Can Ultra-fast High Temperature Sintering (UHS) Boost Transparency of Alumina?

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Abstract

Established routes for consolidation of transparent alumina ceramics by pressure-less sintering requires several hours of dwelling in a reducing atmosphere at a temperature exceeding 1600 °C. Here, for the first time, we report on low temperature and ultrafast consolidation of translucent alumina ceramics.

Transparency was promoted by the synergistic of high initial green density (62.7 %) and rapid sintering using Ultra-fast High Temperature Sintering (UHS) technique. The proposed approach, using a heating rate of 430 °C/min and dwelling time of 15 minutes, resulted in ultra-fine-grained translucent alumina ceramics at 1359 ± 57 °C with a grain size of 0.39 μm , and an in-line transmittance of 28.7 % at a wavelength of 700 nm. For comparison, conventionally fired counterparts were opaque due to their incomplete densification, pore coalescence.

1. Introduction

Approximately 83 % of processing energy is consumed during the firing step and modern ceramic processing is pushing towards reduction of firing energy [1]. In this regard, research on development of new techniques, which can reduce the required energy for firing of ceramics by lowering either temperature or dwelling time has become of paramount importance. Apart from solving environmental and economic concerns, low energy firing brings additional benefits in terms of suppressed grain growth [2, 3], improved mechanical properties and optical properties [4, 5].

Firing using a high heating rate allows to considerably reduce processing time. High-heating-rate firing can also be efficient in sintering/ densification of fine-grained ceramics whose activation energy of densification is higher than that for coarsening [6, 7]. Different sintering techniques, including Fast Firing (FF) [6], Microwave Sintering (MWS) [8], Spark Plasma Sintering (SPS) [9], Flash Sintering (FS) [10], and Ultra-fast High-temperature Sintering (UHS) [11] offer high heating rates in the range of $10^2 - 10^4$ °C/min [12–14]. Among these sintering techniques, FF [15], MWS [16], and SPS [17, 18] have been employed to sinter translucent/ transparent ceramics. For instance, transparent MgO-doped alumina ceramics with a real in-line transmittance of 64 % at a wavelength of 1100 nm have been sintered by FF under vacuum at 1670 °C for 5 min [15]. MWS also allowed to sinter translucent MgO-doped alumina with an average grain size of 20–40 μm and in-line transmittance of 15–35 % (between 500 to 1000 nm) at 1750°C for 15–45 min in an ultrahigh pure hydrogen atmosphere [16]. SPS, which takes advantage from simultaneous application of a high heating rate ($> 10^2$ °C/min) along with a mechanical pressure, is also one of the most widely employed sintering technique for sintering of transparent alumina ceramics [19–21]. For instance, transparent AlF_3 -doped alumina ceramics with a grain size of 1.55 μm and with an in-line transmittance of 30–40 % at the wavelength of 600 nm were sintered at 1350°C under a pressure of 73 MPa after 5 min [20], while by application of pressures as high as 500 MPa for 10 min at 1000°C highly transparent alumina ceramics with an average grain size of 0.2 μm and in-line transmittance of 64 % at 645 nm have been sintered [19].

Ultra-fast High-temperature Sintering (UHS), developed by Wang et al. [11], is a pressure-less sintering technique, which has enabled synthesis and sintering of a wide range of materials (metallic or intermetallic compounds [22], oxide [2, 3, 23–25] (including solid state electrolytes [11, 26–28]) and non-oxide [29] ceramics, for instance) in few seconds to minutes. In a typical UHS process, a green compact is sandwiched between carbon felt(s). Joule heating of carbon felt provides sufficient heat for synthesis/sintering. In addition to a high heating rate ($10^3 - 10^4$ °C/min), and high temperatures (up to 3000 °C), a high cooling rate ($10^3 - 10^4$ °C/min) can also be achieved during UHS [11, 22].

Although translucent/ transparent alumina ceramics have been produced by pressure-less sintering [15, 16, 30], up to now there has been no report on low temperature (less than 1400°C) sintering of ultra-fine-grained translucent/ transparent alumina ceramics using a pressure-less sintering technique without a need for hydrogen atmosphere or vacuum. In this context, in a previous study [31], we performed a parallel comparison between sinterability of alumina ceramics prepared by Dry cold isostatic Pressing (DP) and Water Assisted cold isostatic Pressing (WAP). It was demonstrated that the WAP approach allowed to prepare green bodies with a higher relative density than the traditionally employed DP approach (62.48 % vs. 57.86 % for WAP and DP [31], respectively). WAP compacts showed an enhanced sinterability/ densification and obtained a relative density of 99.32 % after Conventional Sintering (CS) at 1350 °C for 15 min with small porosities remained (< 0.26 μm) in the microstructure, featuring a weak translucent appearance. By contrast, under the same processing conditions, DP artifacts only densified up to 97.75 %, while porosities as large as 0.63 μm were still present in the microstructure of the specimen, resulting in an opaque appearance [31]. In this work we combined synergistic effects of WAP with UHS technique. Targets were to fabricate translucent/ transparent alumina ceramics by maximizing densification while limiting grain growth.

2. Experimental Procedures

α - Al_2O_3 powder (Super-TM-DAR, Taimei Chemicals Co. Ltd., Japan, purity: 99.99 %, average particle size: 130 nm, BET: $17.5 \text{ m}^2/\text{g}$) mixed with 0.1 wt% MgO (Shanghai Aladdin Biochemical Technology, purity: 99.9 %, average particle size: 50 nm) in ethanol using a magnetic stirrer to avoid possible introduction of impurities from milling media. The mixture was ultrasonicated for 1 h after mixing process. Afterwards, the mixture was dried for 24 h at 80°C in an oven, and then passed through an 80-mesh sieve. The sieved powder mixed with 17 vol% distilled water using a vortex mixer for 30 min, and then uniaxially pressed at 50 MPa to prepare specimens with a diameter of 13 mm and thickness of 3 mm. Uniaxially pressed pellets were then CIPed at 600 MPa for 15 min. Finally, green compacts were dried in an oven for 24 h at 120 °C. Samples had an initial green density of 62.73 ± 0.24 % (averaged over 10 samples).

UHS was adopted to sinter prepared green specimens. Samples were either or not presintered at 800°C for 3 h (heating rate: 10°C/min). Specimens were placed between two pieces of carbon felt (Sigratherm® GFA) with a size of $30 \times 5 \times 80$ mm. All experiments performed in an argon filled box. The electrical power was supplied by a programmable power supply (0–30 V, 0-100 A, DM Series, Shanghai Soy power Co., Ltd., China). The electric current increased in a stepwise profile to avoid cracking, as previously

reported in Ref. [2]. Samples prepared in our previous study [2] had a grayish color, so in this study alumina fiber boards were not employed in the UHS set-up. In a separate sets of experiments, a dummy sample with the same size and weight similar to that of translucent UHSed specimens was used to measure temperature by means of an infrared dual color pyrometer (Dikai, DIT613) which was directly focused on the sample. Temperature measurement was repeated at least for 5 times. Consequently, reported values are average of 5 measurements. As discussed in the following, carbon felts cannot be employed for a long discharge time, not allowing to perform a sintering practice with a low heating rate. Therefore, a sample with a comparable relative density as that of translucent specimens was sintered by CS using a heating rate of 5 °C/min at 1350 °C for 15 min under Ar atmosphere.

The density of sintered specimens was measured by Archimedes' principle. In case of translucent specimens, the density was measured using an image processing software by considering results from 5 micrographs. Phases present in as-sintered specimens were characterized by Raman spectroscopy (Horiba Scientific LabRAM HR evolution) using an Ar laser ($\lambda = 514$ nm). Translucent specimens were polished up to a thickness of 1 mm and further mirror polished using a diamond paste. In-line transmittance was measured by a UV-VIS-NIR spectrophotometer (Cary 5000; Varian Inc., Lake Forest, CA) for the wavelength from 200 to 800 nm. Bright Field Transmission Electron Microscopy (TEM, ZEISS Libra 200 FE) was done on as-sintered specimens. Specimens for TEM studies prepared by an ion beam milling system (EM RES102, Leica). Samples were thermally etched at 1150°C to examine the microstructure by a Field Emission Scanning Electron Microscopy (FESEM, S-8220; Hitachi, Japan). The apparent grain size measured by obtaining average cross section area of each grain, assuming spherical grains. To determine the true grain size, the measured apparent grain size was multiplied by 1.225 [5]. In the case of ultra-fine-grained specimens ($< 0.5 \mu\text{m}$), grain size was measured using TEM micrographs as thermal etching could have caused grain growth.

3. Results And Discussion

Table 1 summarizes processing conditions and resulting properties of UHSed and CSed specimens. Initial attempts on attaining translucent alumina ceramics were made without incorporation of a presintering step before the final sintering step. UHS 1 (Table 1) was UHSed for 2 min at a maximum current of 29 A, corresponding to a peak value of 258 W for electric power (Fig. 1 (a)). However, as substantiated by Raman spectra (Fig. 1 (b)), presence of carbon contaminations affected physical appearance of UHS 1. Carbon contaminations resulted from degradation of carbon felts during UHS process. Figure 2 compares changes in microstructural features of the carbon felt before and after a 2 min discharge at 29 A. As can be seen, even though the discharge time was as short as 2 min, the initially smooth surface of carbon threads changed to a slightly rough surface, suggesting degradation of carbon felt. With further extension of discharge time, carbon felt experiences a considerable degradation, affecting the electrical conductivity of the heater. As an example, the discharge time at the maximum electric current of 29 A raised to 30 min, and results are represented in Fig. S1.

As compared in Fig. 1 (b), Raman spectra of UHS 1 and UHS 2 (Table 1) suggests that upon integration of a presintering step (800°C for 3 h (5°C/min)) in processing conditions of UHS 2, the specimen was not further affected by carbon contaminations after being UHSed for 2 min at the maximum current of 29 A (Fig. 1 (a)). Therefore, to avoid carbon contaminations from carbon felts during UHS process, an additional presintering step (800 °C, 3 h (5°C/min)) was necessary. It is likely that presintering improved the thermal conductivity of the sintering sample and it also enhanced the heat transfer with the carbon felt.

Table 1
Processing parameters, physical and microstructural characteristics of UHSed specimens.

Sample ID	Presintered (P)/ Not Presintered (NP)	Electric current (A), and peak value of electric power (W)	Temperature (°C)	Dwell time at maximum electric current (min)	Physical Appearance, and in-line transmittance (%)	RD (%)	Grain Size (µm)*
UHS 1	NP	29, 258	—	2	Opaque, black	68.14 ± 0.62	—
UHS 2	P	29, 258	—	2	Opaque, white	67 ± 0.54	—
UHS 3	P	33, 317	1290 ± 54	2	Opaque, white	84.17 ± 0.65	0.15 ± 0.03
UHS 4	P	33, 321	1312 ± 55	5	Opaque, white	94.56 ± 0.51	—
UHS 5	P	33, 334	1359 ± 57	15	Translucent, 28.7	99.92 ± 0.01	0.39 ± 0.05
UHS 6	P	33, 348	1454 ± 61	30	Translucent, 20.1	99.91 ± 0.01	0.78 ± 0.06
UHS 7	P	33, 385	1517 ± 64	45	Translucent, 5.2	99.94 ± 0.02	1.49 ± 0.05
CS 1**	P	-	1350	—	Weakly translucent	99.68 ± 0.12	0.66 ± 0.04

* Grain sizes of UHS 3 and UHS 5 were measured using TEM micrographs.

** Fifteen minutes dwell time at 1350 °C. Sample prepared using WAP. Dry Pressed specimens were opaque [31].

As UHS 2 did not experience a considerable densification ($67 \pm 0.54\%$, Table 1) by applying an electric current of 29 A for 2 min, the electric current further increased to 33 A. Consequently, by ramping the electric current within 60 s followed by a dwell time of 2 min at a maximum electric current of 33 A, corresponding to peak value of 317 W and 1290 ± 54 °C for electric power and temperature, respectively (Fig. 3 (a) and (b)), UHS 3 densified up to 84.17 ± 0.65 % (Table 1). Subsequently, UHS 4 (Table 1) experienced an effective densification (94.56 ± 0.51 %) after being UHSed for 5 min at the maximum electric current of 33 A (peak value of electric power: 321 W), corresponding to a temperature of 1312 ± 55 °C (Fig. 3 (a) and (b)). Making a comparison between processing conditions and relative densities of UHS 3 and UHS 4 (Table 1) suggests a rise in the dwell time at maximum electric current resulted in an effective densification, while temperature did not rise significantly (Fig. 3 (b)).

Figure 4 and Fig. S2 show TEM images of UHS 3. It should be emphasized that necks can clearly be distinguished. This implies that a high surface energy and/ or surface area was preserved up to temperatures as high as 1290 ± 54 °C. Some of developed grain boundaries for UHS 3 featured a curved/ non-equilibrium shape. Development of curved/ non-equilibrium grain boundaries during ultra-fast sintering of alumina with a heating rate of $\cong 1650$ °C/min was also reported by Zhang et al. [32]. Such curved/ non equilibrium grain boundaries feature a diffuse, an open structure, a high in-plane diffusion coefficient and a high diffusion cross section [32]. It is interesting to note presence of Moiré fringes (Fig. 4 (d)), resulted from superposition of two Al_2O_3 lattices. Considering the initial particle size (130 nm) and grain size of UHS 3 (0.15 ± 0.03 μm , Table 1), it can be pointed that UHS 3 almost experienced a minimal grain growth. Such analysis justifies the absence of grain coarsening experienced by UHS 3.

Recalling established literature on FF of alumina (Al_2O_3) [6], the activation energy for surface diffusion (230–280 kJ/mol) is less than that of the densification through lattice diffusion (580 kJ/mol), a high heating rate allows to rapidly bypass the low temperature window where coarsening is dominant. These effects were investigated in our previous work (see Fig. 5 in ref [2]). Coarsening mainly proceeds through low activation energy mechanisms like surface diffusion, evaporation/ condensation at low temperatures, while densification requires grain boundary and/or lattice diffusion [7]. The suppressed grain growth during UHS processing is likely to enhance grain boundary diffusion, as a reference oxygen self-diffusion coefficient in polycrystalline alumina at 1500 °C is (at least) more than two orders of magnitude higher when compared to single crystal [33].

The preserved high surface energy/ area, and curved/ non-equilibrium grain boundaries developed as a result of the ultra-fast heating rate during UHS process, favor an enhanced densification, allowing a rapid densification within a 3 min from UHS 3 to UHS 4 without the need for a noticeable rise in temperature. The heat modulation based on a heating rate of $\cong 430$ °C/min followed by $\cong 5$ °C/min (Fig. 3 (b)) was a deliberate attempt to minimize grain growth (UHS 3) and maximize densification for translucent specimens (UHS 5). The dwell time at the maximum electric current of 33 A further prolonged. Sintering of UHS 5 (Table 1) with a dwell time of 15 min at the maximum electric current of 33 A (peak value of electric power: 334 W) and a temperature of 1359 ± 57 °C (Fig. 3 (b)) resulted in translucent alumina ceramics with an average grain size of 0.39 ± 0.05 μm , as shown in Fig. 5 (a). TEM images of UHS 5

(Fig. 6 (a-d) and Fig. S3) confirms limited grain growth of the ultra-fine-grained translucent UHS 5. During the transition from a partially to nearly fully dense microstructure (UHS 3 to UHS 5), most of the curved/non-equilibrium grain boundaries evolved into flat or smoothly curved grain boundaries with dihedral angles close to equilibrium (Fig. 6 (d)), similar to that is of conventional sintering [30, 34]. Non-equilibrium features were not observed, contrary to a previous study by Luo et al. [29] on UHS of Si_3N_4 , involving the presence of a meta-stable liquid phase. The bimodal particle size distribution, with very fine grains (Fig. 6 (b-d), between 100–200 nm) and large grains (Fig. 6 (a and b), $\cong 500$ nm) reflects the dynamic evolution of grain growth induced by the ultra-rapid UHS process.

A photograph of translucent specimens is shown in Fig. 7. The in-line transmittance of UHS 5 for a wavelength of 700 nm was 28.7 % (Fig. 8). With a rise in dwell time from 15 min to 30 min for UHS 6 (Table 1), and subsequent increase in electric power (peak value of electric power: 348 W, Fig. 3 (a)) and temperature (1454 ± 61 °C, Fig. 3 (b)), the grain size increased to 0.78 ± 0.06 μm (Fig. 5 (b)) and in-line transmittance decreased to 20.1 % (Fig. 8). Finally, UHS 7 (Table 1) after being sintered for 45 min at the maximum electric current of 33 A (corresponding to a value of 385 W and 1517 ± 64 °C for the electric power and temperature, respectively (Fig. 3)), obtained the largest grain size (1.49 ± 0.05 μm , Fig. 5 (c)) and the lowest in-line transmittance of 5.2 % at 700 nm (Fig. 8). The high in-line transmittance of UHS 5, compared to UHS 6 and UHS 7, is attributed to the small grain size, since at a small grain size, reflectivity and light refraction are negligible [5].

To substantiate the pivotal role of ultra-fast heating rate in sintering of translucent alumina ceramics, a comparable specimen was sintered by CS. Figure 5 (d) shows microstructure of the CSed specimen (CS 1 (Table 1)). It can be seen that the CS 1 well densified up to a relative density of 99.68 ± 0.12 % with a grain size of 0.66 ± 0.04 μm . However, porosities were still present in the microstructure of the sample. In this regard, Kim and Hee Kim [35] compared evolution of pore shrinkage using a fast and a low heating rate and demonstrated that as result of rapidly passing through low temperatures, porosities only shrank, and pore coalescence was effectively suppressed by the high heating rate of 500 °C/min. On the contrary, during sintering with a low heating rate (10 °C/min) sufficient time is provided for porosities to coalesce together [35]. Therefore, it can be speculated that sintering under an ultra-fast heating can effectively prevent pore coalescence. Large pores exceeding a few hundred nanometer are detrimental for the in-line transmittance.

Ultra-fine grained translucent alumina ceramics were obtained by the synergistic effect of high green density (attained by WAP, which in our previous study [31] was demonstrated to enhance sinterability/densification of alumina ceramics through improving green density) followed by consolidation under an ultra-fast heating rate using UHS technique. The mechanisms behind such a strong effect deserve further clarifications. In literature on FF of alumina ceramics, formation of a liquid phase at grain boundaries has been reported [36]. In addition, it has also been speculated that an ultra-fast heating rate results in dislocation formation which can improve densification by enhancing diffusion rate [37]. However, in case of alumina ceramics prepared in the current study, neither a liquid phase nor dislocations were observed (Fig. 4 and Fig. 6).

Grain sizes of translucent UHS 5 and CS 1 (Table 1), prepared in the current work using Super-TM-DAR (BET: 17.5 m²/g) and other published contributions [38–43] on CS and Two Step Sintering (TSS) of opaque MgO-doped and undoped alumina using TM-DAR powder (BET: 14.5 m²/g), sintered at temperatures ≤ 1500 °C, are compared in Fig. 9. Grain sizes of compacts prepared by WAP and CSed at 1350 °C using Super-TM-DAR and TM-DAR powders [31] are 0.66 ± 0.04 μm and 0.60 ± 0.05 μm , respectively, suggesting no significant difference.

Regardless of transparency, several attempts have been made on sintering of fine grain fully dense alumina ceramics using CS and TSS [38–43]. In this regard, Bodišová et al. [40] sintered undoped dense alumina ceramics with a relative density of 99 % and a grain size of 0.76 μm by CS at 1420°C without dwelling. Razavi Hesabi et al. [38] studied suppression of grain growth of undoped alumina by TSS and reported that samples with a grain size of 0.5 μm can be sintered by heating to 1250°C (for the first step) and then holding at 1150°C (for the second step); however, dwell time as long as 64 h for the second step was necessary to achieve a relative density of 98 %. In case of MgO doped alumina ceramics, fully dense specimens (99.4 %) with a grain size of 0.81 μm have been obtained by CS at 1500°C without dwell time, while samples with a relative density of 99.3 % and a grain size of 0.8 μm have been sintered by TSS after being heated up to 1400°C and being held for 8 h at 1275°C [40].

The proposed comparison in Fig. 9 suggests that by exploiting the synergistic effect introduced in the current study (high initial green density and sintering with an ultra-fast heating rate), translucent alumina ceramics with smaller grain size (UHS 5, 0.39 ± 0.05 μm) than that obtained by CS and TSS (larger than 0.5 μm) for opaque alumina ceramics [38–43] can be prepared at comparable temperatures (1350 ± 57 °C for UHS and ≤ 1500 °C for CS and TSS). Harmer and Brook [6] proposed that the higher the firing temperature, the higher the ratio of densification to coarsening, provided that the activation energy of grain boundary/ lattice diffusion is higher than that of surface diffusion. Although the proposed discussion still holds true and has widely been acknowledged in studies on FF of alumina ceramics [36, 38], results of the current study suggest that new sintering trajectories can be defined for FF of ultra-fine-grained (translucent/ transparent) alumina ceramics.

A comparison between processing conditions, in-line transmittance and grain size of the translucent UHS 5 (Table 1) and that of previously published literature [15, 16, 44] on translucent/ transparent alumina ceramics, prepared by pressure-less sintering techniques has been provided in Table 2. Both the reduced UHS processing time and lowered temperature are indicative of the potential energy saving of UHS compared to other routes. Attempts on sintering of translucent/ transparent alumina ceramics using pressure-less sintering have mainly been performed at temperatures higher than 1670 °C either in vacuum or in pure hydrogen atmosphere [15, 16, 44]. Zhang et al. [15] reported on fabrication of translucent/ transparent MgO-doped alumina by performing a presintering step at 1200 for 2 h and rapid vacuum sintering at 1670°C for 5 min with an average grain size of 11.4 μm and in-line transmittance of ≈ 54 % at a wavelength of 700 nm. Cheng et al. [16] also reported on sintering of translucent/ transparent MgO-doped alumina ceramics, presintered at 1100 °C for 2 h, and microwave sintered in a highly pure hydrogen atmosphere at 1750°C for 45 min with an average grain size of 40 μm and an in-line

transmittance of $\approx 30\%$ at the wavelength of 700 nm. By presintering at 800 °C for 50 h and employing CS at 1700°C for 3 h in vacuum, translucent MgO-doped alumina ceramics with an average grain size of $\approx 8\ \mu\text{m}$ and an in-line transmittance less than 6 % at a wavelength of 700 nm have been sintered by Kim et al. [44]. By contrast, the proposed approach in the current study suggests that translucent alumina ceramics with an in-line transmittance of 28.7 % for a wavelength of 700 nm and grain size of $0.39 \pm 0.05\ \mu\text{m}$ can be produced by presintering at 800 °C for 3 h and ultra-fast sintering at a temperature of $1359 \pm 57^\circ\text{C}$, which is approximately 300–400 °C less than what have been reported [15, 16, 44]. Furthermore, UHS required Ar atmosphere without a need for hydrogen atmosphere or vacuum.

Table 2

Comparison between processing conditions, in-line transmittance and average grain size of the translucent UHS 5 (Table 1), prepared in the current study, and published literature [15, 16, 44] on translucent/ transparent alumina ceramics.

Ref	Cold to cold time (min/ h)	Sintering temperature (°C) and dwell time (min)*	In-line transmittance at 700 nm (%)	Average Grain size (μm)
Current work (UHS 5, Ar atmosphere)	Less than 20 min	1359 ± 57 , 15	28.71	0.39 ± 0.05
Rapid vacuum sintering [15]	Approximately 5–6 h	1670, 5	≈ 54	11.4
Microwave sintering (hydrogen atmosphere) [16]	Approximately 3–5 h	1750, 45	≈ 30	40
Pressure-less CS (vacuum) [44]	Approximately 10–12 h	1700, 180	≈ 6	8
* Dwell time in the last stage of sintering process.				

4. Conclusions

Ultra-fast High-temperature Sintering greatly simplifies the fabrication of translucent alumina ceramics. Ultra-fine-grained translucent alumina ceramics with a grain size of $0.39 \pm 0.05\ \mu\text{m}$ were obtained under the synergistic effects of improved green compaction, low temperature presintering (to avoid carbon contaminations) and ultra-fast heating using UHS technique. The sintered compact had an in-line transmittance as high as 28.7 % at a wavelength of 700 nm.

Compared to traditional pressure-less sintering techniques for sintering of translucent/ transparent alumina ceramics [15, 16, 44], the UHS facilitates the sintering process by eliminating the need for a reducing atmosphere (i.e., hydrogen), and by shortening the processing times from several hours to a few minutes.

The ultra-fast heating rate has a decisive role in full densification of ultra-fine-grained translucent alumina ceramics, as it favors densification by rapidly passing through low temperatures, storing the high

surface energy and/ or area of the starting powder for high temperatures and developing curved and diffuse grain boundaries. It also avoids pore coalescence experienced during conventional sintering.

Declarations

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Figures

Figure 1

(a) Electric current (A), and electric power (W) profiles of UHS 1 and UHS 2 (Table 1), (b) Raman spectra of corresponding specimens. Raman spectra of carbon felt is also included. Insets in (b) show macroscopic photographs of specimens after final sintering stage by UHS. The presence of carbon contaminations was observed in Raman spectra of not-presintered UHS 1, while no carbon contaminations were detected in UHS 2, presintered at 800 °C for 3 h (5 °C/min).

Figure 2

SEM micrographs of carbon felts, (a and b) before UHS heating, (c and d) after 2 min discharge at 29 A (corresponding to experimental conditions of UHS 1 (Table 1)). Changes in the appearance of fibers from a smooth to a rough surface suggests a degradation during UHS process.

Figure 3

(a) Electric current (A) and electric power (W) profiles for UHS 3 to UHS 7 (Table 1), (b) Temperature profile (°C) and grain size (μm) of corresponding samples. Average heating rate for the first 3 min was $\cong 430$ °C/min, afterwards the heating rate was stabilized to $\cong 5$ °C/min.

Figure 4

Representative TEM images of UHS 3 (Table 1) (more images are provided in Fig. S2) (a-b) the ultra-fast heating rate employed to sinter the specimen(s), allowed to suppress grain growth, (c) neck(s) can be distinguished, implying a high surface energy/ area was preserved up temperatures as high as 1290 ± 54 °C, (d) Moiré fringes.

Figure 5

Representative SEM micrographs of (a) UHS 5, (b) UHS 6, and (c) UHS 7, (d) CS 1, yellow circles represent residual porosities. Low magnification micrographs for UHS 5 and UHS 6 is provided in Fig. S4. Samples' IDs are according to Table 1.

Figure 6

Representative TEM images of translucent UHS 5 (Table 1). Presence of large grains ($\cong 500$ nm, (a and b)) along with fine grains (encompassed by yellow circles (b-d)) indicate the dynamic evolution of grain growth during ultra-rapid UHS process. Some defects were formed during the FIB milling stage, as UHS 5 was a translucent specimen with an in-line transmittance of 28.7 % at 700 nm (Fig. 8). More images are provided in Fig. S3.

Figure 7

Macroscopic photograph of translucent alumina ceramics, from left to right: UHS 5, UHS 6, UHS 7 (Table 1). Samples were rested directly on text (not retro luminated). For comparison samples processed using conventional sintering were opaque. Samples diameter was 10.90 mm.

Figure 8

In-line transmittance (%) of translucent alumina ceramics with thickness of 1 mm. Samples' ID are according to Table 1.

Figure 9

9 Comparison between mean grain sizes (μm) and relative densities (%) of pressure-less sintered alumina ceramics (USH 5 and CS 1 (WAP)) prepared by WAP using Super-TM-DAR (BET: 17.5 m²/g) in the current study, with conventionally sintered specimens by WAP (CS (WAP)) [31] and other literature [38–43] on undoped and MgO-doped alumina ceramics using TM-DAR powder (BET: 14.5 m²/g), sintered at ≤ 1500 °C (CS: Conventional Sintering, TSS: Two Step Sintering, WAP: Water Assisted cold isostatic Pressing)).

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