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Synthesis and Crystallization Behavior of 3 mol% Yttria Partically Stabilized Zirconia (3Y-PSZ) Nanopowders by Microwave Pyrolysis Process

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Abstract

A crystalline Nano powders of 3 mol% yttria-partially stabilized (3Y-PSZ) has been synthesized using $ZrOCl_2$ and $Y(NO_3)_3$ as raw materials by microwave pyrolysis with a TE666 resonant mode at 700-900°C. The frequency of the microwave was 2.45 GHz with the maximum power of 10 KW, and a hybrid heating structure was used with insulation of porous mullite and SiC aided heaters. For comparison, conventional heating was performed in air at 750°C for 20 min. The as-synthesized products were characterized by SEM and TEM images, XRD patterns. It was found that microwave energy promotes the conversion of tetragonal ZrO_2 (t-ZrO_2) to monoclinic ZrO_2 (m-ZrO_2) phase compared with conventional pyrolysis. TEM images showed that highly dispersed 3Y-ZrO_2 powders with ~23 nm in size were obtained by microwave pyrolysis at 750°C for 20 min.

Keywords: 3Y-ZrO_2 powders; Microwave pyrolysis; Highly dispersed nano powders

Introduction

Zirconia (ZrO_2) is an important material possessing many excellent properties, including high melting point, high hardness and strength, high fracture toughness, low thermal conductivity, high chemical stability, ionic conductivity, and excellent corrosion and abrasion resistance [1,2]. It is thus extensively used in many important areas, e.g., functional ceramics, high-temperature and corrosion resisting components, abrasive and insulating material, dielectric element, catalysts, and ion exchanger [3,4].

To date, several techniques have been developed to prepare ZrO_2 nanoparticles, including sol-gel, flame spray, combustion, glycothermal process, hydrothermal processing, precipitation and other techniques [5-10]. Unfortunately, these techniques all suffer from various disadvantages, such as strong agglomerates, difficulty in particle size control, complex drying procedures, requirement of high energy and/or long reaction time, and low production efficiency. To overcome these drawbacks, it is necessary to develop other alternative techniques.

Microwave method has recently attracted an increasing amount of interest [11-15] owing to the advantages, such as cost-effective, energy efficient, rapid and convenient method of heating, and results in higher yields in shorter reaction times. In this work, 3Y-ZrO_2 Nano powders were prepared by microwave pyrolysis combined with a co-precipitation process using $ZrClO_2{\cdot}8H_2O$ as the starting material, and NH_4OH as the mineralizer.

Experimental Procedure

Preparation of 3Y-ZrO_2 powders

Commercially available zirconium oxychloride octahydrate ($ZrClO_2{\cdot}8H_2O$, purity: 99.2%, Zibo Huantuo Chemical Co. Ltd., Shandong, China), yttriumnitrate hexahydrate ($Y(NO_3)_3{\cdot}6H_2O$, A.R., Tianjin Guangfu Fine Chemical Research Institute, Tianjin, China), and ammonia solution (NH_4OH, A.R., Xilong Chemical Co., Ltd., Guangdong, China) were used in the preparation of the precursor. $ZrClO_2{\cdot}8H_2O$ and $Y(NO_3)_3{\cdot}6H_2O$ were used as received and dissolved in DI water. The concentration of zirconium ion was 1.0 mol/L, to which 3 mol% Y $(NO_3)_3{\cdot}6H_2O$ was added. 1 M ammonia solution was added dropwise in given solution with continuous stirring, adjusting its Ph. value at 12-13. After co-precipitation, the precursor solution was filtered and washed with ethanol repeatedly until no Cl^- was detected in the filtrate by an $AgNO_3$ solution. The resulting precursor powder was oven-dried at 80°C for 24 h. Subsequently, the dried powder was kept in a microwave chamber with the resonant mode of TE666 (WXD20S-07, Nanjing Sanle Microwave Technology Development Co., Ltd., Jiangsu, China) at 700-900°C for 20 min. The frequency of the microwave oven was 2.45 GHz with the maximum power of 10 KW. The temperature was monitored by using an infrared radiation thermometer (OI-T6I2-B-1-type, GOIDSUN, USA) with initial display of 700°C. A thermal insulation structure based on a hybrid heating mode was well designed with the wall material of porous mullite and aided heaters of SiC rods. For comparison, 3Y-ZrO_2 was also prepared via conventional pyrolysis at 750°C for 20 min.

Characterization of 3Y-ZrO_2 powders

Phases in the as-prepared product powders were identified by powder X-ray diffraction (XRD) analysis (XD-3, Persee, China) with Cu K_a radiation (λ=1.5406 Å). Morphologies and microstructures were observed by using a field emission electron microscope (SEM) (JSM-7001F, JEOL, and Japan) and a transmission electron microscopy (HRTEM) (Tecnai G2 F20, Philips Co. Holland).

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Results and Discussion

Microwave pyrolysis behavior

It is known that microwave absorption strongly depends on the dielectric loss factor of the material of interest [16]. At low temperature, ZrO_2 precursor cannot effectively absorb microwaves. However, with the help of hybrid heating by SiC aided heaters, heat is transferred to the precursor in the low temperature region, heating at this stage is analogous to conventional processes. After reaching the critical temperature, ZrO_2 precursor couples with the electromagnetic field and a higher heating rate is obtained due to the increased dielectric loss factor [17]. This change is consistent with the heating curve shown in Figure 1. In Figure 1, the temperature was well monitored by manual control of input powder. Owning to the rapid sintering characteristics of microwave sintering, it took only 30 min to reach the temperature of 750°C. It was observed that an efficient forward power profile requires a high power initial segment, the reflected power increases synchronously with input power. After about 4 minutes, the reflect power was suitably reduced; thermal runaway could be prevented albeit quite fast temperature increase to 750°C. After about 35 minutes, the input power of the system decreased as well as reflected power concomitantly decreased, but the temperature was kept at 750°C, which was due to selective absorbing phenomena, which is the unique heating feature of microwave sintering.

The DTA/TG curves of the 3Y-PSZ precursor amorphous powders at heating rate 10 k/min in air are shown in Figure 2. An endothermic peak at about 94°C is accompanied with a weight loss of 10.5% which is attributed to the evaporation of water. The exothermic peak at 305°C is attributed to the dehydration of precursors. The second exothermic peak at 450°C due to the formation of the tetragonal phase of ZrO_2 in the 3Y-PSZ freeze dried precursor powders.

X-ray diffraction analysis

As shown in Figure 3, the observed diffraction peaks at 2θ=30.2°, 35.0°, 50.4°, 58.9°, and 62.9° are associated with -111, -200, -220, -311, and -222 plane of t-ZrO_2 (JCPDS No. 50-1089). The peaks are at around 28.2 and 31.4 correspond to the (-111) and -111 planes of m-ZrO_2 (JCPDS No. 37-1484). According to the XRD analysis, both the 3Y-ZrO_2 powders obtained by microwave pyrolysis and conventional pyrolysis are a mixture of monoclinic and tetragonal phases. As shown in Figure 3a, the metastable tetragonal phase is the main phase in the powders by CS method when the heating temperature is less than

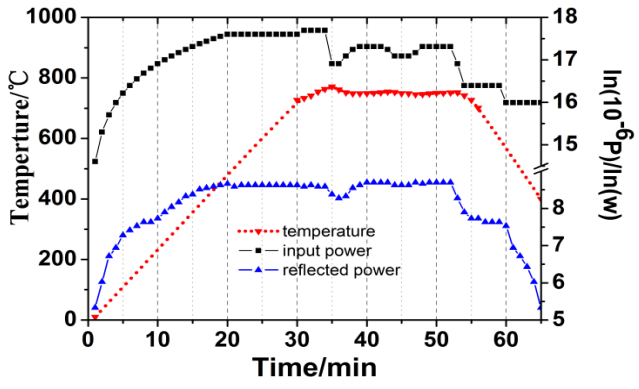

Figure 1: Heating cycle and power variation during microwave pyrolysis of ZrO_2 precursor.

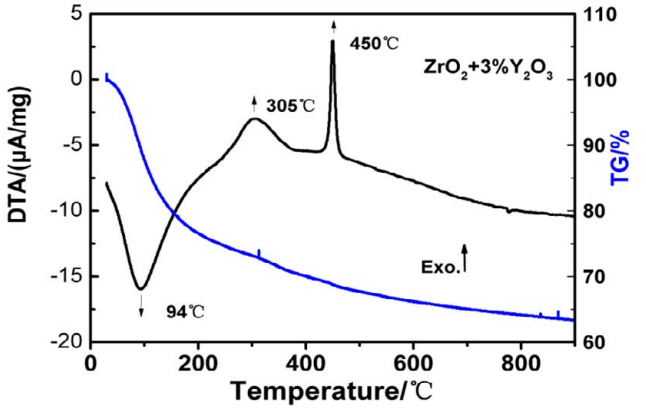

Figure 2: DTA/TG curves for 3Y-PSZ precursor amorphous powders at heating rate 10 k/min in air.

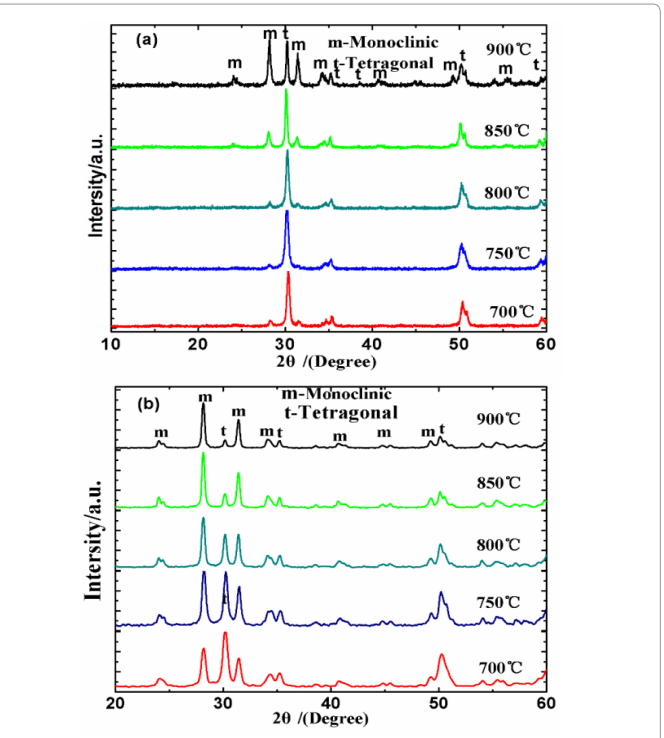

Figure 3: X-ray diffraction patterns of 3Y-ZrO_2 powders synthesized at different temperature by (a) conventional sintering (CS), (b) microwave pyrolysis (MP).

900°C, whereas the monoclinic phase of zirconia appears when the heating temperature is enhanced to 900°C, the prepared powder is a mixture of m-ZrO_2 and the t-ZrO_2, which is the same with the reference [18]. Compared with the CS method, the obtained powders by the MP method are multi-phase, and m-ZrO_2 with t-ZrO_2 are coexisted at every heating temperature, as shown in Figure 3b. It is found that the intensity of the tetragonal phase reflection peaks is greater than the monoclinic phase peaks at 700°C, the relative peaks of tetragonal phase decreased with increasing calcinations temperature, which means the m-ZrO_2 content increased while the t-ZrO_2 content decreased. This results show that the microwave energy can accelerate the formation of m-ZrO_2 phase.

Morphology of ZrO_2 powders

The surface morphology and particle sizes of the prepared ZrO_2 were examined by SEM images, as shown in Figure 4. Figure 4a shows that ZrO_2 powders have been partially crystallized and some residue of precursor coexist with ZrO_2 powders. Powders sintered at 750°C show fine crystalline and high dispersity with uniform particle size, as shown in Figure 4b, the average particle size of ZrO_2 powders is found to be less than 25 nm. In Figure 4c and 4d, the images of the zirconia powder obtained at 800°C and 850°C. It can be clearly seen that there is an appreciable formations of agglomeration. As the temperature further increases, non-uniform powders with agglomeration and abnormal growth of crystalline grain are observed from Figure 4d and 4e. Therefore, it is clearly understood that in the MP method, the molecular dipoles are induced to oscillate by microwave. This oscillation caused a higher rate of molecular collision which generates enormous amounts of heat. Consequently, the temperature distribution is homogeneous and is transferred to the materials interior, making the ZrO_2 particles synchronous growth, and with high dispersity.

The sample sintered by conventional method at 750°C shows bigger particles with serious agglomeration in Figure 3f. Other samples obtained at different temperatures by conventional sintering method present the similar phenomenon (not shown here). There is a temperature gradient between the heat source and the mass to be heated. Thus, during the heating process, the temperature distribution is not homogeneous and cannot be transferred to the materials interior, but spreads more to the particle surface. Consequently, an increase in the size of the crystals takes place owing to solid-state diffusion.

To provide further evidence for the formation mechanism, TEM analysis was carried out only for MP samples. A TEM image of ZrO_2 obtained at 750°C is presented in Figure 5a and 5b, indicating that the nearly spherical nano-crystals are uniformly formed. Moreover, the average size of Nano crystallites obtained from the SEM is in a relative agreement with the TEM studies which show the size in the range of 15-30 nm, the average size was 23 nm. The high-resolution TEM image of nanoparticles in Figure 5b shows that the nanoparticles are highly crystalline. The marked lattice fringes correspond to (-111) plane in m-ZrO_2 with a d-spacing of 3.1 Å.

Conclusion

The 3Y-ZrO_2 Nano sized powder prepared by microwave pyrolysis was thoroughly investigated. Optimized microwave pyrolysis condition is around 750°C for 20 min. The powders were characterized by a narrow particle size distribution, high dispersive and the average size was 23 nm. The microwave pyrolysis YSZ powders consisted of tetragonal and monoclinic phase. In the microwave field, the stability of tetragonal phase was weak and debased during the formation of ZrO_2.

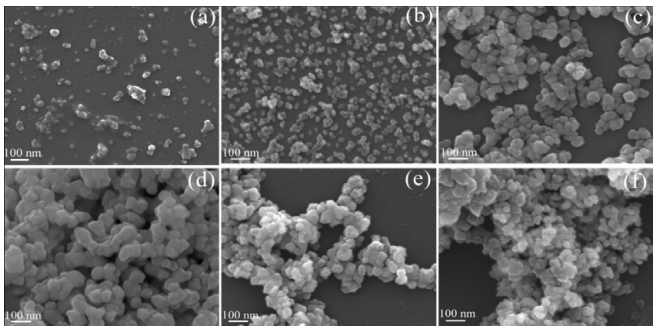

Figure 4: The SEM micrographs of 3Y-PSZ powders by microwave pyrolysis at different temperature: 700°C (a); 750°C (b); 800°C (c); 850°C (d); 900°C (e) and conventional sintered at 750°C (f).

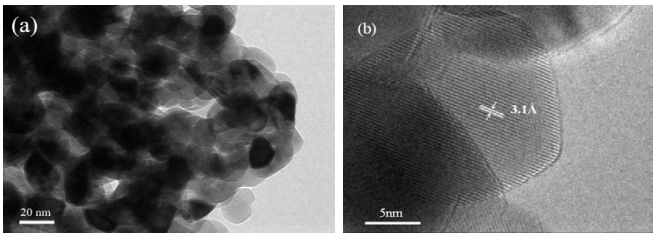

Figure 5: TEM image (a) and HRTEM image (b) of ZrO_2 powders.

The results of the XRD, Raman spectra, and SAED show the tetragonal ZrO_2 formation when the 3Y-TZP freeze-dried precursor powders calcined at 773-1273 K for 5 min. Moreover, the RAMAN spectrum shows that the tetragonal ZrO2 had already formed in the 3Y-TZP freeze-dried precursor powders. The crystallization activation energy of the tetragonal phase from the 3Y-TZP freeze-dried precursor powders when using a non-isothermal method was 169.2 ± 21.9 kJmol^{-1}. The crystallite growth morphology parameter (n) and crystallization mechanism index (m) were approximated as 2.0. This result means that the tetragonal ZrO_2 crystallites have a growth mechanism with a plate-like morphology.

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