Pulsed Electric Current Sintering and Its Employment for Transparent Polycrystalline Al_2O_3

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Pulsed electric current sintering (PECS) has become more and more important in material sciences as an advanced method for material fabrication. The remarkable features of PECS in comparison with almost other sintering techniques are the direct heating method and the assistance of pressure. Therefore, PECS has fast heating rate and this sintering technique can densify samples fast without a severe grain growth. The high density and fine microstructure of materials sintered by PECS help a lot in improving properties of materials, especially mechanical properties. Recently, transparent polycrystalline Al_2O_3 has attracted much attention among ceramic materials. PECS is a superior sintering method for fabrication of transparent polycrystalline Al_2O_3. This article briefly summarizes the features of PECS and its recent advances in fabrication of transparent polycrystalline Al_2O_3.

Keyword: Pulsed electric current sintering, two-step PECS, transparent polycrystalline Al_2O_3, microstructural heterogeneity.

Sintering is one of the excellent methods for shaping of materials. Conventionally, the powder is shaped by pressing in mold in order to form a green body. After that, the green body is sintered by an electrical furnace at high temperature. Conventional sintering showed low ability of densification due to the lack of assistance of pressure. Currently, powder materials can be densified by numerous pressure-assisted sintering techniques such as hot pressing (HP), hot isostatic pressing (HIP) or pulsed electric current sintering (PECS). Among those procedures, HP and HIP methods, which use an indirect heating structure like conventional sintering, often need a long heating time of several hours in order to lead to full densification [1]. In contrast, PECS, the latest developed pressure-assisted-sintering process, is a rapid densification process. This feature of PECS not only derives from the support of the uniaxial pressure but also comes from its direct heating method by electrical current. Therefore, it has a big potential to fully densify the powder compact with minimizing grain growth [2].

The schematical design of PECS is showed in Fig. 1. It simultaneously applies an electric pulse along with a uniaxial mechanical pressure in order to sinter the powder compact in a mold set. The applied electric pulse and mechanical pressure can be controled by a preset program by controlling the electric current or the in-situ measured temperature.

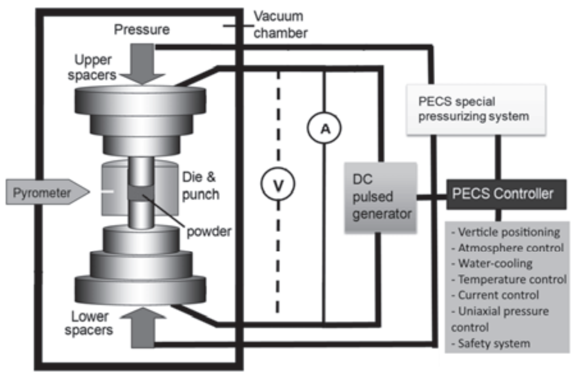

Fig. 1: Schematic diagram of pulsed electric current sintering.

In typical PECS methods, powders are poured into a die/punch set and heated by an electric current flowing through the die/punch and powder compact system. Either the powder or the die/punch set or both of them should be electrical conductive in order to guarantee the flow of the electrical current as a close circuit. For the same reason, all of the blocks, including electrodes, spacers, plungers and punches inserted in the circuit (if necessary), should be made of electrical conducting material (stainless steel, graphite, carbides, etc).

Conducting powders are heated both by Joule effect from the current within the powder compact and by heat transfer from the container and electrodes. Otherwise, the non-conductive powders are heated only through the second route. The use of graphite containers, which are mostly applied in the current PECS processes, limits the mechanical pressure levels to low values, generally 100 MPa. In spite of that limitation, the uniaxial pressure of 100 MPa is still high enough to densify in most cases. Therefore, the graphite dies and punches are the most widely used in PECS because of its simple machining. When the ultra high density is required, other materials with stronger mechanical properties can be applied for dies and punches, such as WC or SiC. Grasso et al. used a special design for the mold set of PECS, in which a smaller SiC mold was inserted inside a bigger graphite mold [3], as shown in Fig. 2. With the same design, they could sinter with PECS at a pressure of 500 MPa. Using the same design, Sokol et al. applied 400 MPa or Ratzker et al. applied 800 MPa in their PECS processes [4, 5].

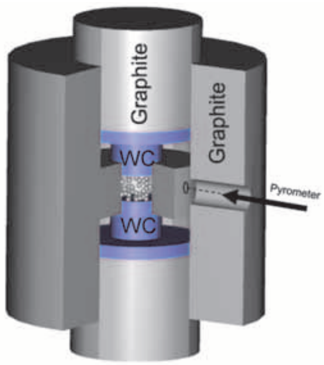

Fig. 2: Schematic of the high-pressure SPS device. Reprinted with permission from J. Am. Ceram. Soc., 93 (2010) 2460-2462. Copyright: @ John Wiley and Sons.

The biggest difference of PECS to other pressure-assisted sintering techniques is the application of the electric current through a power supply as the heating source, which leads to a very rapid and efficient heating. The heating rate during the PECS process depends on the geometry of the container and the sample ensemble, its thermal and electrical properties, as well as the electric power supply. Heating rates as high as 100 up to 600°C/min were achieved. In contrast, the typical heating rate of HP or HIP processes is often limited at a few °C/min. As a result, the processing duration of PECS is shortened in compared with HP or HIP processes, which suppresses grain growth in sintering. Hence, the most advantage of PECS processes is the efficient usage of the heat input, particularly when electrically insulating container is used and the electric current is applied for extremely short duration.

The temperature of PECS process is in-situ measured by a radiation thermometer focused on the surface of the die or by a thermocouple inserted into a hole on the outer surface of the die. The measured temperature at the surface of the die is usually lower than the actual temperature of the sample inside it. Depending on a number of factors such as thermal conductivity of the die and the sample, the heating rate, the applied pressure, the thermal insulation of the die from surrounding, etc, this temperature difference may become quite large. One of the problems of PECS processes is to achieve homogenous temperature distribution in order to achieve a homogeneous sample. In fact, current distribution and consequent temperature distributions within the sample are very sensitive to the homogeneity of the sample after sintering [6-8]. The inhomogeneity of temperature in PECS can be considered as its weakness in comparison with other sintering techniques. However, PECS is still used widely to prepare many kinds of materials such as advanced alloy materials, functional gradient materials, advanced ceramics, thermoelectric materials, nano-composites because of its advantages - rapid densification with minimized grain growth.

Aluminum oxide (Al_2O_3), commonly referred to alumina, is one of the most widely used engineering ceramic materials, especially its most stable crystalline structure, α-Al_2O_3 (or corundum/sapphire). Table 1 lists some of the properties of α-Al_2O_3, which was reported by Doremus [9].

Table 1: Physical and mechanical properties of α-Al_2O_3.

Properties	Typical values
Density	3.99 g/cm^3
Flexural Strength	379 MPa
Compressive Strength	2600 MPa
Hardness (HV)	1440 GPa
Fracture Toughness K_{IC}	3 $MPam^{1/2}$
Melting point	2054°C
Thermal Conductivity	35 W/mK
Coefficient of Thermal Expansion	$8.4\times10^{-6} K^{-1}$

With high melting point and chemically inertness, Al_2O_3 is widely used as high-temperature components, catalyst substrates or biomedical implants. Al_2O_3 is useful for bearings and cutting tools because of its excellent hardness, strength and good abrasion resistance. Electrical insulators and components are also applications for Al_2O_3 based on high electrical resistance. With good optical transparency, Al_2O_3 is used as transparent substrates for many optical parts. With the additions of other materials such as graphite, even higher temperatures, harsh environments, and severe applications are envisaged, such as pouring spouts and sliding gate valves at high temperature. For laser host applications, by using ceramic materials, such as Al_2O_3, significantly higher temperatures are potentially attainable.

Pure Al_2O_3 is colorless, while the addition of transition metal ions to Al_2O_3 creates spectacular colors, gem stones, and practical applications such as ruby lasers. Cr_2O_3-doped Al_2O_3, known as ruby, shows a beautiful red color with about 0.1 mol% of Cr_2O_3 [10]. The red color of ruby results from the transitions of electrons between energy levels in its crystal structure. Doping other transition metal ions into Al_2O_3 crystals may produce other colors within the range of visible light. For example, the deep blue color is resulted by the addition of a few hundred ppm of Fe^{2+} and Ti^{4+} impurities [11]. A wide variety of other colors are found in natural and synthetic Al_2O_3 crystals. Fig. 3 shows the various colors of transparent Al_2O_3 and $MgAl_2O_4$ ceramics, fabricated by adding an amount of transition metal ions and sintering by PECS.

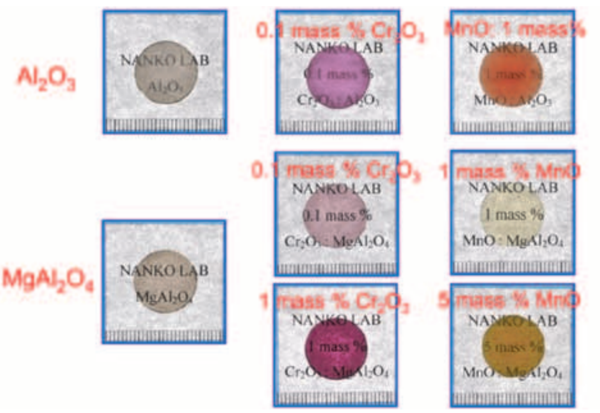

Fig. 3: Appearance of various transparent Al_2O_3 and $MgAl_2O_4$ added with different dopants. Reprinted with permission. Copyright: © 2015 Nanko M, Dang KQ. Published in [Pulsed electric current sintering of transparent alumina ceramics, in "Sintering techniques of materials", InTech] under CC BY 3.0 license. Available from: http://dx.doi.org/10.5772/59170.

In preparation of transparent polycrystalline Al_2O_3, the two most important factors are density and grain size [3, 12-40]. Due to the negative influences of pores on the optical transparency, transparent polycrystalline materials requires extremely low porosity (<0.01 vol%) [18, 20, 21, 37]. It is also believed that polycrystalline materials with nano-scaled grain size would provide better transparency than materials with a grain size in the micrometric range. Moreover, the nano-scaled grain size of the polycrystalline matrix also accomodates with significant improvement in the mechanical properties. Traditional transparent polycrystalline Al_2O_3 are prepared by sintering in hydrogen gas at temperature generally above 1700°C [13, 41]. That high sintering temperature produces polycrystalline Al_2O_3 with low porosity but causes extensive grain growth, which dramatically reduces the mechanical strength, hardness and optical transparency of the material. As a result, the optical in-line transmission of polycrystalline Al_2O_3 produced by conventional sintering is typically below 10%, and the material appears translucent rather than transparent [14]. This matter obviously indicates that conventional sintering is not appropriate to produce transparent polycrystalline Al_2O_3.

Currently, fine-grained transparent Al_2O_3 have been successfully prepared by HP, HIP or PECS at a low temperature range, from 1150 to 1400°C [15, 18-20]. By HP or HIP techniques, the sample is heated by the heat transfer from the external surface of the container to the powder compacts. The consequent heating rate is then typically slow, as lower as 10°C/min and the process normally lasts for hours. The long process at high temperature causes serious grain growth, which is unfavorable for fabrication of transparent Al_2O_3. The large grain size of Al_2O_3 bulks leads to the reduction of the optical inline transmittance and the mechanical properties, such as hardness, fracture toughness or abrasion resistance [21, 22, 26]. The common strategy for sintering of transparent polycrystalline Al_2O_3 is to improve the densification without significant grain growth. The fine-grained microstructure (< 1 μm) provides polycrystalline Al_2O_3 with a significant improvement in both mechanical strength and optical transparency. It is reported that bending strength of the fine-grained transparent Al_2O_3 is up to 400-600 MPa with a high in-line transmission up to 60% for visible light [14, 26]. The addition of MgO is well-known to annihilate normal and abnormal grain growth during sintering of polycrystalline Al_2O_3 [16, 25]. As the result, polycrystalline Al_2O_3 has finer microstructure in grain size with higher final density.

Recently, PECS has become an alternative method to fabricate transparent polycrystalline Al_2O_3 [3, 15-20, 37-41]. It is reported by Kim et al. [19] and Dang et al. [38, 39] that a slow heating rate, such as 2°C/min, was the critical PECS parameter for densification of transparent Al_2O_3 prepared by PECS. Grasso et al. reported that a high pressure of 500 MPa promoted densified and transparent Al_2O_3 in PECS at low temperature of 1000°C [3]. Langer et al. reported about comparison between HP and PECS processing for the Al_2O_3 powder (TM-DAR). By fixing the sample geometry, the heating program, the applied pressure and the atmosphere for both sintering processes, at a given constant time, PECSed samples achieved higher relative density than HPed ones [42].

In consideration of the most impressive advantage of PECS as the high heating rate, Nanko et al. established a new technique with PECS to prepare polycrystalline Al_2O_3 with high transparency: two-step PECS (TS-PECS) [37]. Fig. 4 shows the comparison of some common temperature programs of the PECS processes. A normal one-step PECS process with high heating rate, indicated as the line no. 1, was used commonly but resulted in an opaque or translucent polycrystalline Al_2O_3 samples. To obtain transparent polycrystalline Al_2O_3, the one-step PECS program, which is indicated as line no. 2, employed a much slower heating rate, such as 2°C/min, leading to high density with small grain size [19]. TS-PECS was established with two stages of temperature holding.

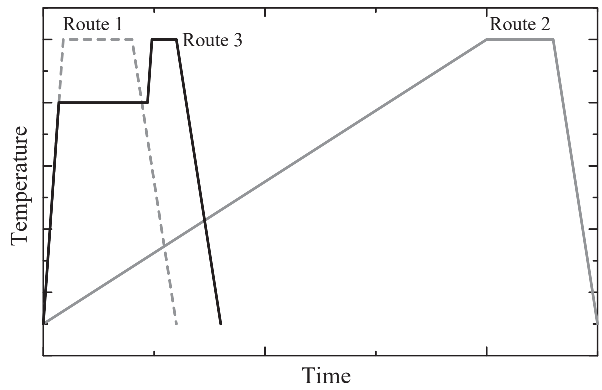

Fig. 4: PECS temperature programs: (1) Normal PECS with high heating rate, (2) Normal PECS with low heating rate, (3) TS-PECS with high heating rate.

Instead of slowly heating up to the final sintering temperature, a holding stage with lower temperature was applied for partial densification without serious grain growth. After that, the temperature was risen to the final sintering temperature to complete densification with preserving the grain size. In both two steps of heating up, the heating rate was 100°C/min [37]. By using TS-PECS, polycrystalline Al_2O_3 with comparatively high transparency was fabricated in a shorter total time than by using a normal PECS process with low heating rate.

Otherwise, Grasso et al. used PECS with two-step pressure pattern to improve the transparency of polycrystalline Al_2O_3 [43]. Fig. 5 shows their Al_2O_3 samples densified with two PECS pattern: constant pressure and two-step pressure. Based on the consideration that the heterogeneous densification was

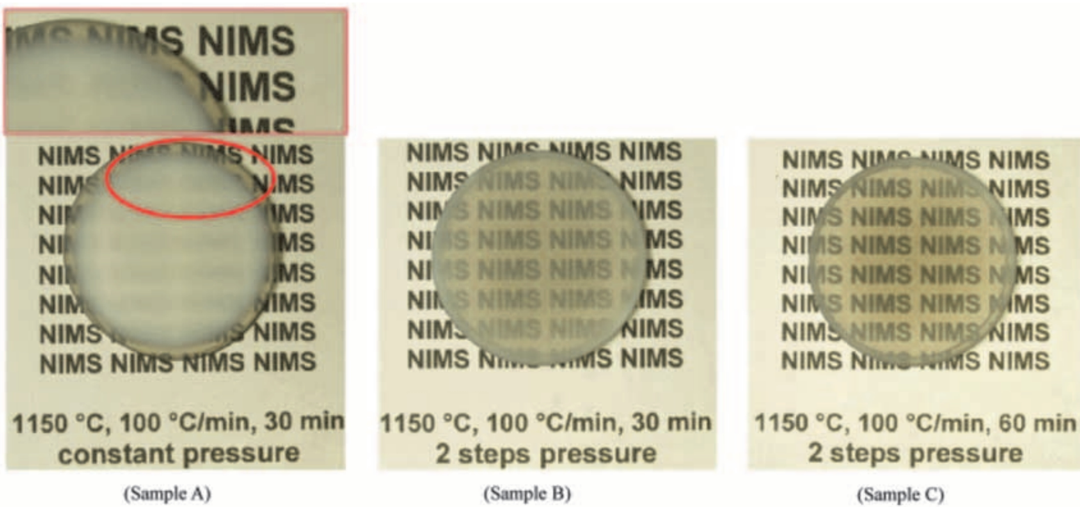

Fig. 5: Photograph of alumina ceramics disks sintered by SPS at 11501C with a heating rate of 100 C/min. The sample shown in figure (a) was sintered for 30 min under 80 MPa constant pressure. The sample (b) and (c) were sintered with pressure two steps pressure application for 30 and 60 min. The samples are 3 cm in diameter and are on top of the text. Reprinted with permission from J. Am. Ceram. Soc., 94 [5] (2011), 1405–1409. Copyright: @ John Wiley and Sons.

the reason for the heterogeneous transparency, they applied low pressure (35 MPa) before the dwelling stage to slow down the densification of the sample's border at the beginning, consequently leading to a more uniform densification at the final stage with higher pressure (80 MPa).

As transparent Al_2O_3 has been a remarkable material in industries, there are a huge amount of publications on transparent polycrystalline Al_2O_3 fabricated by PECS. However, almost of the polycrystalline Al_2O_3 with high transparency fabricated by PECS were sintered at small and thin size, such as 15 mm in diameter or even smaller. The disadvantage of PECS is that there is always a heterogeneity distribution of temperature and pressure inside the powder compact. At small size, this heterogeneity should be not severe but it may be more serious at larger sizes. Among many ceramic materials, transparent polycrystalline Al_2O_3 is a temperature-sensitive material. If the temperature increases just a little, the grain growth may occur seriously and reduce the optical transparency. Because of this characteristic of the PECS process and the strict requirements of transparent polycrystalline Al_2O_3, this material fabricated by PECS is often limited on sintering size. For larger sizes, there have been many reports on the heterogeneity of the transparent polycrystalline Al_2O_3 samples produced by PECS [3, 12, 15, 17-20, 37, 40, 43-45]. Roussel et al. showed their polycrystalline Al_2O_3 samples with a diameter of 20 mm [44]. Their samples were transparent in the center but translucent at the edge and the optical transmittance was improved by using La dopant, as shown in Fig. 6. However, there was no discussion on that heterogeneity of polycrystalline Al_2O_3 samples. Grasso et al. reported a reverse situation about heterogeneity of sintered Al_2O_3 samples with a diameter of 30 mm. The border of sample was denser, had smaller grain size and was transparent while the center was opaque [43]. Wang et al. reported the heterogeneity of grain size of polycrystalline Al_2O_3 samples with various diameters from 12 to 50 mm [46]. Regardless of the differences in grain size at different positions along the radial axis of the samples, the grain size at corresponding positions between various samples was totally different, even the sintering conditions were the same for all of Al_2O_3 samples with various diameters. This heterogeneity of the polycrystalline Al_2O_3 samples derived from the heterogeneous distribution of the sample temperature and pressure during PECS processes. The difficulty of controlling the distribution of temperature and pressure during sintering processes by PECS causes the difficulty of sintering large-sized specimens. Some studies on simulation of PECS processes by finite element method (FEM) reported the heterogeneous distribution of the temperature during PECS processes [46-48]. In almost cases, the distribution of temperature was reported more heterogeneously with the larger sintering size. Therefore, sintering of large-sized transparent polycrystalline Al_2O_3 by PECS is still a challenge.

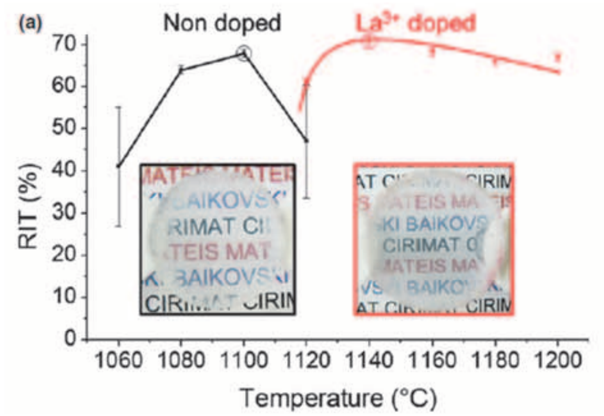

Fig. 6: Dependence of sintering temperature on the real inline transmittance of nondoped and La^{3+}-doped sintered α-Al_2O_3, showing a larger range of densification temperatures with La^{3+} dopant. Reprinted with permission from J. Am. Ceram. Soc., 96 [4] (2013), 1039–1042. Copyright: @John Wiley and Sons.

In our studies of TS-PECS, the transparent polycrystalline Al_2O_3 samples with a diameter of 30 mm were successfully fabricated by the combination of two strategies: the optimization of sintering parameters and the usage of dopants. The sintering temperature of TS-PECS program was 930°C for the first soaking step and 1130°C for the second one. In order to decrease the heterogeneity of grain size of polycrystalline Al_2O_3 samples, the slow heating rate, such as 10°C/min, was applied.

The commercial high-purity Al_2O_3 powder (Taimei Chemicals, TM-DAR) was sintered by TS-PECS with three temperature patterns at the same heating rate of 100°C/min. The heterogeneity of grain size of those three samples was shown in Fig. 7a. The Al_2O_3 sample sintered at 900-1100°C had the smallest and most homogeneous grain size but low relative density, at 99.6%. Due to its low transparency, that sample was not further investigated. On the other hand, Al_2O_3 sample sintered at 950-1150°C obtained higher density, at 99.8%, but was opaque because of the large grain size.

Therefore, the sintering temperature pattern of 930-1130°C was chosen to conduct further investigation of the heating rate. In Fig. 7b, the heating rate was reduced from 100 to 40 and 10°C/min and the relative density of those three samples were 99.8, 99.9 and 99.9%, correspondingly. With decreasing of the heating rate, the grain size of polycrystalline Al_2O_3 sintered by TS-PECS method became smaller and more homogeneous. The apparent optical transmittance of those three samples is shown in Fig. 8, where the optical transmittance was highest with the lowest heating rate, at 10°C/min. However, the optical transmittance of that sample reduced at the edge of the sample. The appearance of the three samples also showed the consistence with the results of optical transmittance. The grain size at the center was larger than near the edge, which typically led to a lower optical transmittance at the center of the sample. In this result, the lower optical transmittance at the edge of the sample was supposed to indicate a heterogeneity of density or porosity of the sample. Unfortunately, the relative density of the samples was almost 99.9-100% and the common Archimedes's method is not accurate enough to detect the difference in relative density at the level of 0.01%. The reasons of the heterogeneous transparency in this case were still unknown and need further studying in the future.

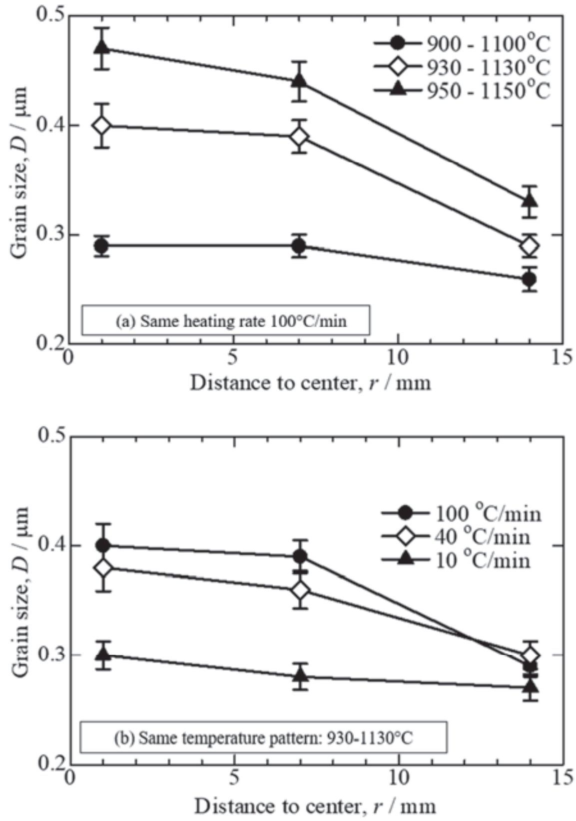

Fig. 7: Distribution along the radial axis of grain size of polycrystalline Al_2O_3 samples with different sintering parameters.

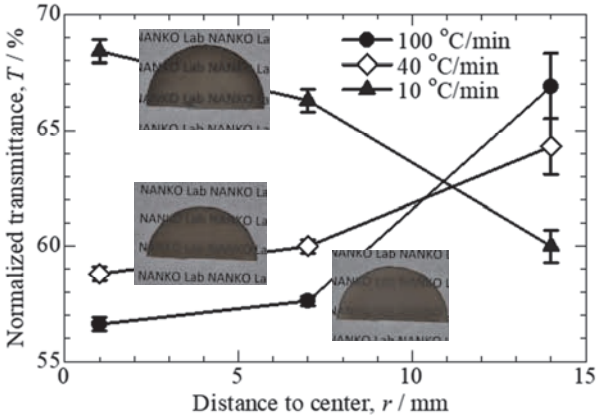

Fig. 8: Appearant optical transmittance of polycrystalline Al_2O_3 samples sintered by TS-PECS with different heating rate.

With the optimization of the temperature and heating rate of TS-PECS program, the large-sized transparent polycrystalline Al_2O_3 was fabricated with good transparency and significantly homogeneous grain size. However, there was still a heterogeneity of the optical transmittance along the radial axis of the transparent bulk samples.

The other routes to control the fine and homogeneous microstructure of the polycrystalline Al_2O_3 are using dopants. Some studies reported that the usage of dopants could slow down the grain growth behavior of the polycrystalline Al_2O_3 during sintering process, so that the fine microstructure could be preserved after sintering [17, 44, 47, 49-53]. One of the most common dopants to reduce grain growth rate of polycrystalline Al_2O_3 was MgO. The influences of MgO dopant in controlling microstructure of polycrystalline Al_2O_3 were reported by Wang et al. [47] and Stuer et al. [17]. Besides MgO, Stuer also used Y and La as the dopants and they reported that the triple doping of Mg-Y-La gave the best results in decreasing the grain size and consequently increasing the optical transmittance of polycrystalline Al_2O_3. Voytovych et al. reported that yttrium doping inhibited both densification and coarsening of sintered α-Al_2O_3 at 1450°C [52]. The influences in densification behavior were suggested relating to the transition from grain boundary diffusion to lattice diffusion controlled densification with increasing temperature. Bojarski et al. also reported a similar influence of Y and La co-doping on the grain growth behavior of Al_2O_3 [51]. Yoshida et al. investigated the densification of Ti-doped Al_2O_3 and showed that the densification and grain growth of Al_2O_3 sintered in N_2+H_2 gas atmosphere was depressed by Ti dopant [54].

Table 2: Sintering temperature of TS-PECS of undoped and doped polycrystalline Al_2O_3 samples, and relative density of sintered bodies.

Dopant	Sintering temperature 1st – 2nd step	Annotation	Relative density	Notes
-	930-1130°C	Undoped	99.9%	Transparent - White
TiO_2 (0.1 mol%)	930-1130°C	Ti1130	98.1%	Opaque
	1000-1200°C	Ti1200	99.9%	Transparent - Dark
Y_2O_3 (0.1 mol%)	930-1130°C	Y1130	80.6%	Opaque
	1000-1200°C	Y1200	92.8%	Opaque
	1100-1300°C	Y1300	99.8%	Translucent
	1150-1350°C	Y1350	99.9%	Transparent - White

Trunec et al. also successfully reduced the grain size as well as improved the optical transparency of polycrystalline Al_2O_3 by using zirconia-spinel co-doping with a combination of PECS and HIP [49].

In our own study, the microstructure of polycrystalline Al_2O_3 was controlled by using two dopants, TiO_2 and Y_2O_3. With both dopants, the densification and grain growth of polycrystalline Al_2O_3 were depressed. However, the influences of Y_2O_3 dopant was much stronger than TiO_2. The influences of Ti and Y dopants in densification of polycrystalline Al_2O_3 can be seen obviously in Table 2. While the undoped Al_2O_3 was fully densified at the sintering temperature of 930-1130°C for TS-PECS, the Ti-doped and Y-doped ones required 1000-1200°C and 1150-1350°C, respectively, for fully densification.

Fig. 9 shows the distribution of grain size of transparent samples along the radial axis. Even at much higher sintering temperature, 1150 and 1350°C for the two steps, the grain size of Y_2O_3-doped Al_2O_3 sample was still equivalent with that of the undoped ones. The grain size of TiO_2-doped Al_2O_3 was a bit smaller than the undoped one, although the sintering temperature was a bit higher. Both results indicated that the dopants dramatically depressed the grain growth of polycrystalline Al_2O_3. Fig. 10 shows the appearent optical transmittance of the three transparent samples, measured at different positions along the radial axis. Although the detailed understanding of the heterogeneity of optical transmittance has been still not clear yet, the results showed obviously that the dopant of Y_2O_3 could improve optical transparency of polycrystalline Al_2O_3 as well as its homogeneity. It indicated that the additive of Y_2O_3 dopant is excellent to control the microstructure of polycrystalline Al_2O_3. Although TiO_2 dopant could depress the grain growth of polycrystalline Al_2O_3 and improve the homogeneity of grain size, it caused a dark color for polycrystalline Al_2O_3 samples, which led to a significantly lower optical transmittance. Therefore, the TiO_2 dopant is not good for transparent polycrystalline Al_2O_3.

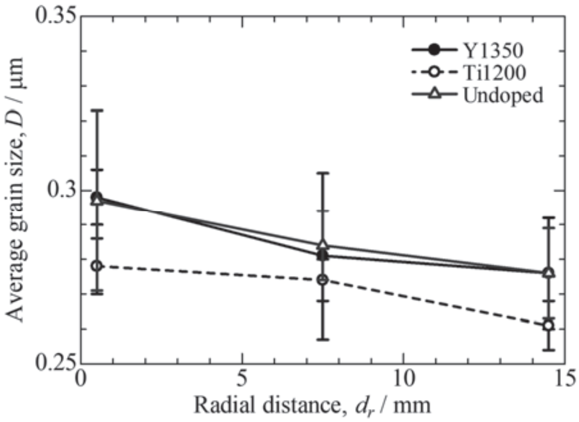

Fig. 9: Distribution along the radial axis of grain size of polycrystalline Al_2O_3 samples with different dopants.

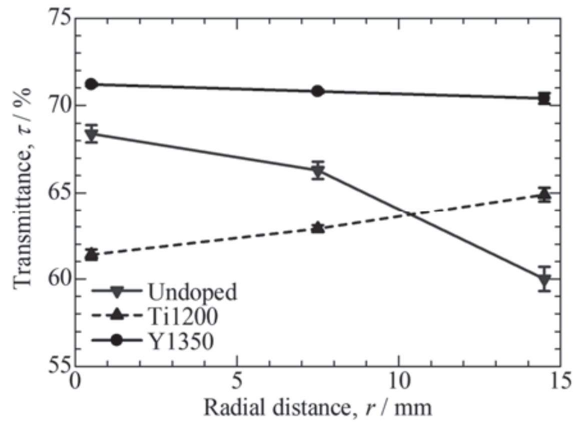

Fig. 10: Appearant optical transmittance of polycrystalline Al_2O_3 samples sintered by TS-PECS with different dopants and corresponding sintering temperature.

In summary, PECS has been used widely to fabricate transparent polycrystalline Al_2O_3. However, the optical transparency of polycrystalline Al_2O_3 is still not as good as single crystal sapphire, which has been applied in many industries for many years. The necessary improvements for transparent polycrystalline Al_2O_3 includes the optical transparency, the homogeneity of the sintered bodies as well as the sintering size. Along with the developments of PECS and its related sintering methods, polycrystalline Al_2O_3 is expected to substitute single crystal one in variety of industrial applications.

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