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2004

Khor, K. A., Yu, L. G., & Murakoshi, Y. (2004). Spark plasma sintering of Sm2O3-doped aluminum nitride. Journal of the European Ceramic Society, 25(7), 1057–1065.

https://hdl.handle.net/10356/95602

https://doi.org/10.1016/j.jeurceramsoc.2003.12.020

Spark Plasma Sintering of Sm_2O_3-Doped Aluminum Nitride

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ABSTRACT

The high sintering temperature required for aluminum nitride (AlN) at typically 1800°C, is an impediment to its development as an engineering material. Spark plasma sintering (SPS) of AlN is carried out with samarium oxide (Sm_2O_3) as sintering additive at a sintering temperature as low as 1500-1600ºC. The effect of sintering temperature and SPS cycle on the microstructure and performance of AlN is studied. There appears to be a direct correlation between SPS temperature and SPS cycle with the density of the sintered sample. The addition of Sm_2O_3 as a sintering aid (1 and 3 wt. %) improves the properties and density of AlN noticeably. Thermal conductivity of AlN samples improves with increase in number of SPS cycle (maximum of 2) and sintering temperature (up to 1600 °C). Thermal conductivity is found to be greatly improved with the presence of Sm_2O_3 as sintering additive, with a thermal conductiviy value about 118 W/m·K for the 3 wt. % Sm_2O_3-doped AlN sample SPS at 1500ºC for 3 minutes. Dielectric constant of the sintered AlN samples is dependent on the relative density of the samples. SPS cycle and sintering aid do not, however, cause significant elevation of the dielectric constant of the sintered samples.

Microstructures of the AlN samples show that, densification of AlN sample is effectively enhanced through increase in the operating SPS temperature and the employment of multiple SPS cycles. Addition of Sm_2O_3 greatly improves the densification of AlN sample while maintaining a fine grain structure. The addition of Sm_2O_3 modifies the microstructures to more faceted AlN grains, resulting in the flattening of AlN-AlN grain contacts.

Keywords: *AlN, Samaria (Sm_2O_3), Relative Density, Thermal Conductivity, spark plasma sintering*

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1. INTRODUCTION

The recent advancement in the density and performance of integrated circuits at higher and faster clock speeds have infused an acute prerequisite for enhanced thermal management of electronic chips [1]. The commercial importance of aluminum nitride (AlN) stems from its high thermal conductivity and moderately low dielectric constant [2]. Moreover, the thermal coefficient of AlN matches well with that of silicon, and it displays desirable properties like high thermal resistance and thermal shock behavior. With these exceptional properties, AlN is considered to be a potential candidate for high performance substrate material.

However, due to the covalent bonding, and small self-diffusion coefficients of constituent elements, full densification of pure AlN is difficult to attain, even at high temperatures above 1800ºC [3]. Furthermore, sintering at high temperatures is undesirable from the vantage point of a mass production system [4], and may promote significant grain growth, causing a reduction of mechanical strength [5]. In addition, as oxygen can easily dissolve in the AlN lattice, or at the grain boundary to form defects that scatter phonons, the thermal conductivity of sintered AlN ceramics is inadvertently reduced, significantly [6]. In order to lower the sintering temperature and remove the oxygen atoms from the grain boundaries and lattices, so as to improve the thermal conductivity, sintering additives such as Y_2O_3 [7], CaO [8,9], Sm_2O_3 [7,10,11] and CaF_2 [12,13] have been studied. Among these additives, rare earth oxides (Y_2O_3, Sm_2O_3 et al.) are proved to be the most effective sintering additives [7]. Yet, due to the high liquidus temperature of AlN-Al_2O_3-Re_xO_y (Re: rare earth), purification of AlN lattice still initiates at a temperature higher than 1800ºC [5].

In order to enhance AlN densification at relatively low processing temperatures, one can introduce some less effective low melting point additives like CaO [5], or use fine grain size raw powders. But the side effect of these methods is obvious [5, 14]. Another way is to introduce advanced sintering methods such as spark plasma sintering.

Spark Plasma Sintering (SPS) is a recently developed densification method by which powders can be sintered very rapidly at relatively low temperatures [15-20]. In the SPS process, a pulsed direct current (dc) is applied to the sintering powder, and the activation of powder particles is thought to be achieved through the application of electrical discharges. The sintering mechanism of SPS can be found elsewhere in the literatures [21-23]. The application of an external electric field leads to improved densification during sintering and requires a considerably shorter time cycle compared with

conventional methods of sintering. Thus, this process holds promise to consolidate difficult-to-sinter materials faster, and offers the opportunity to retain the fine microstructure of the powders. Groza et al. [22,24,25] reported plasma activated sintering of additive free sub-micron AlN powders to near theoretical density with clean boundaries in 5 min. The rapid activation and concentrated heating of the powder particle surfaces due to sparking may be very effective in debonding the surface oxides and cleaning the sintering particle surfaces, thereby creating activated AlN surfaces [22,26,27]. The freshly cleaned and activated surfaces enhance diffusion and shrinkage during subsequent densification. In a previous study [21], the SPS technique was applied to sinter CaF_2-doped AlN, and the result is promising. A thermal conductivity value of 129 $W \cdot m^{-1} \cdot K^{-1}$ was obtained for a 3 wt.% CaF_2 sample sintered at 1800ºC for 5 min.

In this paper, relatively low temperature (1500 and 1600 °C), and rapid densification of Sm_2O_3-doped AlN is carried out using the SPS technique. The objectives of the article are: to prepare dense AlN ceramics with high thermal conductivity through Spark Plasma Sintering (SPS) technique, and to study the effect of various SPS conditions on the properties like microstructure, relative density, thermal conductivity and dielectric constant of the sintered AlN samples, where different soak times, sintering temperatures and number of SPS cycles are employed as sintering conditions.

2. EXPERIMENTAL PROCEDURE

Commercial AlN powder (A1120, Cerac Specialty Inorganic, purity 99.8%) was used for the study. The average grain size was measured with a laser particle size analyzer. The analysis was performed with ultrasonic option to de-agglomerate the powder. Figure 1(a) shows the particle size distribution of AlN powder, which has an average particle size of 2.77 μm. Particle morphology of the as-received powder is shown in Figure 1(b). The powder particles are generally irregular in shape

Samarium oxide (Sm_2O_3) powder was chosen as the sintering aid in the sintering of AlN. The powder was commercially obtained from Cerac Specialty Inorganic, USA, and it has a purity of 99.9%. Samaria contents of 1 wt. % and 3 wt. % are used for the AlN-Sm_2O_3 system. To prepare the AlN-Sm_2O_3 system, the powders were added in the required concentration in a plastic milling jar, and the mixture was dry-mixed by ball milling for 24 hours at 30 rpm. The particle size distribution of the milled powder is shown in Figure 2(a), and the morphology of the milled powders is shown in Figure 2(b).

The spark plasma sintering of pure AlN and AlN-Sm_2O_3 powder system was carried out using the Dr. Sinter® Model 1050 SPS system by the Sumitomo Coal Mining (SCM) Pte. Ltd. SPS is performed in low vacuum for 1 and 3 minutes, and at 1500° and 1600°C. Both the heating and cooling rates of 100°C/min are used, and up to 2 SPS cycles are employed in the SPS study. The SPS conditions for different powder systems are listed in Table I. The densities of the samples were measured using the density measurement apparatus. The dielectric constant is measured with the Impedance/Network analyzer to obtain the parallel capacitance of the sample. The thermal diffusivity of the sample was measured by the laser flash method, which was performed at an average room temperature of 25°C. X-ray diffraction (XRD) analysis was performed to identify the phases present in the samples. And scanning electron microscopy (SEM) was used to evaluate the microstructure of the samples.

3. RESULTS AND DISCUSSION

Figure 3 shows the effect of the variation of temperature and SPS cycle on the relative density of pure AlN and 1 wt.% -Sm_2O_3-doped AlN samples. The level of densification increases significantly with increasing sintering temperature for pure AlN samples with a relative density about 96% at 1600°C. This value is higher than that of the AlN samples sintered by conventional N_2 protected process, which is about 75% at 1800°C for 3 hours [28]. Furthermore, conventional sintering of AlN resulted in a final relative density of about 95% at 1930°C for 30 hours [29]. These comparisons clearly indicate that higher densification can be achieved through the SPS process. It has been suggested that the high density achieved in the process may be attributed to high temperature spark plasma generated by pulsed high dc current and resistance heating together with pressure application [25].

Figure 3 also shows that, for a given temperature, higher density was attained in the double (3+3) SPS cycle than that achieved in single SPS cycle. This is because, in the double SPS cycle process, the powder compact is exposed to high temperature plasma for a longer period of time. It is thought that the high temperature plasma helps the purification of the powder surface and the bulk mass transport between powders, and hence contributes to further densification. Therefore, in this case, double SPS cycle has shown improvement in the final density of AlN. But for pure AlN samples, single SPS cycle process with longer soaking time (3 minutes) still gives a higher densification than the double SPS

cycle process with shorter soaking time (1 minute). This tendency is changed for 1%wt Sm_2O_3 doped sample. The double SPS cycle process with shorter socking time (1 minute) gives higher densification than the single SPS cycle process with longer socking time (3 minutes) at the sintering temperature of 1550˚C and 1600˚C. This indicates that multiple SPS cycles can enhance the further densification of highly densified AlN samples.

Figure 4 demonstrates the effect of Sm_2O_3 sintering aid on relative density. It can be seen that the addition of Sm_2O_3 as sintering aids improved the relative density at the same sintering temperature of 1500°C. Compared to the relative density of pure AlN, the greatest improvement of 13.1% in relative density was observed in the 1 wt. % Sm_2O_3-doped AlN sample sintered at 1500°C. This clearly demonstrates that a mere 1 wt. % addition of Sm_2O_3 greatly improves the sinterability of AlN. The further additional amount of additive content (3 wt. % Sm_2O_3) did not result in further increase in the final density of the sample.

The XRD results for 1 wt. % Sm_2O_3 doped AlN sample sintered at 1500°C with both single and double SPS cycle shows that, no observable peaks of the secondary phase ($SmAlO_3$) were identified for the 1 wt. % Sm_2O_3 sample sintered in single SPS cycle whereas peaks of $SmAlO_3$ can be seen in the pattern derived from the double SPS cycle of the sample. Similar observation was obtained for the patterns of 3 wt. % Sm_2O_3 doped sample sintered at 1500˚C with single and double SPS cycles.

Apparently, peaks of $SmAlO_3$ were not found in the sample prepared by single SPS cycle, indicating that the amount of the secondary phase was probably too minute in these samples. The formation of the second phase is due to the reaction between Sm_2O_3 and Al_2O_3 on the particle surface. Hence, significant amount of Al_2O_3 would have been eliminated if the amount of secondary phase present in the sintered bulk were high. Consequently, thermal conductivity would correspondingly be enhanced significantly due to the precipitation of the Al_2O_3 compounds.

During the sintering of AlN, the use of Sm_2O_3 leads to formation of a liquid phase in the Sm_2O_3-Al_2O_3–AlN system. These liquid phases facilitate densification by liquid phase sintering. It is known that the presence of the liquid phase improves mass transport rates during sintering [30]. In addition, the liquid phase exerts a capillary pull on the particles that is equivalent to a large external pressure [30]. And higher density can be achieved in the samples prepared under the same sintering conditions with the use of Sm_2O_3.

It is worth noting that the SPS processing temperature applied in this research is decidedly lower than the liquidus temperature in the conventional AlN-Al_2O_3-Sm_2O_3 phase diagram. It is reported that

the AlN-Al_2O_3-Y_2O_3 system has a liquidus temperature of 1685°C, while the Al_2O_3-Y_2O_3 system has a solid-liquid line at 1750°C. As the Sm_2O_3-Al_2O_3 system has a solid-liquid line at 1825°C, it is expected that the liquidus temperature of the AlN-Al_2O_3-Sm_2O_3 is higher than 1680°C. But, from the XRD results, it is obvious that the liquid secondary phase $AlSmO_3$ is formed at 1500°C in the SPS process. This abnormal phenomenon may be caused by two mechanisms: first, SPS is carried out under reduction vacuum atmosphere, the solid-liquid line may goes down under this atmosphere; second, the existence of high temperature plasma generated by pulsed dc current may bring the solid-liquid line of the system even lower, so that the liquidus temperature of the AlN-Al_2O_3-Sm_2O_3 system is below 1500°C in SPS process.

In order to quantitatively estimate the amount of $SmAlO_3$ in the sintered sample, the Rietveld refinement is carried out for the XRD result of 1 wt. % Sm_2O_3-doped and 3 wt. % Sm_2O_3-doped AlN samples sintered at 1500°C with 1 minute for double SPS cycle, as shown in Figure 5. The refinement result is given in Table II. It is shown that the concentration of $SmAlO_3$ in the sample is still less than the concentration of Sm_2O_3 added into the powder systems.

Figure 6 shows the thermal conductivity of both AlN and 1 wt. % Sm_2O_3–doped AlN as a function of sintering temperature. The thermal conductivity of 3 wt. % Sm_2O_3–doped AlN sintered at 1500ºC for 3 minutes (single SPS cycle) is as shown in Figure 6. It can be seen that the thermal conductivity of samples increased with sintering temperature. AlN samples exhibited thermal conductivity ranging from 47 W/m·K to 56 W/m·K. On the other hand, 1 wt. % Sm_2O_3-doped AlN samples showed significantly higher thermal conductivity as compared to that of pure AlN samples for a given temperature. The 3 wt. % Sm_2O_3–doped AlN sample shows a thermal conductivity value as high as 119 W/m·K after SPS at 1500ºC for 3 minutes, one SPS cycle. This confirmed that Sm_2O_3 is effective in improving the thermal conductivity of AlN by low temperature SPS.

Sm_2O_3 can react with Al_2O_3 on the surface of AlN powder to form aluminium samarium oxide ($SmAlO_3$). As a result, the amount of dissolved oxygen in AlN lattice is reduced and this leads to higher thermal conductivity in the sintered sample. Thermal conductivity is expected to improve when greater amount of additive is added.

The effect of SPS cycle on the thermal conductivity of 1 wt. % Sm_2O_3–doped AlN is also shown in Figure 6. The enhancement in thermal conductivity by double SPS cycle is evident in the sintering of 1 wt. % Sm_2O_3.

The dielectric constant of 1 wt. % Sm_2O_3-doped AlN samples exhibited a bit higher than that of pure AlN sample at a given temperature, as can be seen in Figure 7. The dielectric constant of 1%wt Sm_2O_3 increased with sintering temperature, attaining a value of 9.77 at 1600°C. This value is relatively high as compared to the earlier specified range. Although dielectric constant is dependent on the second phase, the additive content of 1% in weight is relative low and its effect is considered negligible.

Figure 7 also shows the effect of SPS cycle on the dielectric constant of pure AlN and AlN with 1 wt. % Sm_2O_3. The dielectric constant slightly increased with double SPS cycle.

Figure 8 show the dependence of elastic modulus of 1 wt. % -Sm_2O_3–doped AlN on sintering temperature and SPS cycle. It can be seen that the elastic constant generally increased with temperature expect those sintered by single SPS cycle. Samples prepare by single SPS cycle showed no significant difference in the measured values for the studied temperature. On the other hand, the elastic modulus increased with temperature for samples prepared by double SPS cycle. In the case of 1+1 SPS cycle, the elastic modulus attains a value of 338GPa at 1600°C. The above observation could probably be explained by the dependence of elastic modulus on the relative density of the sample. The decrease in the measured value could be due to the surface porosity induced by the volatilization of the liquid phase. The presence of such porosity reduced the effective resistance of the sample to the applied load exerted by the indenter tip, and thus it leads to a lower elastic modulus.

Figure 9 presents the effect of temperature, Sm_2O_3 additive and SPS cycle on the microstructure of AlN samples. It can be seen that porosity and pore size decreased as the sintering temperature and the number of SPS cycles increased. Figure 9(d) shows the SEM micrographs of 3 wt. % Sm_2O_3 sintered at 1500°C. The microstructure contained rather fine grains with an average particle size of 6 μm. The density is greatly improved by the addition of additive. The distinct faceted features of the grains clearly demonstrate the sintering in the presence of liquid phase. The well faceted nature of the AlN grains leads to a better planar contact between the AlN grains. It is expected that the high planarity of the grain lead to the flattening of the AlN-AlN grain contacts, which can result in an increase in thermal conductivity [31]. This improvement in the topological feature of the grain, coupled with the elimination of the oxide layer by the liquid phase could probably account for the high thermal conductivity (~118 W/m·K) achieved in this sample.

4. CONCLUSION REMARKS

Spark plasma sintering (SPS) of aluminum nitride (AlN) is carried out with 1 – 3 wt % samarium oxide (Sm_2O_3) as sintering additive. The effect of SPS temperature and SPS cycle on the microstructure, density, thermal conductivity and dielectric constant of AlN is studied. The result shows that, SPS process is capable of producing dense AlN ceramics at a low sintering temperature in a relatively short sintering time. Increasing temperature and SPS cycle number have been shown to directly increase the density of the sintered samples. The addition of 1 wt. % Sm_2O_3 improves density of AlN significantly. Thermal conductivity of AlN samples improves with increasing SPS cycle and sintering temperature. Addition of Sm_2O_3 greatly improves the thermal conductivity of AlN sample, with the 3 wt. % -Sm_2O_3-doped AlN sample giving a thermal conductivity about 118 W/mK at a sintering temperature of 1500°C for 3 minutes. Dielectric constant of the sintered AlN samples is dependent on the relative density of the samples.

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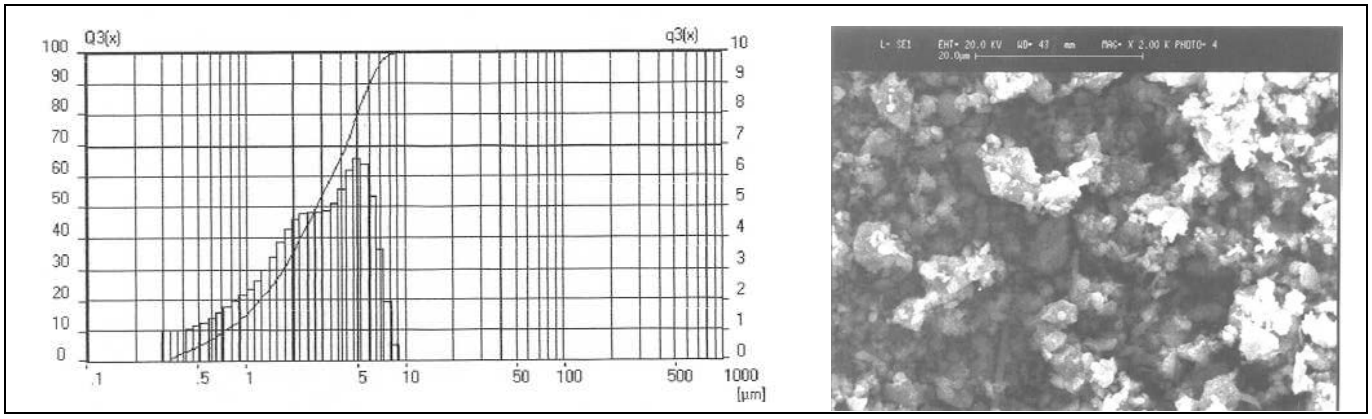
Table I SPS conditions for various powder systems

Sample	SPS Temperature (ºC)	Soak Time (minutes)	SPS Cycle
AlN, Single	1500, 1550, 1600	3	1
AlN, 1+1	1500, 1550, 1600	1	2
AlN, 3+3	1500, 1550, 1600	3	2
1%Sm_2O_3, Single	1500, 1550, 1600	3	1
1%Sm_2O_3, 1+1	1500, 1550, 1600	1	2
1%Sm_2O_3, 3+3	1500, 1550, 1600	3	2
3%Sm_2O_3, single	1500	3	1
3%Sm_2O_3, 1+1	1500	1	2

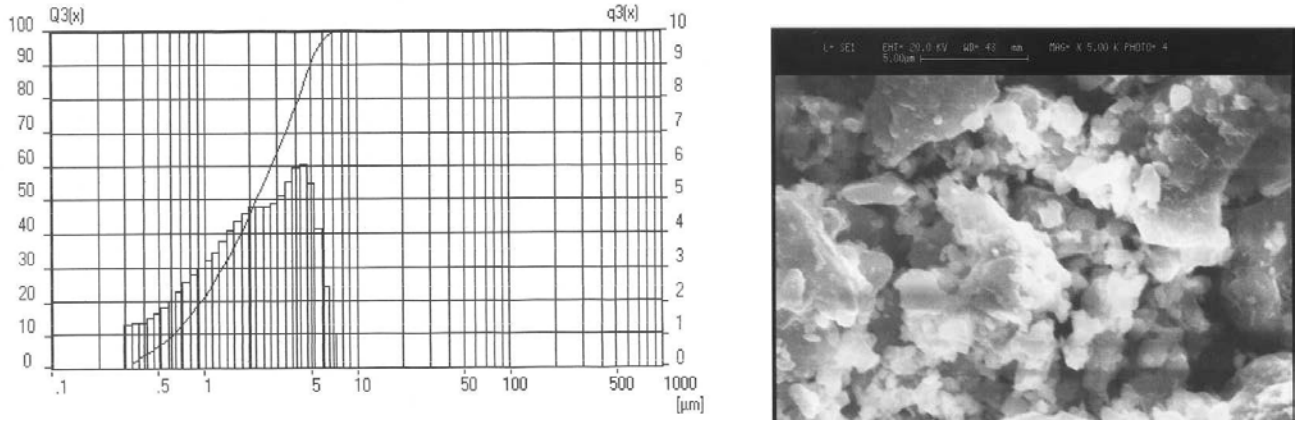
Table II. Phase composition of Sm_2O_3-doped AlN system after double-cycle spark plasma sintering with sock time of 1 minute for each cycle

Phase	1%wt $\mathbf{Sm_2O_3}$, 1+1	3%wt $\mathbf{Sm_2O_3}$, 1+1
AlN	99.5 ± 0.1%	98.1 ± 0.2%
$SmAlO_3$	0.5 ± 0.2%	1.9 ± 0.5%



(a) Particle size distribution **(b) morphology**

Figure 1. Particle size distribution and morphology of AlN powder



(a) Particle size distribution **(b) Morphology**

Figure 2. Particle size distribution of AlN-1%wt Sm_2O_3 system

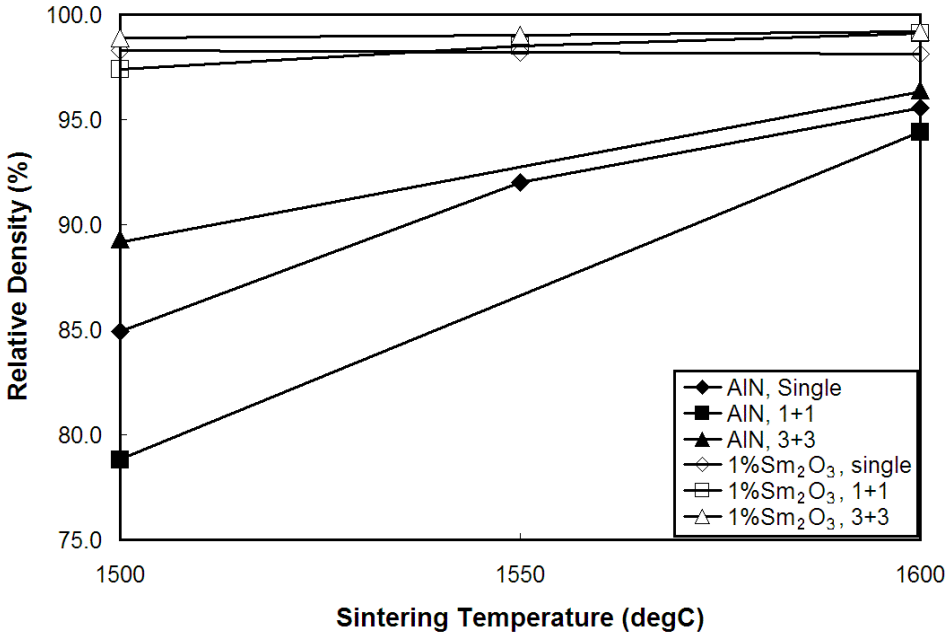


Figure 3. Relative density as a function of sintering temperature, SPS cycle and Sm_2O_3 addition (1%wt). The theoretical density for pure AlN and 1%wt Sm_2O_3 doped AlN is 3.261 g/cm^3 and 3.299 g/cm^3 respectively.

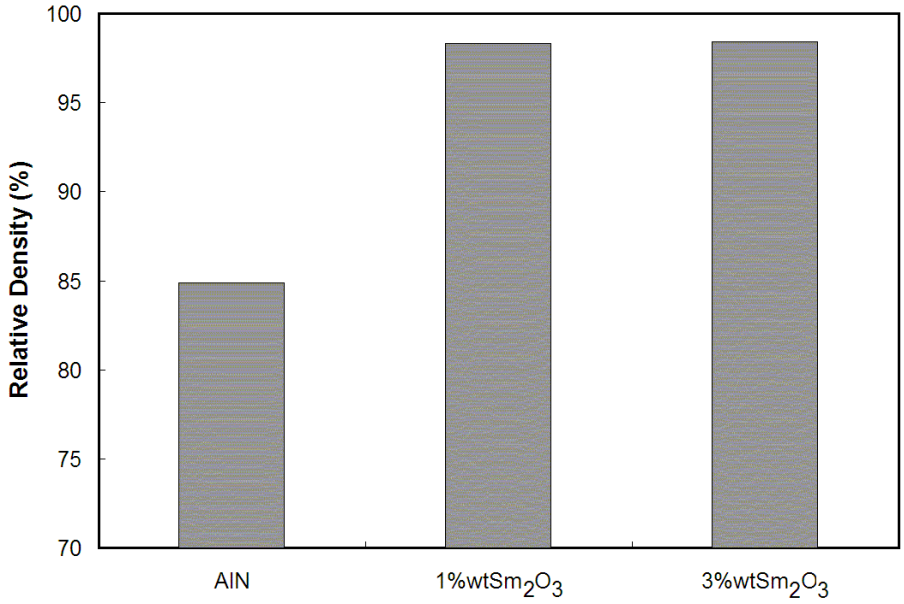
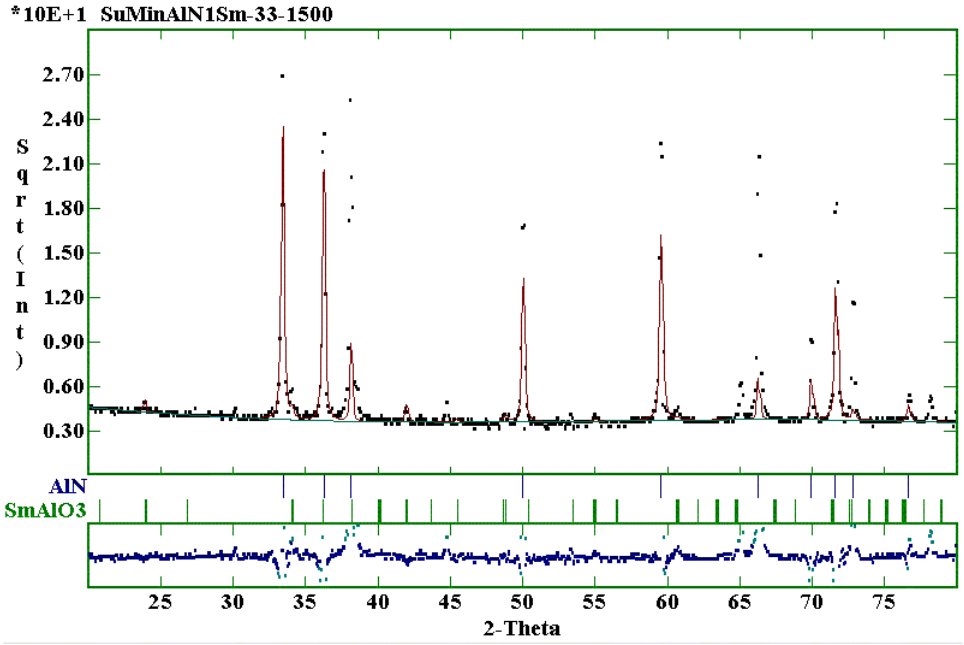
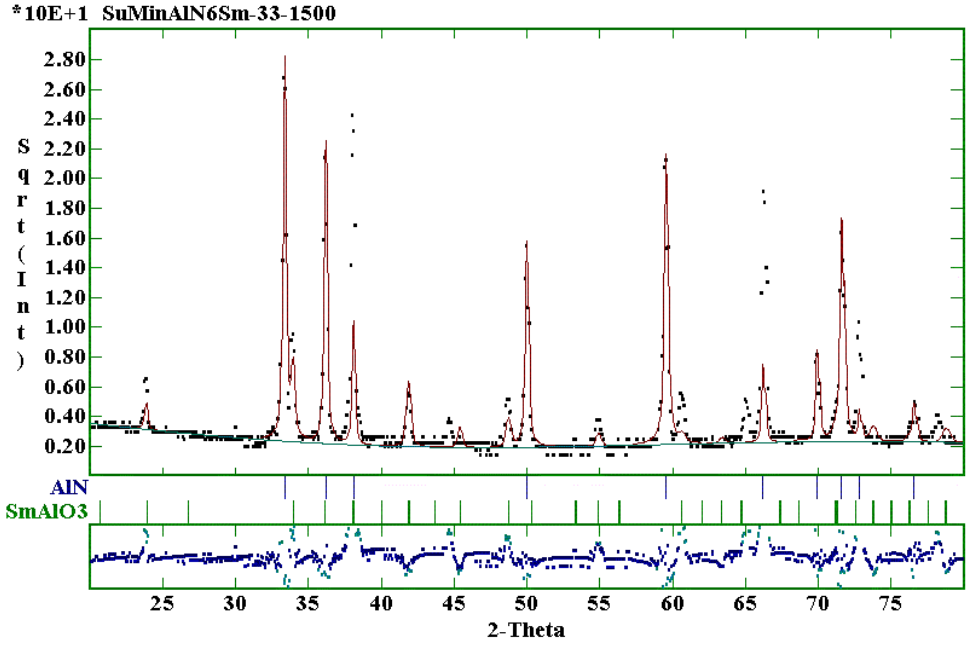


Figure 4. Relative density of various powder systems sintered at 1500°C for 3 minutes, single cycle. The theoretical density for pure AlN, 1%wt and 3%wt Sm_2O_3 doped AlN is 3.261 g/cm^3, 3.299 g/cm^3 and 3.367 g/cm^3 respectively.



(a) 1%Sm_2O_3, 1+1,



(b) 3%Sm_2O_3, 1+1,

Figure 5. Reitveld refinement of XRD result of 1%wt-Sm_2O_3-doped and 3%wt-Sm_2O_3-doped AlN samples sintered at 1500°C with 1 minute for double SPS cycle

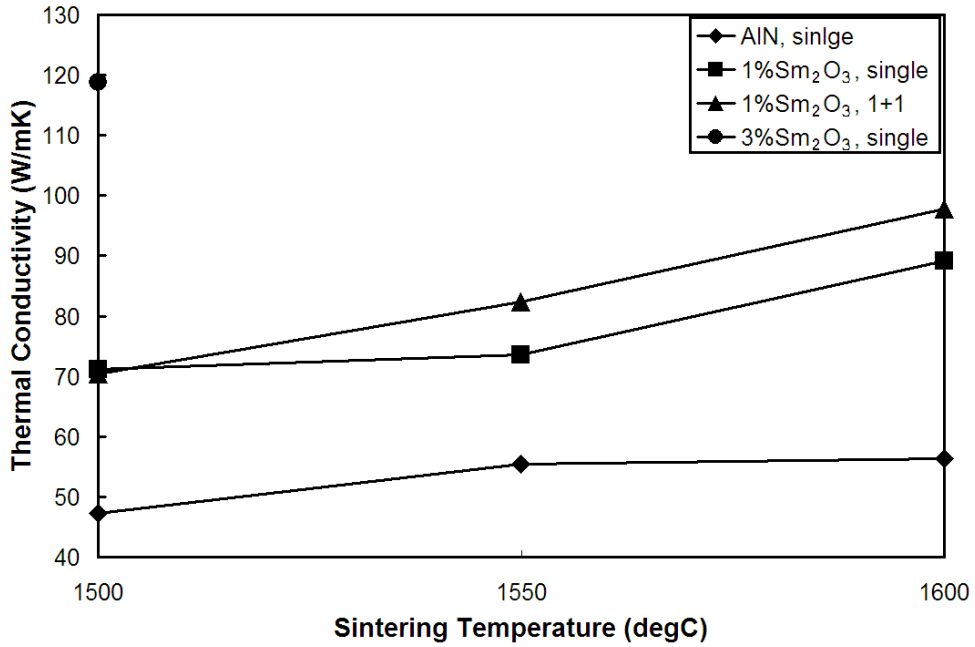


Figure 6. Thermal conductivity as a function of sintering temperature, SPS cycle, and Sm_2O_3 addition

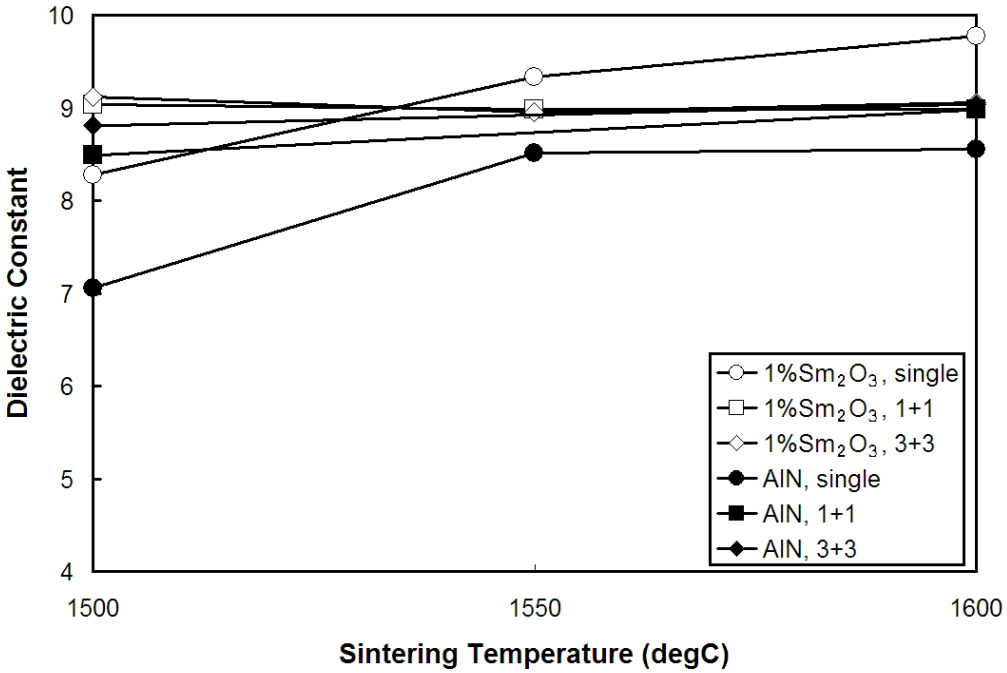


Figure 7. Dielectric constant of AlN and 1%wtSm_2O_3 as a function of temperature and SPS cycle

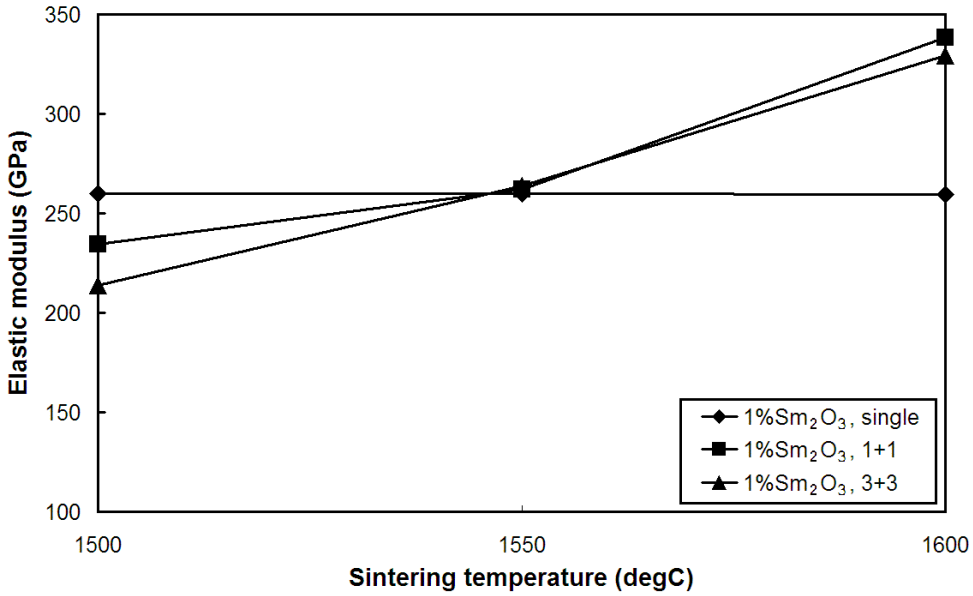
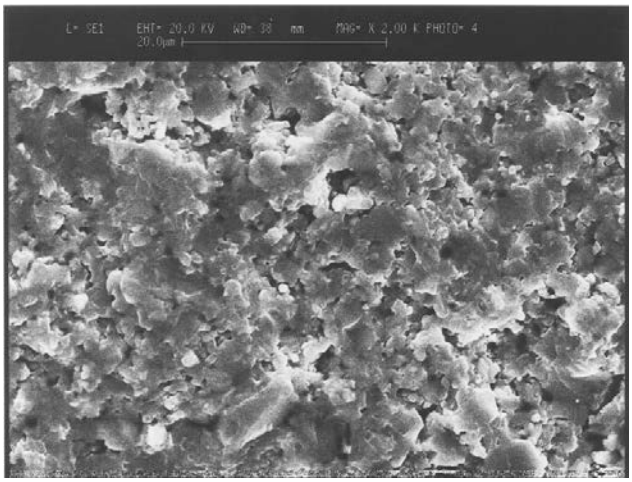
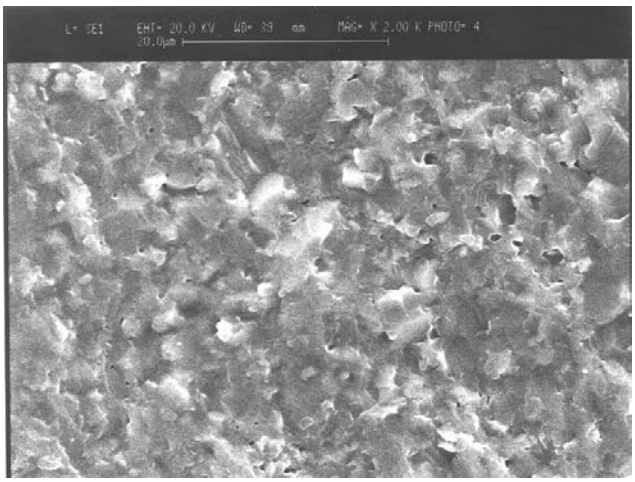


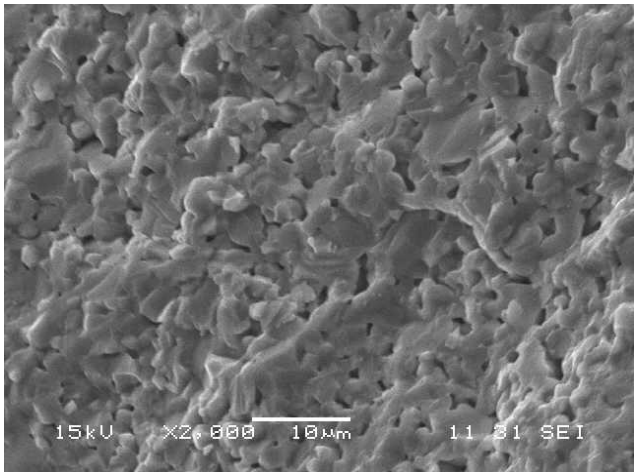
Figure 8. Dependence of elastic modulus on temperature and SPS cycle for 1%wt-Sm_2O_3-doped AlN samples



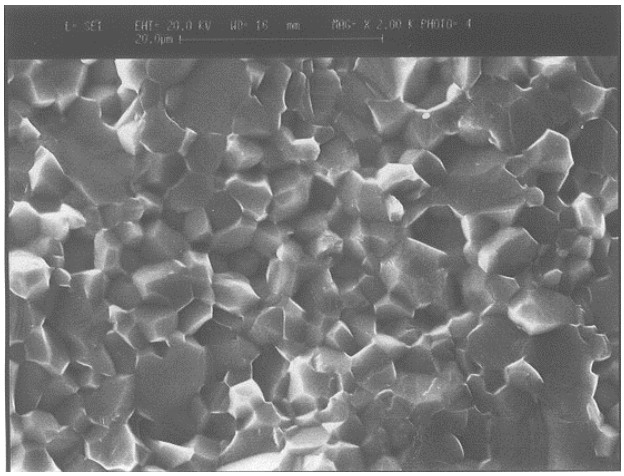
(a)AlN, Single, 1500°C



(b) AlN, Single, 1600°C



(c) AlN, 1+1, 1500°C



(d) 3%Sm_2O_3, Single, 1500°C

Figure 9. Effect of SPS parameters on the microstructure of AlN sample