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Synthesis of an alumina–YAG nanopowder via sol–gel method

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Abstract

Alumina–YAG ($Y_3Al_5O_{12}$) composite has attracted a great deal of attention as a high temperature structural ceramic due to its good creep resistance. In the present article, an Al_2O_3–YAG nanopowder was synthesized by aqueous sol–gel method using $AlCl_3{\cdot}6H_2O$, aluminum and Y_2O_3 powders as raw materials. The dried amorphous gel was heat treated in the range of 600–1500 °C. The influence of heat treatment on crystallization and phase transformation of the dried gel was investigated using X-ray diffractometery (XRD), scanning electron microscopy (SEM), thermogravimetry and differential thermal analysis (TG–DTA) and Fourier transform infrared spectroscopy (FTIR). It was shown that the gel heat treated at 1300 °C resulted in the formation of a crystalline Al_2O_3–YAG nanopowder with particle size distribution ranges from 70 to 130 nm.

Keywords: Nanostructured material; Sol–gel processes; Phase transitions; X-ray diffraction

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1. Introduction

Alumina is an important material in industrial applications and is widely used in high temperature conditions. Aluminum yttrium garnet ($Y_3Al_5O_{12}$, YAG) is a good candidate for hardening of Al_2O_3 because of its excellent creep resistance, similar coefficient of thermal expansion and no reaction with alumina. YAG has a large unit cell, which make dislocations generation difficult and suppress of them [1–3]. Many researchers investigated the creep behavior of alumina–YAG composite and found that it indicates a lower creep rate than pure alumina [4–7].

The quality of the sintered ceramic depends on the properties of the initial powder, such as sinterability, purity, homogeneity and particle size [8,9]. More recently, the synthesis of nanocrystalline ceramics have provided new opportunities. Nanocrystalline ceramics are structurally characterized by a large volume fraction of grain boundaries, which may significantly alter their physical, mechanical and chemical properties in comparison to those of conventional course-grained polycrystalline materials [10]. The former materials can be made from nano size raw powders [11]. Conventional processes for synthesizing ceramic nanopowder involve mechanical synthesis, vapor phase reaction, precipitation, combustion and sol–gel methods. There are some limitations in all aforementioned methods. Mechanical synthesis requires extensive mechanical ball milling and easily introduces impurities. Vapor phase reaction for preparation of nanopowder from a gas phase precursor demands high temperatures. The precipitation method takes too much time. The nanopowder obtained from the combustion method is usually hard aggregated [12–16]. Sol–gel technology has been developed for the fabrication of high quality nanopowders. For complex powders, it achieves ultra-homogeneous of the several components on a molecular scale [17,18]. But in this method the metal alkoxides usually used as raw materials are expensive.

In the present work, an alumina–YAG nanopowder was synthesized through an aqueous sol–gel method without using metal alkoxides. Decomposition of the gel, phase transformation and morphology of the synthesized alumina–YAG powder were investigated. No organic additives have been used to stabilize the sol because of their deleterious effects on product properties.

2. Experimental procedure

The precursor solutions for Al_2O_3–20 wt.% YAG nanopowder were prepared by sol–gel method using $AlCl_3{\cdot}6H_2O$ (Merck), Al powder, Y_2O_3 (Sigma–Aldrich) and HCl (Merck). Y_2O_3 powder was first dissolved in aqueous HCl. The main solution was prepared by dissolving aluminum chloride hexahy-

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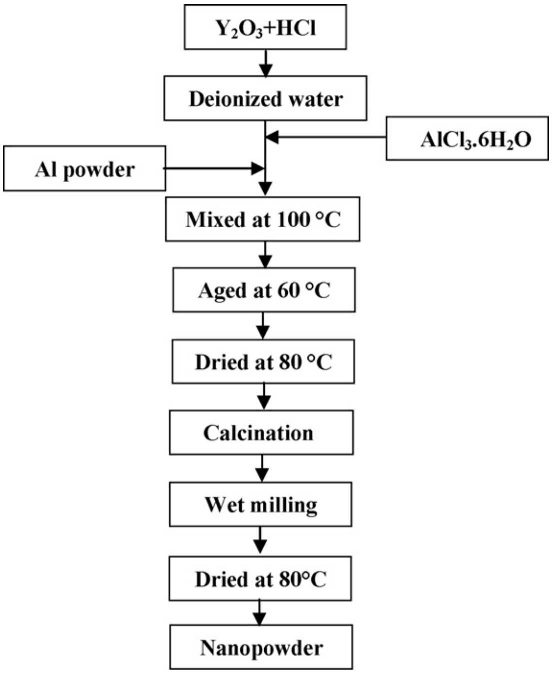

Fig. 1. The flow chart of the processing of Al_2O_3–YAG nanopowder.

drate, aluminum powder and yttrium oxide solution into the deionized water. The amount of yttrium oxide was 6 wt.% of premixed raw materials. The precursor solution was then continuously stirred at 100 °C for 4 h to completely dissolve the starting materials and was aged at 60 °C. The viscosity of the batch gradually increased and finally the batch set to a rigid gel. It was then dried at 80 °C for 48 h. The dried gel was calcined in a muffle furnace at various temperatures and wet ball milled in ethanol media using a high dense alumina jar and high pure alumina balls. The obtained powder was again dried at 80 °C. The flow chart of the powder preparation is shown in Fig. 1. In order to study the effects of yttrium on the phase transformations in the gel, one specimen was prepared without yttrium oxide powder (pure alumina gel).

X-ray diffraction was carried out for phase analysis of the dried gel and calcined powder using Philips X-pert model with Cu Kα radiation. Differential thermal analyses (DTA) and thermogravimetric (TG) were used in the range of 25–975 °C with a rate of 10 °C/min with the STA 1460 equipment. The powder morphology was investigated using a Phillips XL30 scanning electron microscope. Fourier transformation infrared spectroscopy analysis of dried gel and calcined powders were carried out in a Nicolet Nexus 6700 for studying the chemical groups of the dried gel and calcined powder.

3. Results and discussion

The synthesis mechanism may be described by the following reactions. Yttrium oxide first reacts with HCl and forms yttrium chloride (reaction (1)):

$$Y_2O_3 + 6HCl \Leftrightarrow 2YCl_3 + 3H_2O \quad (1)$$

Then aluminum chloride hexahydrate and yttrium chloride may be hydrolyzed to produce the sol (reactions (2) and (3)):

$$AlCl_3 + 3H_2O \Leftrightarrow Al(OH)_3 + 3HCl \quad (2)$$

$$YCl_3 + 3H_2O \Leftrightarrow Y(OH)_3 + 3HCl \quad (3)$$

Table 1
The effect of water on sol formation and gelation time

Molar ratio of H_2O/Al	Solution description	Gelation time (min)
40	Transparent	275
35	Transparent	215
30	Transparent	180
25	Opaque	–
20	Opaque	–

Reaction (4) shows that Al powder reacts with HCl to produce aluminum chloride and hydrogen gas and therefore Al can be used as a source of $AlCl_3$:

$$2Al + 6HCl \Leftrightarrow 2AlCl_3 + 3H_2 \quad (4)$$

Finally, the hydroxides groups produced in reactions (2) and (3) aggregate together to form the gel. The amount of water in the mixture must be precisely controlled. Table 1 presents the effect of water on sol formation and gelation time. This shows that the molar ratio of H_2O/Al should be fixed at about 30 to form a sol. The color of sol would be opaque when the ratio of H_2O/Al is lower than 30 due to insufficient amount of water for hydrolysis reactions. Adding more water than 30 results in an increase of the gelation time which may be attributed to the dilution effect. The pH value of precursor solution gradually reduced from 5 to 2 after heating for 4 h. This is related to the increase of the amount of HCl in the sol. Color of gel was yellow after drying because of the residual Cl.

The influence of the weight ratio of Al/$AlCl_3{\cdot}6H_2O$ on the gelation time and color of the resulting solutions are summarized in Table 2. As can be seen, the gelation time decreases by increasing aluminum powder. This is due to the increase of the rate of hydrolysis and condensation reactions. In addition, aluminum powder is cheaper than aluminum chloride hexahydrate, therefore, it is desirable to increase the Al/$AlCl_3{\cdot}6H_2O$ ratio. But increasing the amount of aluminum content in the sol decreases its transparency. Evidently this is due to insufficient solubility of aluminum in the sol. It seems that the 0.2 wt.% of Al/$AlCl_3{\cdot}6H_2O$ is an optimum ratio.

Fig. 2 presents thermogravimetry and differential thermal analysis (TG–DTA) curves of the dried gel. The DTA curve shows two overlapping endothermic peaks at 182 and 292 °C which are attributed to evaporation of absorbed water and dehydration of the dried gel, respectively. Corresponding weight loss of 40% appears in the TG curve.

An endothermic peak is also detected at 655 °C and the corresponding weight loss is about 4%. This peak may be caused by

Table 2
Dependence of sol formation and gelation time on weight ratio of Al/$AlCl_3$

Weight ratio of Al/$AlCl_3{\cdot}6H_2O$	Y_2O_3 content (wt.%)	Solution description	Gelation time (min)
0.10	6	Transparent	1500
0.12	6	Transparent	900
0.14	6	Transparent	660
0.16	6	Transparent	450
0.20	6	Transparent	180
0.25	6	Translucent	90 (not suitable)
0.50	6	Opaque	30 (not suitable)