Fast densification mechanism of bimodal powder during pressureless sintering of transparent AlON ceramics

Yingchun Shan a,*, Zhaohui Zhang a, Xiannian Sun a, Jiujun Xu a,*, Qinghua Qin b, Jiangtao Li c

a Department of Materials Science and Engineering, Dalian Maritime University, Dalian 116026, China
b Research School of Engineering, Australian National University, Acton, ACT 2601, Australia
c Technical Institute of Physics and Chemistry, Chinese Academy of Sciences, Beijing 100080, China

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Both bimodal and unimodal AlON powders were pressureless sintered and their phase assemblages and microstructure evolutions at 1400–1900 °C were monitored to reveal the fast densification mechanism of bimodal powder. Phase assemblages analysis of samples sintered at 1400 °C suggests that fine AlON particles were transformed into hexagonal α-Al2O3 and AIN, while coarse AlON particles were transformed into cubic γ-Al2O3 and AIN. Correspondingly, only a small fraction of the unimodal mixture was γ-Al2O3, while the relatively unstable γ-Al2O3 kept dominating the bimodal samples and most of them were directly reformed into AlON at a lower temperature of 1600–1700 °C for their similarity in crystal structure. As a result, a bimodal grain size distribution was kept in the whole sintering process of the bimodal sample, which led to its fast densification. Using the bimodal AlON powder, highly transparent AlON ceramics were fully pressureless sintered at 1820 °C after holding for 2.5 h.

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1. Introduction

In recent years, transparent aluminum oxynitride (γ-AlON) with cubic structure has attracted a growing interest due to its excellent strength and high optical transparency, for that has many superior applications [1–5]. Transparent AlON can be fabricated by using either pressing or pressureless sintering technique. Pressureless sintering technique is favored for its low cost and easy shaping in manufacturing transparent AlON compared to hot pressing or hot isostatic pressing techniques [6,7]. Nevertheless, pressureless sintering transparent AlON by using traditional powder requires a high sintering temperature about 1920–2000 °C and a long holding time of ≥7 h, which is still high energy consumption.

Recently, we proposed a new starting powder having bimodal particle size distribution (PSD) to further cut the pressureless sintering costs of transparent AlON. Our work has demonstrated that this powder can be fast sintered for its fast heating of 40 °C /min (compared to ~10 °C /min in other works) and a marginally lower sintering temperature of 1880 °C. More importantly, the holding time of pressureless sintering transparent AlON using this bimodal powder can be remarkably shortened to 1.5–2.5 h [8]. The fast sintering of the transparent AlON ceramics using the new powder is mainly attributed to fast surface diffusion during heating, due to the initial bimodal PSD of starting powder, and fast grain boundary diffusion during both heating and holding, due to the transient bimodal grain size distribution of AlON. It was noticed that fast heating and high relative density up to 99.11% once heating to 1880 °C without holding indicated that the powder can be fast densified. However, the fast densification mechanism of this bimodal powder was not very clear because no observation and measurement have been made before 1880 °C in that work. In this paper, a serial of experiments were performed at 1400–1900 °C for both the bimodal and unimodal powder, and the densification process of both powders was monitored to reveal fast densification mechanism of the bimodal powder.

Generally, contact fraction of solid particle, nucleation rate and sintering temperature are recognized as the key factors affecting the solid state reaction rate. Contact fraction of solid particle is controlled by both particle size and particle size distribution. Meanwhile, experiment and theoretical analysis revealed that the phase transformation kinetics is largely governed by the heterogeneous nucleation that is strongly sensitive to the driving force, dictated by
the majority phase of the starting powder, and that coherent nucleation is often favored when abundant isotrostructure nucleation sites are available, i.e., nucleation will be easier if one or more reactants have the similar structure with the product [9–11]. Higher temperature and long holding may be needed to eliminate the defects in the product if grain boundary and volume diffusion during holding is slow. Therefore, increasing contact fraction of powder or grain, controlling the crystal structure of intermediate reactants can accelerate nucleation and phase transformation at low temperature. Fast grain boundary and volume diffusion during heating and holding is also favored to shorten holding time.

To obtain high contact fraction of powder, reducing particle size is a good choice in some cases. However, it is very costly to obtain the particle fine enough for most of ceramics, and the large shrinkage caused by fine particle sintering usually are harmful to sample. Alternatively, one can manipulate the particle size distribution by mixing the fine particle with a fraction of coarse particle to improve contact fraction of raw powder. As a result, a mixture having bimodal PSD has been widely studied and applied in solid state reaction. Research work on the effects of powder PSD on compact density suggested that bimodal PSD mixtures provide higher green densities due to its improved gap filling and particle rearrangement ability [12–17]. According to theoretical calculation, particle rearrangement is mainly carried out by surface diffusion in its sintering process, which is associated with three types of particle contact, i.e., large–large, small–large and small–small [16]. Moreover, the rearrangement parameters θ (representation of rearrangement, the angle between the real motion vector of particle and the mean field strain vector associated to particle) calculated by Martin [17] also showed that the greatest value of θ was reached for the particle assemblies characterized by a domination of large–large and small–large contacts, which is the exact case for the bimodal powders. Therefore, the proposed bimodal AlON powder can lead to fast pressureless sintering benefited from both its increased contact fraction and improved particle rearrangement ability associated with particle contact type.

In the sintering process of transparent AlON ceramics from AlON powder, AlON particle is firstly transformed into Al₂O₃ and AlN at low temperature which then forms AlON again at high temperature (AlON is thermodynamically unstable below a temperature between 1600 °C and 1640 °C [18], which means that Al₂O₃, AlN or AlON may coexist in the mixtures during heating to sintering temperature). In the homogeneous regions of transparent AlON characterized by cubic crystal structure, the content of Al₂O₃ is 67–80 mol% (depending on the temperature) [19]. It is evident that Al₂O₃ is the main reactant in the mixture to be sintered into AlON. As a matter of fact, Al₂O₃ can be present in many kinds of crystal structures, such as cubic η-Al₂O₃, hexagonal α-Al₂O₃, and etc. It is worth noting that η-Al₂O₃ is more unstable than α-Al₂O₃ at high temperature. In addition, it is well known that transparent AlON ceramics has a cubic crystal structure for its high transparency, which implies that γ-Al₂O₃ has faster phase transformation kinetics than α-Al₂O₃ because of the similarity in crystal structure between η-Al₂O₃ and transparent AlON. Therefore, it appears that γ-Al₂O₃ is favored in pressureless sintering of transparent AlON ceramics because it could be faster transformed into AlON at a lower temperature compared to α-Al₂O₃. However, the reduction mechanism of Al₂O₃ from AlON powder is not well understood, which makes it hard to control the crystal structures of transformed Al₂O₃. Moreover, η-Al₂O₃ may be transformed into α-Al₂O₃ before it can be transformed into AlON at high temperature.

One possible way of controlling the phase of Al₂O₃ transformed from AlON powder is through adjusting the particle size. Study of particle size effect on crystal structure of Y₂O₃ particles synthesis revealed that there is a critical particle diameter of approximate 1.5 μm in the formation of phase of Y₂O₃ particles [20]. Particles significantly smaller than the critical diameter were all monoclinic, while those significantly larger were all cubic. The particle size effect was also reported for heavy rare earth sesquioxides RE₂O₃ (RE = Dy, Ho, Er, Tm and Yb) [21]. This particle size effect was attributed to the interplay between surface energy and polymorphism [22]. Therefore, it is highly possible for the proposed bimodal powder to form unstable cubic η-Al₂O₃ during heating, which in turn contributes to fast sintering of transparent AlON and lowering the sintering temperature. As a result, the phase assemblages of all the sintered samples during heating were characterized in this study. Indeed, unstable cubic η-Al₂O₃ was detected in the samples during heating.

Based on the above analyses and assumptions, in order to reveal the fast densification mechanism of the proposed bimodal powder, both a bimodal PSD AlON powder characterized by a domination of large–large and small–large contacts and a unimodal powder were prepared to study the microstructure evolution and the densification mechanism of AlON ceramics between 1400 and 1900 °C by pressureless sintering. For the purpose of comparison, the particle size of unimodal powder was chosen as same as that of small particle size of bimodal powder. At temperature 1700 °C, majority of hexagonal α-Al₂O₃ were found in the unimodal powder, but a large amount of cubic η-Al₂O₃ combined with a small fraction of α-Al₂O₃ were detected in the bimodal mixtures. By using the bimodal powder, AlON ceramics having high transparency were pressureless sintered at 1820 °C after holding for 2.5 h. The maximum infrared transmittance for 3 mm thickness sample was up to 82.1% (94.0% of theoretical in-line transmittance). The fast densification mechanism of the bimodal AlON powder was presented accordingly.

2. Experiment procedure

2.1. Preparation of bimodal and unimodal PSD AlON powders

AlON powder was firstly synthesized by carbothermal reduction and nitridation (CRN) method (detailed fabrication process were given in [8]), then 0.5 wt% Y₂O₃ (Grade C, Starck, Germany) was added into the obtained AlON powder. Using Si₃N₄ ball (having 5, 8 and 10 mm diameter. weight ratio at 1.7:1.1:1.3) in milling media, keeping the ball-to-powder weight ratio at 1:1. 50 g of mixture powders of AlON and Y₂O₃ were ground in absolute ethyl alcohol (weight ratio: absolute ethyl alcohol/powder = 3.93) by ball mill into both bimodal (at ~1.1 μm and ~2.2 μm) and unimodal (at ~1.1 μm) powder. It should be pointed out that 170 rpm for 24 h of ball mill was performed to obtain the bimodal powder, and 250 rpm for 36 h of ball mill to gain the unimodal powder. The morphology of the milled powders were observed by scanning electron microscope (FSEM; supra 55, Zeiss, Germany) as shown in Fig. 1. The particle size distribution of the obtained powders was measured using laser particle size analyzer (Mastersizer 2000, Malvern, UK) and their PSD are shown in Fig. 2.

2.2. Sintering of AlON powders

Two groups of sample, i.e., the bimodal and unimodal samples, were prepared using the obtained AlON powder. For each sample, 1.6 g of the corresponding AlON powder was individually packed into a pellet of 13 mm in diameter under 50 MPa. The pellet was then pressureless sintered within a graphite furnace in an atmosphere of 0.1 MPa N₂. For the purpose of comparison, two identical samples of the bimodal powder together with another two identical samples of the unimodal powder were put into graphite furnace each time, and were heated at a heating rate of 40 °C/min to 1400 °C, 1500 °C, 1600 °C, 1700 °C, 1800 °C and 1900 °C, respectively.
2.3. Characterization of sintered samples

The phase assemblages of the samples heated to different temperature were characterized by X-ray diffractometry (XRD; D/Max-ULTima+, Rigaku, Japan) using Si as an internal standard. The polished samples of bimodal powder fabricated at 1800 °C, 1820 °C and 1900 °C were hot etched at 1640 °C for 40 min in vacuum for micrographs observation. The SEM and optical microscope (GX51, Olympus, Japan) were employed to observe the morphology of all the samples. Average grain sizes of AlON were statistically calculated by using Image Pro plus software, where the average grain size was the average value of diameters passing through objects’ centroid.

The bulk density of all the samples sintered at different temperature was measured by Archimedes principle. The samples of bimodal powder sintered at 1820 °C for 2.5 h were ground and polished on both sides to a thickness of 3 mm for optical transmittance measurement. Optical transmittance in the wave of 2500–6000 nm was recorded by a Fourier transform infrared spectroscopy (FTIR; Nexus 670, Thermo Nicolet, USA).

3. Results and discussion

3.1. Morphology of AlON powders

From Figs. 1 and 2, it can be seen that the AlON powder synthesized by CRN was successfully milled into both a bimodal (at ~1.1 μm and ~2.2 μm) and a unimodal (at ~1.1 μm) powder in this paper. The unimodal powder has the equivalent size with that of the fine particle in the bimodal powder. As calculated in the previous paper, the bimodal PSD AlON powder produced in this paper is in a domination of large–large and small–large contacts [8], which can improve its gap filling ability and is helpful to its rearrangement resulted from surface diffusion at the early and middle stage of sintering.

3.2. Microstructure and phase assemblages evolution

To better understand microstructure and phase assemblages evolution of the sintered samples at different temperature, the SEM images and the XRD patterns of both the bimodal and unimodal samples after pressureless sintering at 1400–1900 °C without holding are presented in Figs. 3 and 4, respectively.

At 1400 °C, grain morphology of both the bimodal and unimodal powder developed toward sphere (Fig. 3a and b) compared to the starting AlON powders shown in Fig. 1. However, the grain size and grain size distribution was found strongly depends on the PSD of the starting powder. A microstructure characterized by bimodal grain size distribution and limited grain growth was observed in the bimodal sample (Fig. 1a vs. Fig. 3a), on the contrary, a uniform grain size and significant grain growth was observed in the unimodal sample (Fig. 1b vs. Fig. 3b). As a result, the average grain size of the unimodal sample is larger than that of the bimodal sample, as shown in Fig. 5. Further detection of their phase assemblages showed that all the AlON powders were transformed into Al2O3 and AlN at 1400 °C. Both cubic η-Al2O3 and hexagonal α-Al2O3 can be found in both samples. However, it can be seen from Fig. 4a that the bimodal sample was dominated by cubic η-Al2O3, while only a small fraction of η-Al2O3 was detected in the unimodal sample. Moreover, all the AlN detected in both group of sample have a hexagonal crystal structure and its crystal structure didn’t change throughout the whole sintering process (from 1400 °C to 1900 °C).

The fraction of η-Al2O3 and α-Al2O3 in two groups of sample (as shown in Fig. 4a) and the similarity in PSD between the sintered sample at 1400 °C and its own starting powder suggests that

The furnace was cooled to room temperature to take out the sintered samples for testing and characteristic. In addition, one more pellet packed with the bimodal powder was also sintered at 1820 °C and held for 2.5 h.
Fig. 3. SEM images of the fracture surfaces of the samples heated to 1400 °C from bimodal (a) and unimodal powder (b), 1500 °C from bimodal (c) and unimodal powder (d), 1600 °C from bimodal (e) and unimodal powder (f), 1700 °C from bimodal (g) and unimodal powder (h), 1800 °C from bimodal (i) and unimodal powder (j), 1900 °C from bimodal (k) and unimodal powder (l), respectively.
fine AlON particles were transformed into $\alpha$-Al$_2$O$_3$ and AlN, while coarse AlON particles were transformed into $\eta$-Al$_2$O$_3$ and AlN. It is worth noting that the reduction mechanism of Al$_2$O$_3$ from AlON powder has not been thoroughly studied so far. However, the presence of $\eta$-Al$_2$O$_3$ will definitely be helpful to sinter the mixture into transparent AlON ceramics because they all have a cubic crystal structure.

Further heating up to 1600 °C, the observed grain morphology of two powders was respectively similar to that at 1400 °C and the grain size distribution kept its own original pattern of starting powder. It indicates that the main mass transport is surface diffusion for the bimodal powder at the early or middle stage of sintering. Correspondingly, surface diffusion combined with grain boundary diffusion contributed to the grain growth of the fine unimodal powder at this stage due to its high specific surface energy. On the other hand, $\eta$-Al$_2$O$_3$ cannot be detected in the unimodal sample from 1500 to 1600 °C, while $\eta$-Al$_2$O$_3$ still dominated in the bimodal samples. It is reasonable to assume that the small amount of $\eta$-Al$_2$O$_3$ found in the unimodal sample at 1400 °C was transformed into $\alpha$-Al$_2$O$_3$ because $\eta$-Al$_2$O$_3$ is unstable at this temperature. Accordingly, only part of $\eta$-Al$_2$O$_3$ in the bimodal sample may be transformed into $\alpha$-Al$_2$O$_3$ because majority of Al$_2$O$_3$ in the bimodal sample was still $\eta$-Al$_2$O$_3$. However, it is not clear to us why $\eta$-Al$_2$O$_3$ in the bimodal samples remained in a dominated fraction. Further investigation is under way to clarify this observation.

At 1700 °C, only $\alpha$-Al$_2$O$_3$ together with AlN can be found in the unimodal sample, which clearly demonstrates that $\alpha$-Al$_2$O$_3$ cannot be transformed into AlON at this temperature. Similarly, $\alpha$-Al$_2$O$_3$ and AlN can also be detected in the bimodal sample. However, $\eta$-Al$_2$O$_3$ can no longer be detected in the bimodal sample. Instead, a large amount of AlON was presented in the bimodal mixture. It indicates that most of $\eta$-Al$_2$O$_3$ in the bimodal mixture was transformed into AlON at 1700 °C with a possibility of a small amount of $\eta$-Al$_2$O$_3$ being transformed into $\alpha$-Al$_2$O$_3$ in this duration. It is evident that cubic $\eta$-Al$_2$O$_3$ is easier to be transformed into AlON at a lower temperature (between 1600 °C and 1700 °C) compared to $\alpha$-Al$_2$O$_3$ due to its instability at this temperature and similarity in crystal structure with transparent AlON ceramics. In addition, from 1400 °C to 1700 °C, the pores in the bimodal mixture observed from Fig. 3 are much smaller and less than that in the unimodal sample

![XRD patterns of the bimodal and unimodal samples heated to 1400 °C (a), 1500 °C (b), 1600 °C (c), 1700 °C (d), 1800 °C (e) and 1900 °C (f), respectively.](image-url)
due to its improved gap filling and rearrangement ability of bimodal PSD.

Grain morphology at 1800 °C for both samples changed dramatically due to the fast grain growth with all the α-Al₂O₃ being transformed into AlON because no α-Al₂O₃ can be detected in both group of sample (Fig. 4e). It is evident that α-Al₂O₃ can be transformed into AlON at a temperature between 1700 °C and 1800 °C, which is higher than that for η-Al₂O₃ (between 1600 °C and 1700 °C). Moreover, only a few small pores can be observed in the bimodal sample at 1800 °C (Fig. 3i) and many vivid large pores can be seen in the unimodal sample at the same temperature (Fig. 3j), which implies that the bimodal sample is more densified than the unimodal one. As a matter of fact, the curve of relative density vs. temperature shown in Fig. 6 confirms that the relative density of the bimodal sample (98.02%) is much higher than that of unimodal one (65.95%) at 1800 °C. From 1700 °C to 1800 °C, both groups of sample had a quick increase in relative density. Apparently, relative density of the bimodal sample (63.57–98.02%) increases faster than that of the unimodal one (49.25–65.95%). For the unimodal sample, it is the transformation from α-Al₂O₃ to AlON that contributes the most shrinkage in volume leading to densification. However, for the bimodal sample, only a small amount of α-Al₂O₃ was left in the mixture at 1700 °C, which is much less than that in the unimodal sample (Fig. 4d). Therefore it is obvious that the shrinkage in volume due to transformation from α-Al₂O₃ to AlON in the bimodal sample from 1700 °C to 1800 °C is very limited. More densification for the bimodal sample is due to grain diffusion and volume diffusion of AlON grains transformed by η-Al₂O₃ between 1700 °C and 1800 °C, resulting in a rapid growth in AlON grain size, which can be confirmed by Fig. 5.

With all the Al₂O₃ being transformed into AlON for both groups of sample at 1800 °C, only the diffusion of AlON grains can contribute to further densification of the product. The curve of average grain size vs. sintering temperature shown in Fig. 5 demonstrates that the average AlON grain size of bimodal sample grew from 6.76 μm to 15.32 μm, which is much faster than that of unimodal sample (from 3.5 μm to 9.25 μm) in the same duration with temperature being raised from 1800 °C to 1900 °C, i.e., average grain growth rate of 0.09 μm/°C vs. 0.06 μm/°C for bimodal vs. unimodal sample. It implies that AlON grain boundary diffusion and volume diffusion of the bimodal sample is much faster than that of the unimodal sample at the final stage of sintering. The pores observed in the bimodal sample at 1800 °C (Fig. 3i) are much smaller and less than that in the unimodal sample at the same temperature (Fig. 3j), and even much smaller than that in the unimodal sample at 100 °C higher temperature (Fig. 3l). Although a small amount of AlN can be detected in the bimodal sample at 1800 °C (as shown in Fig. 4e), these residual AlN were quickly dissolved into AlON before the sample was heated to 1900 °C (as shown in Fig. 4f). Microstructures of the hot etched bimodal samples at 1800 °C and 1900 °C given in Fig. 7 shows that the AlON grain at this stage has a transient bimodal grain size distribution. It illustrates that AlON grains in the bimodal sample can diffuse much faster (much more grain growth) and better (densified faster and had less and smaller defects, i.e., pores and residual AlN, which can be easier to be eliminated) than those in the unimodal sample in the final stage of sintering, which can only be attributed to its transient bimodal AlON grain size dis-
distribution inherited from the bimodal starting powder. In turn, the pressureless sintering of the bimodal sample into transparent AlON ceramics needs a much less holding time.

3.3. Fast densification mechanism of the bimodal AlON powder

From Fig. 6, it can be seen that the relative density of the bimodal sample is consistently higher than that of the unimodal one at the same sintering temperature from 1400 °C to 1900 °C. This can also be observed from the picture of all the samples sintered at 1400–1800 °C shown in Fig. 8, where the diameter of the bimodal sample was smaller than that of the unimodal one at the same sintering temperature. It is evident that the bimodal AlON powder was densified faster than the unimodal powder in the whole sintering process.

At the early or middle stage of sintering (before the reformation of AlON, <1700 °C), the bimodal sample has a better gap filling and rearrangement ability due to its bimodal PSD [17] of the Al2O3 and AlN mixture, which resulted in a higher relative density compared to that of the unimodal sample at the same temperature. Due to the presence of both α-Al2O3(transformed from the fine AlION particles) and η-Al2O3 (transformed from the coarse AlION particles) in the bimodal sample before the final stage of sintering, the transformation of AlON from Al2O3 didn’t accomplish simultaneously. Instead, η-Al2O3 was transformed into coarse AlION grain at a temperature between 1600 °C and 1700 °C and α-Al2O3 was transformed into fine AlION grain at a temperature between 1700 °C and 1800 °C. Different transform temperature of AlION from two types of Al2O3 enables the mixture to keep its bimodal PSD. As a result, the reformed AlION grains have a bimodal grain size distribution which contributes to a much faster (less holding) and better (less and smaller defects) densification at the final stage of sintering.

Although η-Al2O3 can be transformed into AlION at a lower temperature (between 1600 °C and 1700 °C), a high enough sintering temperature (between 1700 °C and 1800 °C in this paper) is still necessary to transform the α-Al2O3 in the bimodal mixture into AlION. Nevertheless, the sintering temperature of the proposed bimodal powder is lower than that of reported powder due to its improved grain boundary diffusion and volume diffusion at the final stage of sintering attributed to its transient bimodal AlION grain size distribution. Moreover, as shown in Fig. 9, the bimodal sample sintered at 1820 °C holding for 2.5 h shows too much light transmission than that of the present unimodal sample sintered at the same condition for 3 mm thickness sample (In fact, the unimodal sample is opaque). From Fig. 9, an up to 82.1% infrared transmittance (94.0% of theoretical in-line transmittance [8]) at a wavelength of ~3600 nm was achieved for the bimodal sample, which is evident that the proposed bimodal powder can be fully sintered into transparent AlION ceramics at 1820 °C with holding 2.5 h.

![Fig. 8. Optical images of the bimodal (B) and unimodal samples (U) heated to 1400 °C, 1500 °C, 1600 °C, 1700 °C, 1800 °C and 1900 °C.](image)

![Fig. 9. Transmittance of the bimodal and unimodal AlION samples sintered at 1820 °C held for 2.5 h, and the optical images of bimodal and unimodal samples (a) for 3 mm thickness sample, the microstructure of bimodal sample (b).](image)

4. Conclusions

1) Both a bimodal (at ~1.1 μm and ~2.2 μm) and a unimodal (at ~1.1 μm) AlION powder were successfully prepared by using ball mill. After these powders were heated to 1400 °C at a heating rate of 40 °C /min, the AlION powders were transformed into Al2O3 and AlN. Further phase assemblages analysis of both samples sintered at 1400 °C suggests that fine AlION particles were transformed into hexagonal α-Al2O3 and AlN, while coarse AlION particles were transformed into cubic η-Al2O3 and AlN. It indicates that the phase of Al2O3 transformed from AlION powder could be controlled by the AlION particle size.

2) Cubic η-Al2O3 was transformed into AlION at a temperature between 1600 and 1700 °C and hexagonal α-Al2O3 was transformed into AlION between 1700 and 1800 °C, the lower phase transformation temperature of η-Al2O3 to AlION is mainly benefited from the similarity in crystal structure between them.

3) The bimodal powder kept its bimodal PSD throughout the whole sintering process and therefore it can be densified faster and better than the unimodal powder. At the early or middle stage of sintering (before the reformation of AlION, <1700 °C), the bimodal sample has a better gap filling and rearrangement ability due to its bimodal PSD. At the final stage of sintering, AlION grains in the bimodal sample can diffuse much faster and better than those in the unimodal sample, which can only be attributed to the transient bimodal AlION grain size distribution inherited from the bimodal starting powder. In other words, it was the bimodal PSD that played a key positive role in the fast densification mechanism of bimodal powder.

4) The proposed bimodal powder can be fully sintered into transparent AlION ceramics at 1820 °C with holding 2.5 h.
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