of pure CeO₂ sample prepared by the same synthesis method and at the same temperature. Composites were quite stable upon increasing the temperature from 400 to 800 °C. Visible light absorption, reduction, and photocatalytic activity characteristics of CeO₂ were improved upon dispersion on SiO₂. This work can help synthesize supported oxides with high activity and thermal stability.

1. Introduction

In recent years, ceria (CeO₂) has been widely used as active component or support in several applications including three-way catalysts for automotive exhaust treatment [1–3], removal of organic compounds [4,5], cracking of heavy oils [6], and water-gas shift processes [7,8], among others. The unique redox and high oxygen storage/release capacity of CeO₂ through the Ce⁴⁺/Ce³⁺ couple play a major role in these applications [2,4]. However, pure CeO₂ suffers from poor thermal stability [9]. Thus, the high oxygen mobility and catalytic activity of ceria significantly decrease at elevated temperatures [10,11]. One effective approach to overcome this issue involves the utilization of thermally stable inert supports [12,13], which provide CeO₂ with the necessary thermal stability without negatively affecting its special features [13,14]. In this sense, silica (SiO₂) is one of the most widely used supports since it possesses excellent chemical inertness, thermal stability, high specific surface area, and low cost [15]. Thus, SiO₂ allows to improve the dispersion and catalytic activity of most deposited active oxides [12,15].

There are two ways of preparing CeO₂-SiO₂ mixed oxides namely, co-crystallization of cerium and silicon ions [14,16,17] and adsorption of cerium ions on the surface of SiO₂ [10,18–20]. In the former method, intermediate phases such as Ce_{9.33}(SiO₄)₆O₂ can be formed to enhance the textural and thermal stability of the composite [21,22]. Despite

these good characteristics, the adsorption method is more widely used since the structure of SiO₂ can be easily controlled, allowing CeO₂ to be uniformly loaded on its surface and to participate in the catalytic reaction.

Several studies have been carried out on the properties of SiO₂-supported CeO₂, which are affected by the composition of the CeO₂ precursor, the structure of the SiO₂ support, the Ce/Si ratio, and the synthesis method. Bensalem et al. [23] prepared SiO₂-supported CeO₂ composites and found ceria particles of lower size when using C-acetylacetonate as a precursor instead of Ce-nitrate (1–3 vs 5–9 nm). Khalil et al. [24] found that cerium(IV) ammonium nitrate resulted in better ceria dispersions as compared to cerium(IV) isopropoxide, and the crystal size of ceria decreased with the Ce content. Cracium [10] prepared amorphous and crystalline CeO₂ materials supported on SiO₂ by incipient wetness and grafting methods, respectively. Strunk et al. [18] investigated the structure of ceria deposited on meso-porous silicas such as MCM-41 and SBA-15. Despite the large number of works, the role of the SiO₂ support in the composite synthesis has not been sufficiently explored. This knowledge is required to precisely control the structure and properties of the composite.

In this work, pure and SiO₂-supported CeO₂ samples were prepared by Ce(NO₃)₃ decomposition, precipitation, and sol-gel methods. The formation and structural changes of CeO₂ during the preparation were investigated by thermogravimetry and differential thermogravimetry

* Corresponding authors.

E-mail addresses: ningfang.ustb@hotmail.com (N. Fang), czg@ujs.edu.cn (Z. Chen).

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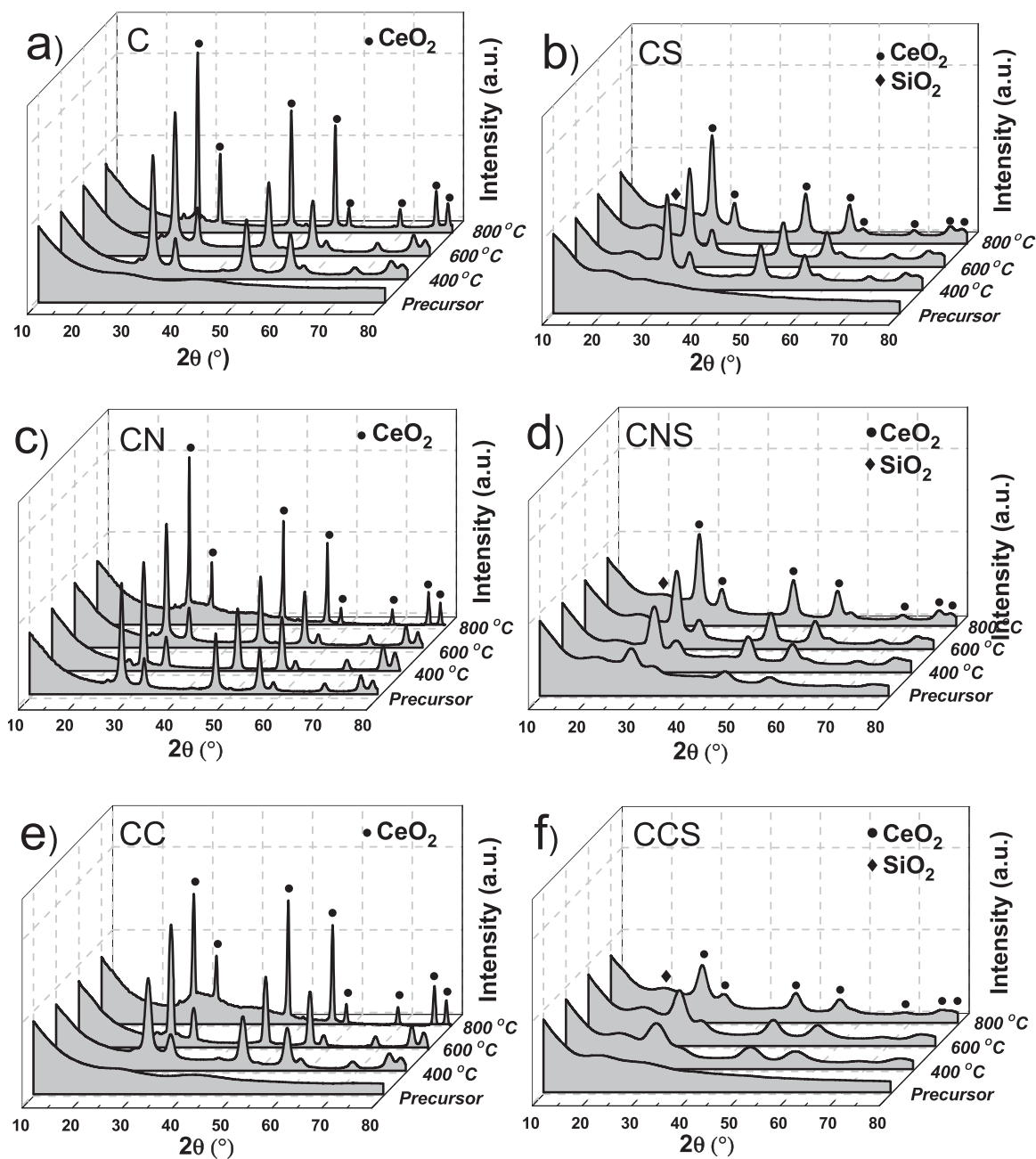


Fig. 1. XRD patterns of: a) C, b) CS, c) CN, d) CNS, e) CC, and f) CCS samples prepared at different calcination temperatures.

coupled with differential scanning calorimetry (TG-DTG-DSC) and X-ray diffraction (XRD) in order to study the role of SiO₂ in the synthesis process. The effect of the SiO₂ support on the light absorption, reduction, and photocatalytic activity characteristics of CeO₂ was also evaluated.

2. Experimental

2.1. Synthesis of samples

Pure CeO₂ samples were prepared by decomposition of Ce(NO₃)₃·H₂O, precipitation, and sol-gel methods and labeled as C, CN, and CC, respectively. The C precursor was obtained by drying Ce(NO₃)₃·H₂O at 60 °C for 24 h. In the precipitation method, Ce(NO₃)₃·H₂O was dissolved in deionized water to form a 0.1 M solution. Dilute aqueous ammonia was subsequently added drop-wise into the solution upon vigorous stirring until the precipitation was complete (pH

= 10). The solvent was subsequently evaporated upon stirring in a water bath at 60 °C for 12 h to obtain the CN precursor. In the sol-gel method, Ce(NO₃)₃·H₂O and C₆H₈O₇·H₂O (citric acid) at a 1:1 M ratio were dissolved in distilled water upon stirring until a stable sol was formed. The sol was aged in air until a gel was formed. The gel was dried at 60 °C for 12 h to obtain the CC precursor. Finally, the C, CN, and CC precursors were calcined in air at 400, 600, and 800 °C for 4 h, respectively.

SiO₂-supported CeO₂ composites (CS, CNS, and CCS) were prepared by the three synthesis methods. The SiO₂ support was impregnated with a Ce(NO₃)₃ solution under ultrasound for 30 min for the decomposition method. Dilute aqueous ammonia (for the precipitation method) or citric acid (for the sol-gel method) solutions were subsequently added. The support consisted of a finely ground (800–1000 mesh) silica-gel after calcination in air at 800 °C for 8 h. The Ce/Si molar ratio was fixed to 0.1 in all composites. The other parameters and steps were similar to those used during the preparation of pure CeO₂.

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