Effects of heating rate on the microstructure and mechanical properties of rapid vacuum sintered translucent alumina

HaiHong Zhang, YaLi Xu, Bo Wang, Xiao Zhang, JianFeng Yang, Koichi Niihara

State Key Laboratory for Mechanical Behavior of Materials, Xi’an Jiaotong University, Xi’an 710049, China
Luoyang Ship Material Research Institute, Luoyang 471023, China
Extreme Energy-Density Research Institute, Nagaoka University of Technology, Nagaoka 940-2188, Japan

Abstract

Translucent polycrystalline alumina ceramics were fabricated by rapid vacuum sintering using commercial α-Al₂O₃ as a starting powder and MgO as an additive. Effect of heating rate on the sintering behavior, microstructure and properties of translucent alumina was investigated. It was shown that with increasing the heating rate from 50 to 150 °C/min, grain growth was inhibited due to the restrained surface diffusion, and densification was promoted due to the enhanced vacancy diffusion. The grain size decreased from 14.8 μm to 4 μm, and the flexural strength increased from 352 MPa to 570 MPa correspondingly. Further increase of the heating rate led to the increase of residual pores, which was owing to the insufficient rearrangement of the Al₂O₃ particles. The in-line transmission was related to the porosity and grain size.

Keywords: Translucent alumina; Flexural strength; In-line transmission; Heating rate

1. Introduction

Because of its great strength, thermal resistance and chemical durability, translucent polycrystalline alumina (PCA) has been widely used in the industrial and military areas since 1960s when it was firstly synthesized by Coble [1]. During recent decades, outstanding properties of PCA like good transmittance, combining with high strength and hardness can be achieved when decreasing its grain size to sub-micrometer [2] or nanoscale [3].

New sintering technologies, such as hot pressing sintering (HP), Spark Plasma Sintering (SPS) [4,5], Hot Isostatic Pressing sintering (HIP) [6] and microwave sintering etc. [7] have been used for the sintering of translucent PCA to achieve the great microstructure of fine grain, of which the sintered bodies are usually characterized by high density, controlled grain size and good transparency. These sintering technologies can be divided into two kinds: pressure utilized sintering and fast sintering. The pressure during sintering process accelerates the sliding of grain boundary which could benefit the densification. While fast sintering aims to get full densification and limited grain growth rate by carefully controlling the sintering schedule. The surface diffusion for the coarsening mechanisms commonly dominates over the lattice and grain-boundary diffusion for densification mechanisms at lower temperatures, thus the shorter time spent at this stage serves to reduce the excess coarsening while the driving force for densification is not affected significantly [8]. Many researchers have proved that high density combined with fine grain size can be achieved through rapid heating to higher temperatures [9,10].

The above mentioned sintering techniques require pretty expensive apparatus and the mold limited the shape of their products which restrict their industrial applications. Rapid vacuum sintering, which proposed by Zhang [11], was a combination of fast sintering and pressureless sintering. It has no external pressure and its fast heating rate was achieved through an induction coil. Rapid vacuum sintering has rapid heating and cooling rates at the sintering temperature below
1700 °C, meanwhile it can achieve considerably high density within very short sintering time (in ten minutes). Thus compared with the above mentioned sintering methods rapid vacuum sintering manifests distinct advantages in preparing ultrafine materials [12–14]. As a result of its lower sintering temperature and shorter sintering time, the grain size and size distribution of its products can be controlled.

The effect of heating rate on sintering is very controversial, which attracts considerable attention. Zhou et al. [9] reduced grain growth rate during the PECS rapid heating process, and concluded that the effect of heating rate on densification was influenced by particle size and maximum sintering temperature. According to the research of Kim et al. [15], after sintering at 1150 °C by SPS and increasing the heating rate from 10 °C/min to 100 °C/min, the sample turned from transparent to opaque and the grain size of aluminum changed from 0.29 μm to 0.55 μm and the porosity changed from 0.02% to 0.59%. Stuer [16] sintered MgO doped alumina through SPS. He considered that high heating rate had a negative effect on real in-line transmittance. However, Aminzare [17] reported an approximately full dense alumina of which the average grain size reduced from 875 nm to 443 nm when the heating rate increased from 2 °C/min to 25 °C/min, and its grain size distribution became narrower. And higher strength and finer grain size were obtained by using faster heating in ZrB2–SiC–ZrC system according to Snyder’s research [18]. Schwarz [19] found that higher heating rates enhanced the densification kinetics, both during the heating and soaking processes when preparing zinc oxide by SPS.

In this work, translucent polycrystalline alumina was prepared by rapid vacuum sintering with different heating rates (from 50 °C/min to 200 °C/min) using 0.25 wt% MgO as an additive. The effect of heating rate on the sintering behavior, microstructure, mechanical properties and transmittance of translucent alumina was investigated.

2. Experiment procedure

Commercial α-alumina powder (mean particle size of 350 nm; 99.995%; Tianli Industry & Trade Co. Ltd., Shandong, China) and MgO powder (99.99%; 30 nm; Nachen S&T Co. Ltd., Beijing, China) were used as starting powder. The alumina was mixed with 0.25 wt% MgO, and the mixed powder was ball-milled using a planetary ball milling machine with anhydrous alcohol as grinding aid in a polyethylene bottle, the rotating rate is 50 r/min and lasting for 8 h.

The ball-milled powders were dried and then passed through a 200-mesh sieve. Afterwards they were put into steel mold and uniaxial pressed at a pressure of 250 MPa. Two types of samples were prepared: Φ15 mm × 1.5 mm cylindrical samples and 50 mm × 5 mm × 5 mm square bar ones. The compacts were pre-heated at 1200 °C for 2 h with a heating rate of 5 °C/min in air furnace (LHT 02/17, Nabertherm Industrial Furnace Ltd. Co., Shanghai, China). The pre-sintered samples were heated at 1650 °C for 5 min with different heating rates (50, 100, 150 and 200 °C/min) in a low vacuum (about 5 × 103 Pa), and then cooled down with the furnace.

The relative density of the sintered samples was measured by the Archimedes method using 3.98 g/cm3 as the theoretical density of alumina. The bar samples (50 mm × 5 mm × 5 mm) were ground and polished into test bars for flexural strength measurement, and tested by a three-point bending method with a span of 16 mm and a cross-head moving speed of 0.5 mm/min at room temperature. Each final value was averaged over five measurements. After flexural strength measurement, the fractured surface of samples was observed using scanning electron microscopy (VEGAAII XMU, Tescan, Czech) to reveal the microstructure. Average grain size was calculated by a line-intercept method. Each value was averaged over 200–300 grains. The as-calculated grain sizes were then multiplied by a ratio of 1.225 [20] to represent the real grain size. The circle samples were ground to a thickness of 1 mm and carefully mirror-polished on both sides for the measurement of optical property. The in-line transmission [20–22] was measured in the wave length range of 350–1100 nm using a spectrophotometer (UV-1800; Mapada, Shanghai, China) by inserting a slide (0.5 mm diameter) in front of the detector in order to allow the detection of only the specularly transmitter portion of the incident light beam with a 0.5 mm diameter. The distance between the sample and the detector is about 55 mm. The scattering range of the measured optical properties is less than 5%.

3. Results and discussion

Fig. 1 shows the effect of heating rate on the porosity and grain size of sintered samples. As can be seen from Fig. 1, with the heating rate increased from 50 °C/min to 200 °C/min, the grain size considerably decreased from 14.8 μm to 4 μm. The porosity reduced slightly with the heating rate increased from 50 °C/min to 150 °C/min, and then it increased when the heating rate reached 200 °C/min. It is well known that densification and grain growth are two competitive processes during sintering. The porosities of less than 0.5% indicated that all the samples reached the final sintering stage. Fast heating
was beneficial to the restraining of surface diffusion for the coarsening mechanisms, resulted in the decreased grain size and more grain boundaries. Consequently vacancy diffusion through the grain boundary was enhanced for the elimination of closed pores at the final sintering stage.

Since fast sintering contributed to the fine grain size and promoted the elimination of pores at lower temperature, consequently near full density can be reached even with a high heating rate of 150°C/min and a short soaking time of 5 min. However, when the heating rate was too rapid, such as 200°C/min, the lack of the particle rearrangement at the first stage and large temperature hysteresis between the heater and sample might be the reason for the high porosity after sintering.

Fig. 2 shows the SEM photographs of the fracture surface of samples sintered at 1650°C via different heating rates. As can be seen, with the increase of the heating rate, the gain size decreased remarkably. For the samples with heating rate less than 150°C/min (Fig. 2(a)–(c)), the sintered product was quite compact and nearly no pores could be detected. However, for the sample with high heating rate of 200°C/min (Fig. 2(d)), some minor pores existed at grain boundaries and inside the grains, which are marked with circle in Fig. 2(d). This phenomenon was consistent with the result of porosity and grain size measurement which is shown in Fig. 1.

Fig. 3 shows the influence of heating rate on the flexural strength of sintered samples. With the heating rate increased from 50°C/min to 200°C/min, the strength of samples increased from 352.2 MPa to 570.1 MPa. As can be seen in Fig. 1 and Fig. 2, the porosities were very low and the pore size was very small. Consequently, the remained pores were no more acted as the fracture flaw, while the grain size was the
dominant factor for the influence of the strength. Usually, the strength varies with grain size. According to the Hall–Petch equation [23, 24], the yield strength $\sigma_{LYP}$ is linear with the inverse square root of the grain diameter $d$: $\sigma_{LYP} = \sigma' + Kd^{-1/2}$, so fine grained material has high strength. Since high heating rate led to the fine grain microstructure, the flexural strength was clearly increased following the Hall–Petch equation.

Fig. 4 shows the in-line transmission of the samples obtained with different heating rates. The transmittance increased with the heating rate. However, after reaching peak value at 100 °C/min, it decreased considerably. For the crystal material, the transparency was related to the porosity and grain boundary proportion. The low transparencies of samples obtained at 200 °C/min was mainly caused by the high porosity, although it had the fine grain size.

Below the heating rate of 200 °C/min, very few pores were caused by the fast heating through the enhanced vacancy diffusion along the boundary of the refined grain size. As a result, the influences of porosity and grain boundary proportion on the transmittance were contradictory for these samples. The low transmittance for the samples obtained at 50 °C/min was due to its high porosity, the same as that at 200 °C/min, although they had fewer grain boundaries. The materials obtained at 100 °C/min and 150 °C/min have a similar low porosity between 0.1% and 0.15%, however, the grain size of the materials obtained at 100 °C/min was much larger than that at 150 °C/min (10.26 μm compare to 7.4 μm), indicating low proportion of the grain boundary, which contributes to the highest in-line transmission value.

4. Conclusion

Effect of heating rate on the sintering behavior and properties of translucent alumina ceramics obtained by rapid vacuum sintering of nano-α-Al2O3 and 0.25 wt% MgO was investigated. As the heating rate increased, the grain size decreased due to the restrained surface diffusion. The porosity decreased with the heating rate increase from 50 °C/min to 150 °C/min due to the enhanced vacancy diffusion along the boundary of the refined grain size, and then it increased as the heating rate reached 200 °C/min because of the lack of particle rearrangement at the first sintering stage. Correspondingly, the sample with the heating rate of 200 °C/min had the highest flexural strength of 570.1 MPa because of the smallest grain size, and the sample with 100 °C/min had highest transmittance due to the lower porosity and fewer grain boundaries.

Acknowledgments

This work was supported by the National Natural Science Foundation of China (Grant nos. 51272205 and 51072157), by China Postdoctoral Science Foundation (Grant no. 2012M521763), and by the Fundamental Research Funds for the Central Universities (xjj2013111).

References


学霸图书馆
www.xuebalib.com

本文献由“学霸图书馆-文献云下载”收集自网络，仅供学习交流使用。

学霸图书馆（www.xuebalib.com）是一个“整合众多图书馆数据库资源，
提供一站式文献检索和下载服务”的24小时在线不限IP图书馆。
图书馆致力于便利、促进学习与科研，提供最强文献下载服务。

图书馆导航：
图书馆首页     文献云下载     图书馆入口     外文数据库大全     疑难文献辅助工具