Y_2O_3-doped Al_2O_3 transparent ceramics prepared by low temperature microwave sintering

Junming Luo*, Jiling Xu

(School of Materials Science and Engineering，Nanchang Hangkong University，Nanchang 330063，China)

Keywords: Ceramics; Optical materials and properties; Microwave sintering; Al_2O_3

Abstract：Y_2O_3- doped Al_2O_3 transparent ceramics have been successfully fabricated by microwave sintering at the lowest temperature. Compared to vacuum sintering processing, the microwave sintering needs a lower sintering temperature and shorter dwelling time to get highly dense Y_2O_3-doped Al_2O_3 transparent ceramics with finer grain size, higher mechanical and better optical properties. As a result, it is concluded that the low temperature microwave sintering has obvious advantages over the conventional sintering method in preparation of transparent Al_2O_3 ceramics.

Introduction

Due to its unique combination of low microplastic deformability, high hardness, corrosion resistance, thermal stability and optical properties, polycrystalline alumina ceramic is widely used in industry. One of its most important applications is as lamp envelopes in lighting equipment, windows and one of the emerging candidate materials for transparent amour applications.In the conventional sintering process, an extremely high sintering temperature (up to 1850-1900˚C) and long dwelling time (several hours) under high vacuum or pure hydrogen atmosphere are needed to fabricate transparent alumina with the highest density and minimum porosity, which, however, results in coarse grains (>15μm). Mechanical properties are important for the envelopes in energy-saving lamps with a high internal pressure of the discharged plasma.

Y_2O_3 is commonly used as grain-growth inhibitor in alumina ceramic for obtaining small grain size, which is an important prerequisite for producing transparent ceramic[1]. Microwave process offers several advantages, such as more rapid and uniform heating, shorter processing time, finer microstructure, enhanced densification and improved materials properties [2].

The transparent ceramics, such as alumina[3], Yttrium (Y_2O_3) [4] have been successfully fabricated by microwave sintering processing in the last fifteen years. Jiping Cheng used microwave sintering method to successfully prepare the first transparent alumina, but the sintering temperature was too high (up to 1750˚C), and the transmittance was low, additionally, the mechanical properties were not studied[5].In this paper, Y_2O_3-doped Al_2O_3 nanocrystalline powders were prepared by chemical co-precipitation method. The microwave sintering（1550˚C）method has been used to prepare the high transmittance Al_2O_3 transparent ceramics at the lowest temperature.

Experiments

High purity Al_2O_3 and Y_2O_3 powders with the mass fraction content of 0.5% were dissolved in nitric acid with the molar concentration of 0.1M under magnetic stirring. PEG2000 liquid was added to improve powder dispersion and ammonia with the molar concentration of 3M was added drop by drop at a rate of 2 ml/min to realize co-precipitation. PH value of the solution was maintained at 5. After aged for 24 h, the precipitates were filtered in a vacuum and washed with distilled water and alcohol, and then dried in an oven at 80˚C for 24 h and crushed in an agate mortar. Finally, the powder was calcined at 1300˚C for 2 h. The obtained powder was dry pressed manually (150 MPa) into small cylinder samples with a diameter of 20 mm and a thickness of 3 mm in a steel die. The samples were isostatically compacted at a pressure of 220 MPa, then heat-treated at 1100°C to remove organic materials. The microwave sintering was carried out in a 2.45 GHz, 5KW microwave furnace (NJZ4-3) with high purity hydrogen as reducing gas. The samples were heated up with the heating rate of 100°C/min and kept in the furnace at 1550°C for 30 min(the optimal process parameters), and then cooled down naturally. The vacuum sintering was carried out in a furnace with molybdenum heating element (VSF-7) at 1850°C for 5 h under vacuum of 1.3×10^{-3} Pa, the specimens were cut and double polished with 1 mm in thickness for spectral analysis.

The phases of the samples were characterized by X-ray diffraction (XRD). The surface topography of the samples was characterized by scanning electron microscopy (SEM), the relative densities of the samples were measured by Archimedes law, the mechanical properties were measured by WDW-50 Intron testing machine and HVS-1000 hardness tester. Moreover, the transmittances of the samples were measured using a DMR-22 fluorescence analyzer.

Results and discussion

Fig.1 is the FESEM image of Al_2O_3 powders calcined at 1300˚C for 2 h, indicating that the particle of the powder is well-distributed and spherical-like with diameter of 40-60 nm.Figure.2 show the pictures of the prepared Al_2O_3 transparent ceramics by microwave sintering and vacuum sintering, respectively. It can be seen that the prepared Al_2O_3 transparent ceramics by microwave sintering shows very good optical qualities, whose transmittance was 79 %, while the prepared Al_2O_3 transparent ceramics by vacuum sintering is translucent, whose transmittance was only 53%.Fig. 3 shows the XRD of the Al_2O_3 transparent ceramics samples processed by microwave sintering (a) and vacuum sintering (b). It can be seen from the figures that there is only α-Al_2O_3 phase in the samples，because amount of added Y_2O_3 is very little. It has been proved that the second phase can cause light scattering, so the single phase state is very useful for enhancing luminescence [6]. Compared to the microwave sintered sample, the positions of the diffraction peaks do not change for vacuum sintered sample, but the diffraction peaks become sharper, indicating that the grains are larger.

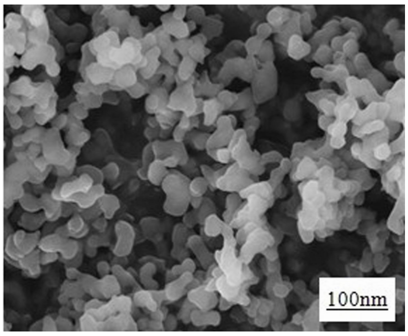


Fig.1. FESEM of the Al_2O_3 powders calcined at 1300˚C for 2h

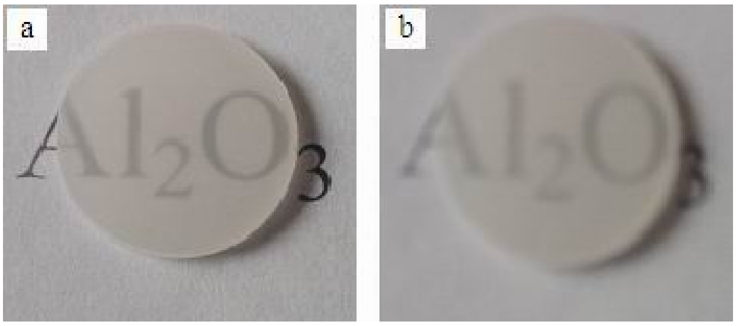


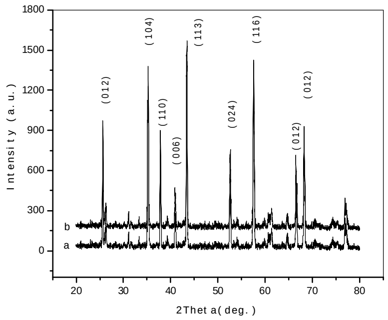
Fig.2. Picture of Al_2O_3 transparent samples prepared by microwave sintering(a), vacuum sintering(b)

Table 1 shows the comparison of the processes and properties of the Al_2O_3 transparent ceramics prepared by both microwave and vacuum sintering. Compared to vacuum sintering, the microwave sintering has many advantages including more than 10 times higher heating rate, about 200-300°C lower sintering temperature, and about 270 min shorter dwelling time (actually, only some 30 min for the specified samples in our present experiments). Furthermore, the microhardness, bending strength and fracture toughness are increased from17.36 GPa to 19.62, 480 MPa to 658 MPa, and3.96 MPa $m^{1/2}$ to 4.81 MPa $m^{1/2}$ respectively, being improved by about 13 %, 37.1 % and 21.5 % in relative amplitudes, and the relative density is increased from 98.1% to 99.5%, the transmittances is increased from 53 % to 79 %.

Table1 The comparison of processes and properties by microwave and vacuum sintering

Processes and properties	Microwave sintering	Vacuum sintering
Heating rate/˚C·min^{-1}	100	10
Temperature/˚C	1550	1850
Holding time/min	30	300
Relative density/%	99.5	98.1
Microhardness/GPa	19.62	17.36
Bending strength/MPa	658	480
Fracture toughness/ MPa.$m^{1/2}$	4.81	3.96
Transmittance/%	79	53

Figs. 4 show the SEM images of the surfaces of the Al_2O_3 transparent ceramic obtained by microwave and vacuum sintering. It clearly demonstrated that the grain distribution of the vacuum sintered sample is not uniform with the smallest and the largest particle size of about 3μm and 15 μm respectively, while the grain distribution of microwave sintered sample is uniform with the average diameter at about 2-4 μm, with no abnormal growth or coarsening phenomenon. We have further found out that the smaller the grain size, the higher the mechanical and optical properties.



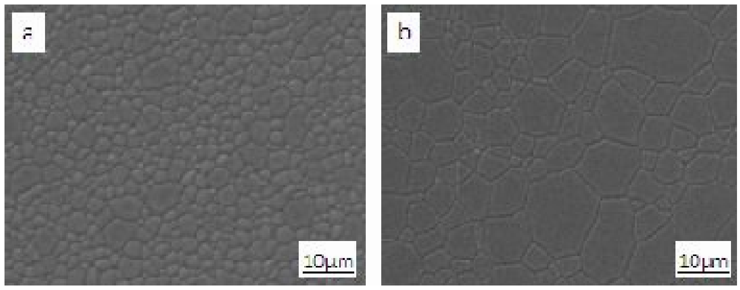


Fig.3. XRD patterns of transparent ceramics surface by microwave
by microwave sintering (a),vacuum sintering (b)

Fig.4. SEM image of of transparent ceramics
sintering (a),vacuum sintering (b)

There are four main reasons that microwave sintered sample under hydrogen atmosphere have high optical and mechanical properties as follow. Firstly, since Y_2O_3 -doped Al_2O_3 powder with particle size of about 40-60nm has high interface energy which could become the driving energy of the atomic movement and promote the shrinkage of interface hole, the sample can get full densification at a lower temperature. Secondly, because the radius of Y^{3+} (0.106 nm) is greater than the Al^{3+} (0.0535 nm), the solid solubility of Y_2O_3 in Al_2O_3 is only 3×10^{-3}. Y_2O_3 mainly exists in the grain boundary of Al_2O_3, which reduces the migration rate of Al_2O_3 grain boundary, inhibits the grain growth, and promotes the sintering [7]. Thirdly, for sintering under hydrogen atmosphere, the pores are basically eliminated since the gas in the pores is displaced and quickly spreads[8]. Finally, the kinetic energy of molecules or ions inside the material increases due to microwave electromagnetic energy during microwave sintering, thus the sintering activation energy increases and the diffusion coefficient increases[9]. As a result, the low temperature and rapid sintering is realized. Compared to vacuum sintering, the sintering temperature for the microwave sintering reduces from 1850^{o}C to 1550^{o}C and the dwelling time shortens from 300 to 30 min. The microwave sintering is an effective method to get fine or superfine crystal and accelerate the ceramic densification. It has been found that the relative density, mechanical and optical properties of the samples prepared by microwave sintering under hydrogen atmosphere have been improved obviously compared to those of vacuum sintered samples.

Conclusions

Y_2O_3-doped Al_2O_3 nanocrystalline powders have been successfully prepared by chemical co-precipitation method. Y_2O_3-doped Al_2O_3 transparent ceramic samples have been successfully prepared by microwave sintering method at the lowest sintering temperature. Compared to the vacuum sintered samples, the microwave sintered samples have finer grains, higher microhardness, stronger bending strength, larger fracture toughness, and better transmittances. It has been found that the microwave sintering technique has high potential for the fabrication of optical ceramics for industrial and military applications.

Acknowledgments

This work has been financially supported by the Natural Science Foundation of China

(51362022), the Department Education Research Project of Jiangxi Province(GJJ13500).

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