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Recent advances of cerium oxide nanoparticles in synthesis, luminescence and biomedical studies: a review

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Abstract: Nanostructured cerium oxide (CeO_2) commonly known as nanoceria is a rare earth metal oxide, which plays a technologically important role due to its versatile applications as automobile exhaust catalysts, oxide ion conductors in solid oxide fuel cells, electrode materials for gas sensors, ultraviolet absorbents and glass-polishing materials. However, nanoceria has little or weak luminescence, and therefore its uses in high-performance luminescent devices and biomedical areas are limited. In this review, we present the recent advances of nanoceria in the aspects of synthesis, luminescence and biomedical studies. The CeO_2 nanoparticles can be synthesized by solution-based methods including co-precipitation, hydrothermal, microemulsion process, sol-gel techniques, combustion reaction and so on. Achieving controlled morphologies and enhanced luminescence efficiency of nanoceria particles are quite essential for its potential energy- and environment-related applications. Additionally, a new frontier for nanoceria particles in biomedical research has also been opened, which involves low toxicity, retinopathy, biosensors and cancer therapy aspects. Finally, the summary and outlook on the challenges and perspectives of the nanoceria particles are proposed.

Keywords: cerium oxide; nanoparticle; controlled synthesis; luminescence; biomedical; rare earths

Generally, CeO_2 crystallizes in the fluorite crystal structure with space group *Fm*3*m*[1]. In the ideal cell of CeO_2, the structure consists of a face-centered cubic unit cell of cations with anions occupying the octahedral interstitial sites. In this structure (Fig. 1)[2,3], each cerium cation is coordinated by eight nearest-neighbor oxygen anions, while each oxygen anion is coordinated by four nearest-neighbor cerium cations[4,5]. When cerium cations are replaced with lower valent elements (M^{2+}/M^{3+}) to form the solid solutions, the lattice oxygen atoms in CeO_2 are removed and oxygen vacancies are subsequently produced to keep the electric neutrality. Actually, the nanoceria crystal usually exhibits a few defects due to the co-existence of Ce^{4+} and Ce^{3+} ions. Based on the thermodynamic principle, the generation of defects will result in the increase of entropy of the system, and therefore reduce the Gibbs free energy and improve the stability of material[3].

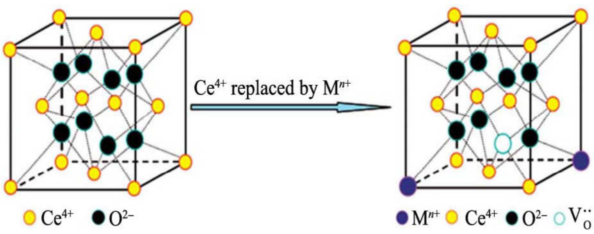

Fig. 1 Unit cell of CeO_2 and variation of unit cell during doping process[3]

Nanoceria and rare earth doped ceria-based nanostructures have been widely investigated as important functional materials due to excellent properties such as superior thermal and chemical stability, high ionic conductivity, good oxygen storage or release capacity and strong UV absorption[6–9]. However, nanoceria has little or weak luminescence, which limits its uses in high- performance luminescent devices and biomedical areas[10–12]. Recently, the study of nanoceria has been paid considerable attention in the luminescence and biomedical fields. Herein, the recent advances of nanoceria particles in synthesis, luminescence and biomedical studies are reviewed.

1 Synthesis

The controlled synthesis of the nanoceria often requires more specialized approaches as compared with traditional solid-state reactions (SSR). Some solution-based techniques have been developed for this purpose and mainly focus on the co-precipitation[13,14], hydrothermal or solvothermal[15,16], microemulsion process[17], sol-gel and solution combustion synthesis (SCS).

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In general, the kinetic control (temperature and concentration), nucleating seeds and the activation energy modulations of surfaces by surfactant molecules are important factors to adjust their anisotropic growth. Some typical examples to prepare nanoceria materials are described as follows:

1D nanoceria materials, including nanowires, nanorods, and nanotubes have been studied owing to their new physical performance and potential applications. To achieve 1D nanoceria, the crystal growth directions should be controlled with different mediators, such as surfactant, solvent, reaction temperature and concentration. Sun et al., for the first time, obtained polycrystalline CeO_2 nanowires by using sodiumbis(2-ethylhexyl) sulfosuccinate surfactant as a structure-directing agent[18]. The typical scanning electron microscopy (SEM) and transmission electron microscopy (TEM) images of the samples are given in Fig. 2[18], which shows clearly that they are nanowire-structure. The CeO_2 nanowires are around 30–120 nm in diameters and 0.2–5 μm in lengths, and the nanowire consists of many tiny interconnected nanocrystallites of about 7 nm in size. This structure perhaps enable the gas to access all the surfaces of nanoceria contained in the device unit.

Li's group synthesized single-crystalline CeO_2 nanorods with well-defined crystal planes by a facile solution-based hydrothermal method[19]. Zhou et al. synthesized CeO_2 nanotubes with large cavities and thin walls via a simple oxidation-coordination-assisted dissolution process of $Ce(OH)_3$ nanotubes/nanorods[20].

Recently, 2D nanoceria materials including nanosheets and nanoplates have attracted great attention due to their intriguing properties. Xia et al. synthesized single-crystalline ceria nanosheets with a thickness of about 2.2 nm and lateral dimension up to 4 μm by a simple aqueous route, as can be seen from Fig. 3[21]. Their synthetic protocol involves the slow, continuous addition of cerium(III) nitrate into an aqueous solution containing 6-aminohexanoic acid using a syringe pump. They found that the nanosheets were formed through 2D self-organization of initially formed small ceria nanocrystals, followed by an in situ recrystallization process.

Additionally, mesoporous ceria has also exhibited tremendous potential in catalytic fields because of its large surface area and the improved dispersibility of active secondary components[22]. Tong's group developed a facile and low-cost approach for the preparation of porous ceria and Gd-doped ceria foam nanostructures in high yield, and the corresponding SEM images are given in Fig. 4. Applying an electrochemical deposition route at room temperature, the as-prepared porous Gd-doped CeO_2 has shown a remarkable enhancement of optical and magnetic properties[23].

Sol-gel technique is an effective method to synthesize 3D nanoceria-based materials. Jeong et al. synthesized $CeO_2:Eu^{3+}$ nanoparticles by a Pechini-type sol-gel proc-

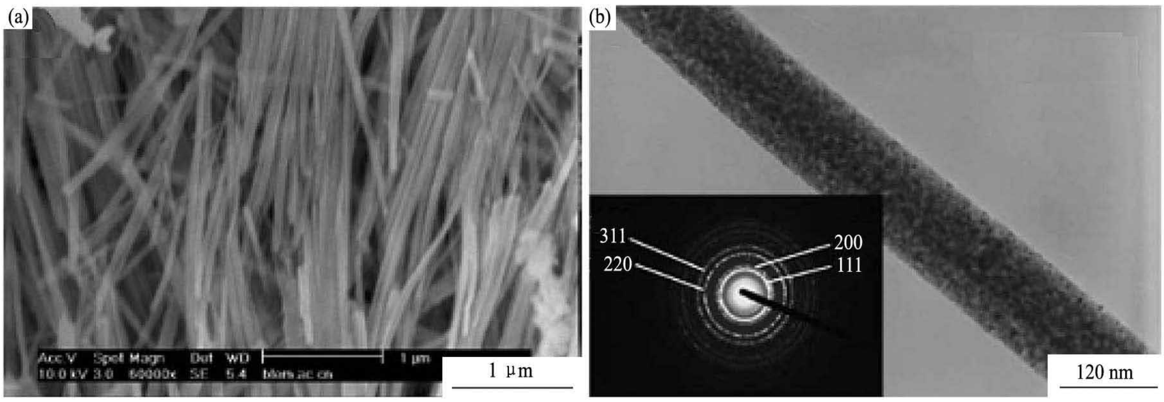

Fig. 2 SEM (a) and TEM (b) images of the CeO_2 nanowires

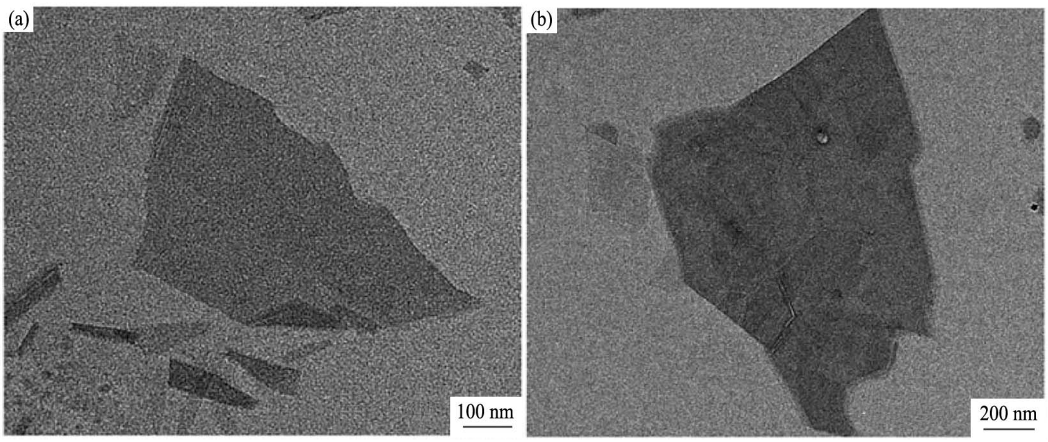

Fig. 3 TEM images of ceria nanosheets with different sizes (a and b)

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