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Development of Translucent Aluminum Nitride (AIN) Using Microwave Sintering Process

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Abstract. Pure aluminum nitride (AIN) has been successfully sintered to highly translucent form by microwave sintering at 1850°C with a dwelling time of 30–60 minutes. The results showed that the sintering temperature should be at least 1850°C or higher to get reasonable translucency in the AIN sample by the microwave sintering process. On the other hand, the conventional sintering method requires much longer sintering time to obtain a translucent AIN ceramics.

Keywords: aluminum nitride, microwave sintering, translucent ceramics

Introduction

Aluminum nitride (AIN) has attracted much attention as a substrate and package material for high-power integrated circuits due to its unique physical properties, such as high thermal conductivity, high electrical resistivity, high dielectric strength and low dielectric constant, and low thermal expansion coefficient which matches with that of silicon [1–3]. Translucent AIN can also be used for some special applications in electrooptics area, for example, as substrates for laser and light-emitting diodes, and infrared seeker window for high-speed missiles [4].

Since AIN is a covalent compound, limited atomic mobility makes it very difficult for complete densification of pure AIN at reasonable sintering temperatures. At higher temperatures ($\geq$1600°C), possibility of decomposition of AIN is another problem to achieve dense sintered bodies. Thus, sintering aids, including rare-earth or alkaline earth oxides or fluorides, such as yttria, calcia, are commonly used to assist sintering of AIN to high density. In 1984, N. Kuramoto and H. Taniguchi reported their efforts to make translucent AIN, and they improved their results further in 1989. The authors concluded that the requirement for the starting AIN powder material was that it should be in high purity, good formability and excellent sinterability, to make it translucent. Using this kind of AIN powder, highly translucent AIN ceramic samples were prepared by conventional sintering at 1850–1900°C with a dwell time longing from 3 to 7 hours [5, 6].

Microwave sintering is a new processing technique for sintering of ceramic materials. It differs fundamentally from the conventional sintering processes. The ceramic green bodies that placed in microwave field absorb microwave energy and convert it to heat directly providing volumetric heating. It has been demonstrated that using microwave process offers several technical and economic advantages, such as faster heating rate, lower sintering temperature, shorter sintering time, finer average grain size, better products properties, and energy/cost savings [7, 8]. In this work, highly translucent AIN samples were prepared by microwave sintering at 1850°C in a duration time of 30–60 minutes under ambient pressure and nitrogen atmosphere. Compared to the conventional sintering method, microwave sintering of AIN into highly translucent ceramics was achieved in much shorter sintering time.

Experimental

AIN powder Grade F from Tokuyama Soda, Japan, with a mean particle size of 1.5 μm was used in this study. The staring powder was blended with 2.0 wt% of polyvinyl alcohol and uniaxially pressed at 30 MPa followed by the cold isostatically pressing (CIP) under a pressure of 280 MPa into pellets with three different sizes: 12.7 mm diameter and 1.5 mm thick samples for a systematically experimental study, 19 mm diameter and 5 mm thick samples for thermal conductivity measurements, and 31.7 mm and 2 mm thick for microwave scale-up study. The compacted pellets were pre-heated using a resistance furnace at 1100°C for 2 hours in flowing nitrogen atmosphere to burn out the binder.

The small (12.5 mm diameter) samples were microwave sintered using a TE_{103} single mode cavity with a 2.45 GHz, 1.5 kW microwave generator power supply, and the sintering of large (19 mm and 31.7 mm diameter) samples was conducted in a 2.45 GHz, 6 kW multimode microwave system. The heating rate was controlled within 150°C/min in the single mode, and 50°C/min for the multimode system. 1-atm flowing ultrahigh purity nitrogen was used as sintering atmosphere for all samples. An optical pyrometer was used to measure the surface temperature of the samples during microwave sintering.

The insulation package arrangement for the microwave sintering AIN is schematically shown in Fig. 1. The compacted AIN green pellets were enveloped with a BN crucible, and a mixture of 50 wt% AIN and 50 wt% BN powder was used as powder bed to separate the pellets and to reduce the evaporation or decomposition of AIN at high temperature. A small hole was made on the BN crucible's top cover for observing the temperature inside. No microwave suscepor materials used in our experiments. It was found that the AIN can be heated very efficiently in the single mode microwave cavity without any auxiliary microwave absorptive materials. The heating in a multimode microwave system was not very efficient, since AIN has low dielectric loss. In this case, a piece of metal reflector was introduced in the cavity [9]. By adjusting the angle and position of the reflector, the microwave power could be focused on the area where the samples located to greatly increase the microwave heating efficiency. In addition, the samples with the insulation package were subjected to rotate during the microwave processing in the multimode cavity, to allow uniform heating and sintering.

The densities of sintered samples were determined using the Archimedes' method. The microstructures were studied with scanning electron microscope (SEM). The thermal conductivity was determined using laser flash method, and the transmittance was measured using Magna-IR 560 Spectrometer (Nicolet Instrument Co., Madison, WI, USA).

Results and Discussion

The densification behaviors of the AIN samples during the microwave sintering are shown in Fig. 2. The starting dimensions of the green samples were 12.5 mm in diameter and 1.5 mm in thickness. All samples were sintered in a single mode microwave cavity at different temperatures with the same dwelling time of 30 minutes. It was observed that there was little

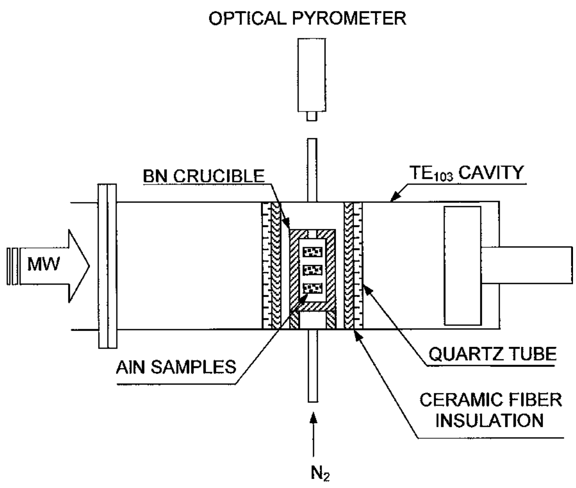

Fig. 1. The scheme of the insulation package arrangements for microwave sintering of AIN.

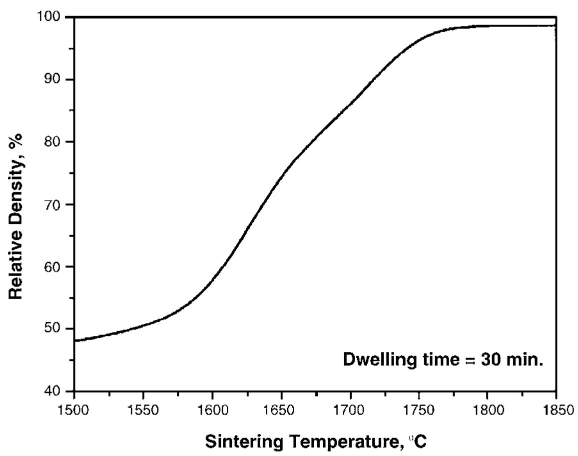

Fig. 2. Densification of AIN samples under different microwave sintering temperatures.

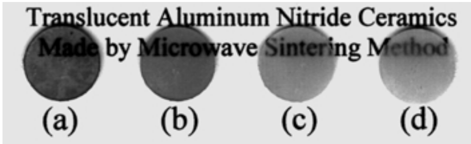

Fig. 3. Appearance of the AlN samples under different microwave sintering conditions. (a) Microwave sintered at 1850°C for 60 minutes, (b) microwave sintered at 1850°C for 30 minutes, (c) microwave sintered at 1750°C for 60 minutes, and (d) microwave sintered at 1750°C for 30 minutes.

consolidation occurred when the sintering temperature was below 1600°C. Due to the covalent nature of the Al—N bond, densification of pure AlN is difficult. However, compared to the coarse AlN powder, fine AlN powders possess favorable densification potential. Hashimoto et al. demonstrated that fully densified samples could be obtained by using starting powder with a specific surface area of 16.6 m^2/g under pressureless sintering at 1700°C, and that adding Y_2O_3 further reduced the sintering temperature to 1600°C [10]. The starting AlN powder used in this work possessed an average particle size of 1.5 μm. The samples microwave sintered at 1750°C for 30 minutes had achieved almost complete densification with densities higher than 98% theoretical density and a very little translucency. A further increase of the sintering temperature did not result a significant increase in the sinter density. However, a longer sintering time or higher sintering temperature can improve the transparency as shown in Fig. 3. The sample sintered at 1750°C for 60 minutes showed higher transparency, compared to the sample sintered at the same temperature for only 30 minutes. A higher sintering temperature (1850°C) resulted into significant improvement in transparency. Table 1 listed the comparison results of the AlN samples under different microwave sintering conditions.

Table 1. Comparison of the AlN samples under different microwave sintering conditions.

Sample	Microwave sintering conditions (min)	Density (g/cm^3)	Transmittance (%) (wavelength = 5.8 μm, sample thickness = 0.5 mm)
A	1850°C × 60	3.22	42
B	1850°C × 30	3.22	38
C	1750°C × 60	3.21	23
D	1750°C × 30	3.20	18

Though the AlN samples sintered at 1750°C were fully densified, but their optical properties varied with sintering temperature and time. The optical transmittance of polycrystalline ceramics is depended not only on the crystal structure, but also microstructure (grain size, porosity, etc.) and any impurity phases. The most efficient approach to improve the optical transmittance of a polycrystalline ceramic is to reduce porosity and eliminate impurities from the boundary area by optimizing the sintering conditions. Figure 4 reveals the microstructures of the sintered AlN samples. The average grain size of the AlN sample sintered at 1750°C was around 4–5 μm (for 30 minutes), and 6–8 μm (for 60 minutes). The samples sintered at 1850°C for 30 minutes had the same grain size as the sample sintered at 1750°C for 60 minutes, the main difference is the pore structures. There were some large pores existed along the grain boundaries in the samples sintered at 1750°C. At a higher sintering temperature, these pores became much smaller or completely disappeared; the sample sintered at 1850°C for 60 minutes exhibited much bigger grain size, and there were only very few and very small pores existed along the grain boundaries. The grain boundary volume also had considerably reduced in the sample sintered at 1850°C for 60 minutes, and therefore it gives the highest degree of transparency among all samples. Those results showed that to make highly translucent AlN ceramics, the sintering temperature should be at least 1850°C.

Table 2 gives the comparison of our microwave sintering and Kuramoto's conventional sintering results. The main difference between microwave and conventional sintering methods is the sintering time to achieve highly translucent AlN ceramic samples. Compared to the conventional sintering of translucent AlN ceramic samples, microwave sintering technique reduces sintering time dramatically from several hours to a few minutes. Many researchers have demonstrated that microwave radiation provides sibnificant enhancement on the reaction kinetics, material diffusion and sintering rates. In our earlier work, it was proved that the diffusion coefficient was much greater (about 3 times higher) in microwave sinerting than that in conventional sintering of Al_2O_3 ceramics [11]. Janney and Kimrey reported that the sintering activated energy of Al_2O_3 ceramics was greatly reduced by microwave processing [12]. Binner et al. showd that diffusion rate constants using microwave were more than treble those achieved using conventional heating for synthesis of non-oxide ceramic powders [13]. For

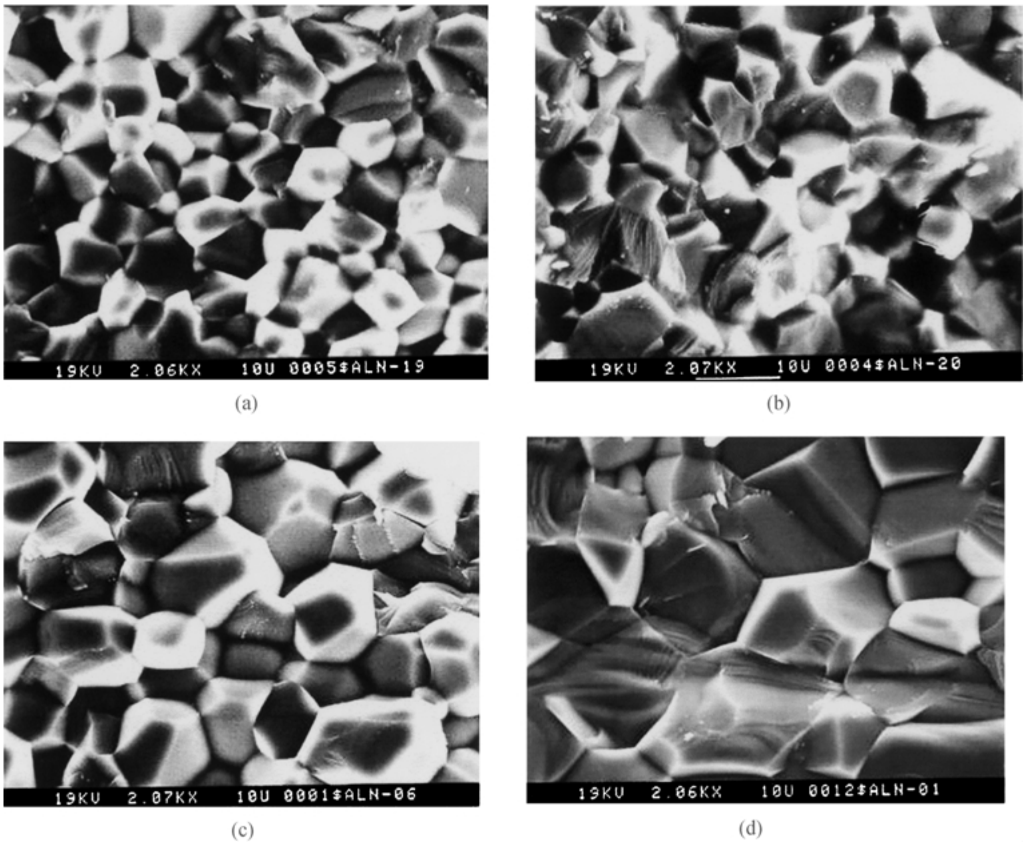

Fig. 4. The comparison of the microstructure (fracture surface) of the AIN samples under different microwave sintering conditions. (a) Microwave sintered at 1750°C for 30 minutes, (b) microwave sintered at 1750°C for 60 minutes, (c) microwave sintered at 1850°C for 30 minutes, and (d) microwave sintered at 1850°C for 60 minutes.

such kind of "microwave effect", the microwave sintering AIN to highly translucent can be achieved in much shorter time as compared to the conventional processing.

The thermal properties measurement results showed that the thermal conductivities of our translucent AIN samples (microwave sintered at 1850°C for 30 minutes) were between 96–102 W/m·K. Since the samples were

Table 2. Comparison of microwave and conventional sintering of translucent AIN.

	Microwave sintering	Conventional sintering[a]
Starting AIN powder	Tokuyama Soda, Japan, Grade F	Tokuyama Soda, Japan, Grade F
	Mean partical size: 1.5 μm	Mean partical size: 1.4 μm
	Oxygen content: 1.0%	Oxygen content: 1.0%
Sintering temperature	1850°C	1850°C
Sintering atmosphere	N_2 (1 atm)	N_2 (1 atm)
Sintering time	0.5 hour	7 hours
Sintered sample's density	3.22 g/cm^3	3.22 g/cm^3
Transmittance (at 5.8 μm)	38%	Translucent (no data)
Thermal conductivity	98–104 W/m·K	124 W/m·K

[a]From Kuramoto, et al.'s results [6].

made of pure AIN powder without any additives, it was expected that these samples would not achieve very high thermal conductivity. There are two ways to increase the thermal conductivity, one is to use lower oxygen content AIN powder (the oxygen content of the AIN powder we used was ∼0.9%), the other is to use sintering aids. In Kuramoto's work, they used two different oxygen contents (0.2 and 0.06%) AIN powder and added 2.1 wt% of $3CaO \cdot Al_2O_3$ (C_3A), and that is how they were able to achieve higher thermal conductivities (170 and 260 W/m·K). We believe that highly translucent with high thermal conductivity AIN samples can be prepared by microwave sintering if reduction of the oxygen content and addition of suitable sintering aids are employed.

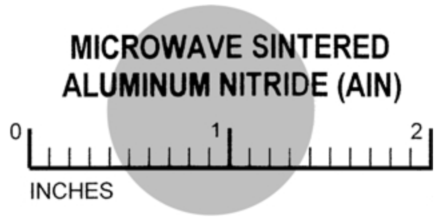

Fig. 5. The appearance of the translucent AIN sample (1-inch diameter) made by microwave sintering at 1850°C for 30 minutes.

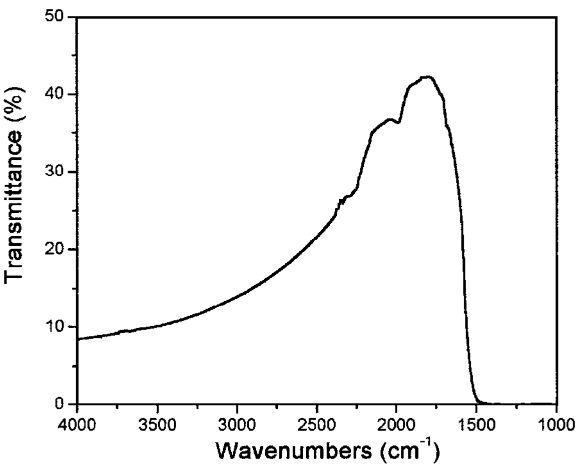

Fig. 6. The transmittance of the translucent AIN sample (25.4 mm diameter, 0.8 mm thickness).

Finally, for the scaling up, some AIN samples with a starting diameter of 31.7 mm were also sintered using the multimode microwave cavity at 1850°C for 30 minutes. The sintered sample had a diameter of around 25.4 mm (1 inch) in highly translucent form as shown in Fig. 5, and its transmittance measurement shown in Fig. 6.

Conclusion

Pure AIN samples with no additives were microwave sintered to highly translucent form. The results showed that one can not produce AIN samples with good translucency at sintering temperatures lower than 1750°C even with longer dwelling time. However, compared to conventional sintering method, microwave sintering of translucent AIN ceramic can be conducted in much shorter sintering time.

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