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Hydrothermal assisted synthesis and hot-corrosion resistance of nano lanthanum zirconate particles

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Abstract

The nano $La_2Zr_2O_7$ (LZ) particles with pyrochlore microstructure were successfully synthesized from a mixture of $La(NO_3)_3$, $Zr(NO_3)_4$ and $C_{19}H_{42}BrN$ (CTAB) using hydrothermal assisted (HTA) synthesis which consists of two steps: hydrothermal treatment and calcination. Transmission electron microscopy (TEM), scanning electron microscopy (SEM), Fourier transform infrared spectra (FT-IR) and X-ray diffraction (XRD) spectroscopy were employed to study morphologies and phase compositions. The results suggest that HTA process led to very rapid synthesis of nano LZ compared to the conventional solid reaction process. The particles produced by HTA synthesis have cubic shape and the distribution of its grain size is from 10 nm to 30 nm. The present work demonstrates that the nano $La_2Zr_2O_7$ produced via HTA synthesis which have better hot-corrosion resistance is an ideal material for thermal barrier coatings.

Keywords: Hydrothermal assisted synthesis; $La_2Zr_2O_7$; Nanoparticle; Hot corrosion

1. Introduction

In recent years, nanopyrochlore type oxide materials have received considerable attention due to its wide industrial application [1,2]. Pyrochlore type oxide (cubic, Fd3m) can be usually expressed by the general formula $A_2B_2O_7$ (A and B are trivalent and tetravalent metal ions). Pyrochlore type of rare earth zirconates ($Ln_2Zr_2O_7$, Ln denotes rare earth) has gained intense interest for their important technological applications. These zirconates exhibit low thermal conductivity, high melting point, thermal expansion coefficient [3,4], excellent photocatalytic activity [5], host materials for luminescence centers [6] and good dielectric property [7]. In view of such excellent properties, $Ln_2Zr_2O_7$ is widely used as thermal barrier coatings (TBCs), possible host for radioactive wastes and surplus actinides and solid electrolytes in high-temperature fuel cells [8]. In the series of the $Ln_2Zr_2O_7$, the $La_2Zr_2O_7$ (LZ) is the typical compound and has been widely used in the thermal barriers research and it has exhibited excellent thermal properties such as low thermal conductivity, high thermal stability and so on [9].

In order to obtain fine LZ crystalline particles which can be used for plasma spraying, various methods have been proposed. As we know, the LZ has been successfully prepared via the traditional high temperature solid-state reaction [10], but the heating temperature is almost above 1700 K and the average size of products may reach to a few microns. In order to obtain LZ coatings with better property, the nano LZ particles were widely synthesized by many methods such as co-precipitation [4], sol–gel [11], stearic acid combustion method [12], and salt-assisted combustion [13]. However, the particles prepared by the above methods were irregular in shape and inhomogeneous in size distribution. In the fabrication process of nanomaterials, the hydrothermal technique is

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promising, which can promote the reaction processing for the system activation has increased under the high temperature and high pressure subcritical conditions, and which is mentioned to have great potential for the near-room temperature manufacture of nanoparticles [14]. The main advantages of this method are related to the homogeneous nucleation processes and extra small grain size due to lower sinter temperature or the elimination of calcination step [15].

However, it is difficult to synthesis LZ only using the hydrothermal technique due to the slow reaction rate. After the hydrothermal treatment, a moderate heating at lower temperature could accelerate the solid reaction and control the particles size.

In the present works, nanocrystalline LZ particles were synthesized via the hydrothermal-assisted (HTA) method. The parameters in processing technique of LZ were be optimized, and properties of LZ particles were characterized in this paper. The study is also to evaluate the thermal corrosion properties of the as-prepared LZ produced by different methods.

2. Experimental procedure

2.1. Synthesis of nano LZ particles

The LZ particles with nanosize were prepared by the hydrothermal-assisted (HTA) method including two steps: hydrothermal treatment and high temperature solid reaction. The $La(NO_3)_3 \cdot 6H_2O$ (AR, Tianjin Kemiou Chemical Reagent Co., Ltd.) and $Zr(NO_3)_4 \cdot 5H_2O$ (AR, Tianjin Kemiou Chemical Reagent Co., Ltd.) were used as the precursors of the La^{3+} and Zr^{4+}, respectively. CTAB ($C_{19}H_{42}BrN$) (AR, Tianjin Kemiou Chemical Reagent Co., Ltd.) was used as a cationic surfactant. NaOH (AR, Tianjin Kemiou Chemical Reagent Co., Ltd.) solution was acted as a pH regulator.

The fabrication procedures are shown in Fig. 1. 10 ml 0.5 mol/L $La(NO_3)_3$ solution and 10 ml 0.5 mol/L $Zr(NO_3)_4$ solution were mixed in the vessel with the magnetic stirrer at room temperature, in which the 2 wt% surfactant CTAB was blended. The pH value of the mixture was regulated to 7 by dropwise adding the 0.1 mol/l NaOH solution. The mixture was added into the hydrothermal reactor, and the reactor was heated in the drying oven under certain temperature conditions for 24 h. Then it was cooled to the room temperature, the suspension mixture was taken out and centrifuged. The solid sediments were dried in the vacuum drying oven and ground to the fine particles in the agate mortar. Finally, the particles were calcined for 2 h in the electric furnace.

The particles produced by the hydrothermal-assisted method were denoted as LZ-HTA, and in comparison, the particles produced only via high temperature calcination were marked as LZ-UHTA.

2.2. Hot corrosion resistance tests

Hot corrosion resistance tests on the LZ-HTA and LZ-UHTA particles were performed simultaneously. The V_2O_5 was used as the hot corrosion agent. The mixture consisting of V_2O_5 (10 wt%) and LZ particles (90 wt%) was heated at 1473 K in an air electric furnace. Following a 2-h dwell time, the specimens were furnace-cooled down to room temperature.

2.3. Characterization of LZ

The phase structure of the as-synthesized LZ was identified by the Fourier transform infrared spectra (FT-IR, model:

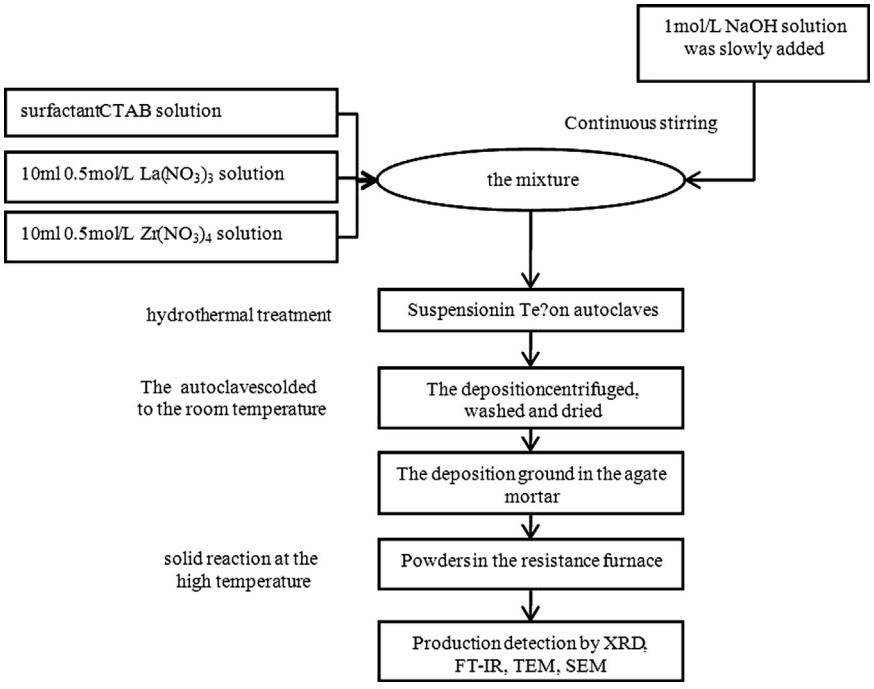

Fig. 1. The fabrication process of the nano LZ particles.