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Use of Focused High-Power 60-GHz Radiation Beams for Advanced Sintering of Ceramics†

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Abstract

The experimental results of radiation focusing and ceramics sintering using high-power millimeter-wave radiation at 60 GHz are reported. Focusing experiments of millimeter-wave radiation from the high-power pulsed 60-GHz gyrotron have been performed using a quasi-optical antenna system (two-dimensional ellipso-parabolic focusing antenna system) to demonstrate the feasibility of power densities as high as 100 kW/cm². Typical heating characteristics using the focused beam were established for this system. Sintering of yttria-stabilized zirconia (ZrO_2-8mol%Y_2O_3) samples resulted in densification to as high as 97% of theoretical density (TD) using focused 60 GHz beam irradiation in pulse operation with a 10-ms pulse duration at a 0.5Hz repetition. The densification temperature for the zirconia could be lowered by 200°C than that expected conventionally. Sintering of silicon nitride with 3%AlO_3 and 5%Y_2O_3 was also performed using the focused 60-GHz beam in pulse irradiation. It was found that the silicon nitride samples sintered at 1600°C for 30 min reached as high as 90% TD, which conventionally requires a sintering temperature as high as 1700°C.

KEY WORDS: (Millimeter-wave radiation) (Gyrotron) (Quasi-optical antenna) (Ceramics heating) (Sintering)

1. Introduction

Remarkable progress in the microwave sintering of ceramics has been made since the 1980's following the pioneering work by W. H. Sutton[1)], T. T. Meek et al.[2)], and M. A. Janny and H. D. Kimrey[3)]. Since then microwave sintering technology has been widely recognized to offer a number of advantages over conventional sintering process; i.e., rapid and selective heating, higher densification rates at lower temperatures, achievement of sintered bodies with finer grain size and improved mechanical properties.

Microwave frequencies from several GHz to several tens of GHz (millimeter wave region) have been used for ceramics sintering. The most commonly employed frequency has been 2.45 GHz. But at this frequency there are problems associated with inefficiency in direct heating of low loss ceramics and difficulty in designing a multi-mode applicator with sufficient uniformity to avoid thermal runaway, which are mainly attributed to the longer wavelength of the radiation. Hybrid microwave heating[3,4)] has been proposed as one of the solutions for these problems, however, limitations exist in the scale length of the workpiece and the choice of materials with appropriate coupling, and so on. Another possible solution for these problems is to employ millimeter-wave radiation, where the power absorption per unit volume increases linearly with increasing frequency and the sufficient uniformity of the wave field can be obtainable in a feasible and/or practical applicator size. Experiments with millimeter-wave radiations from Gyrotron tubes have been carried out in USA[3)] and followed by a group at the Institute of Applied Physics[5)] in Russia. In Japan our group[6,7)] is also extensively studying this process.

In this paper we present the results of sintering experiments performed with a 100-kW 60-GHz gyrotron in pulse operation. The experiments with the 60GHz pulsed millimeter-wave radiation have been aimed at fine focusing of the radiation using the quasi-optical nature of the millimeter-wave radiation to obtain high power density for the efficient coupling of the wave energy to the samples. This method is convenient for heating the samples in a small reactor and has the potential for rapid heating with actively controlled temperature distribution by appropriately scanning the focused beam over the workpiece. Furthermore

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the focused millimeter-wave beam can be employed in advanced processing of ceramics such as sintering and joining of functionally-gradient ceramics-composite structure, which requires a technology capable of selective heating with precisely controlled temperature gradients (see **Fig.1**).

2. Experimental procedures

The quasi-optical antenna system designed for the fine focusing of the 60GHz millimeter-wave radiation, described elsewhere [6, 8] in detail, is schematically illustrated in **Fig.2**. The millimeter-wave radiation (λ=5mm) in TE_{02} mode was generated by a high-power pulsed Gyrotron tube (VGE8060, Varian) with the maximum output power of 200kW and the maximum pulse width of 100ms. The pulse repetition was 0.5Hz, and the average power of the radiation was limited to about 1kW by the capability of the charging DC power supply to the high-voltage condenser bank.

The 60-GHz millimeter wave was radiated through the cut-in-half circular waveguide of 25.6mm in diameter and reflected by the elliptic mirror to the focal point **O'** on the **O_2** axis. When the parabolic reflector additionally intercepts the radiation, the radiation was again reflected to the focal point F to form a nearly Gaussian profile. The power distributions of the radiation were measured at various x-positions on the y-z plane. The radiation was injected into the microwave absorber sheet (Eccosorb AN, Emerson and

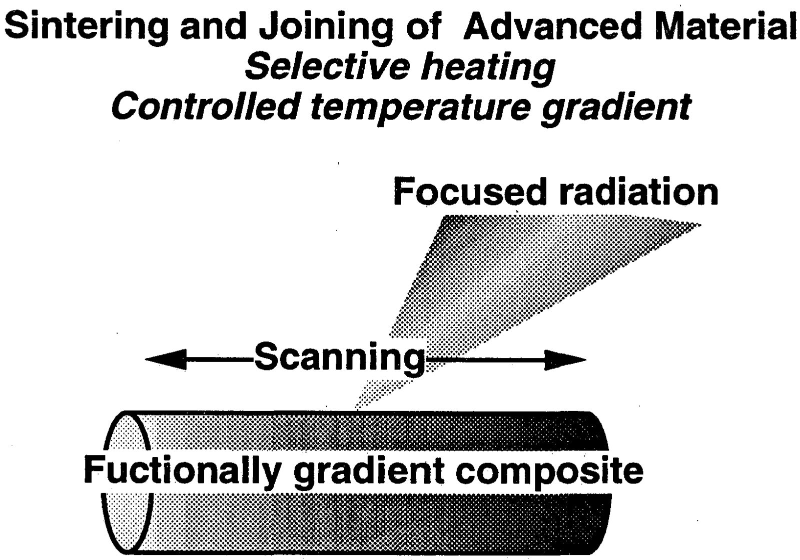


Fig. 1. Focused millimeter-wave radiation can be used for sintering and/or joining of advanced ceramics such as functionally gradient composite materials.

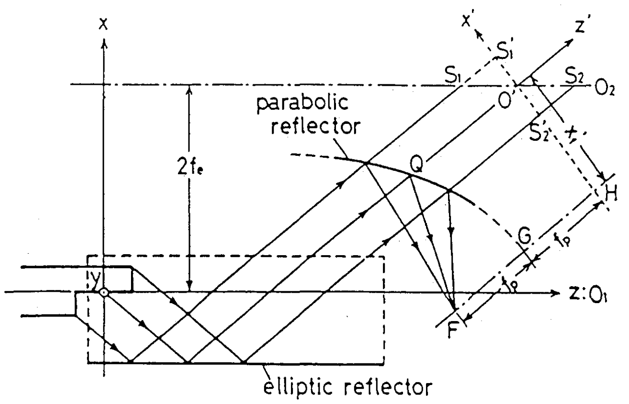


Fig. 2. Schematic diagram of quasi-optical antenna system.

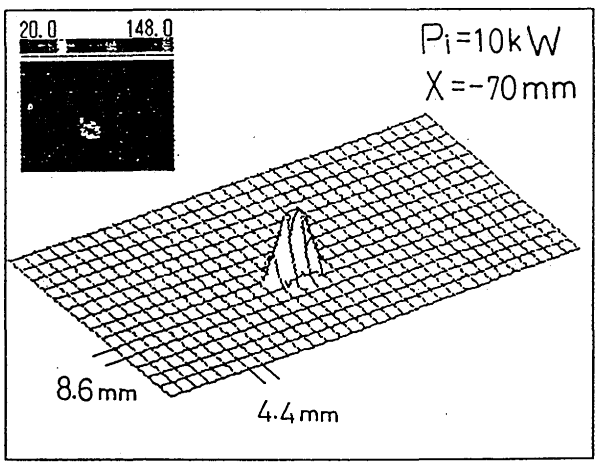


Fig. 3. Beam profile of the focused 60GHz radiation measured at the focal point.

Cuming) located parallel with the y-z plane. The temperature distributions on the sheet were measured using a thermal video system (TVS-3000, Nippon Avionics).

The beam profile observed at around the focal plane (x= -70mm), shown in **Fig. 3**, clearly demonstrates a finely focused and nearly Gaussian beam profile with a full width at half maximum of ~10mm. The focused power distribution showed an excellent agreement with the predicted profile using a simulation code[9]. These results demonstrated the feasibility of a high energy-density beam with 100 kW/cm^2 in the case of 100kW operation.

The heating characteristics have been studied in terms of the pulse width of the irradiated beam. The focused beam was irradiated onto alumina samples with inclusion of 5wt% SiC using the experimental setup schematically illustrated in **Fig. 4**; 74 mm in diameter and 80 mm in length with a beam entrance of 39 mm in diameter and a viewing port of 40 mm in diameter for temperature measurement. The sample temperature was measured using the thermal video recorder which was separately calibrated to thermocouple

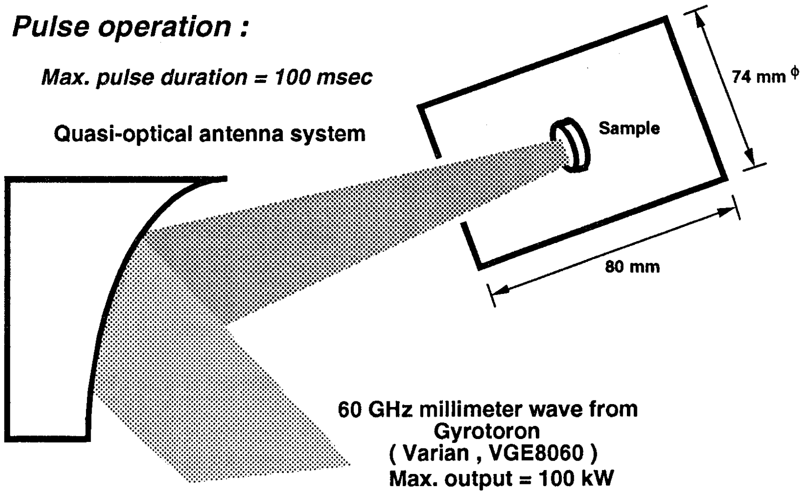


Fig. 4. Experimental setup used for heating and sintering of ceramics with the focused 60 GHz beam.

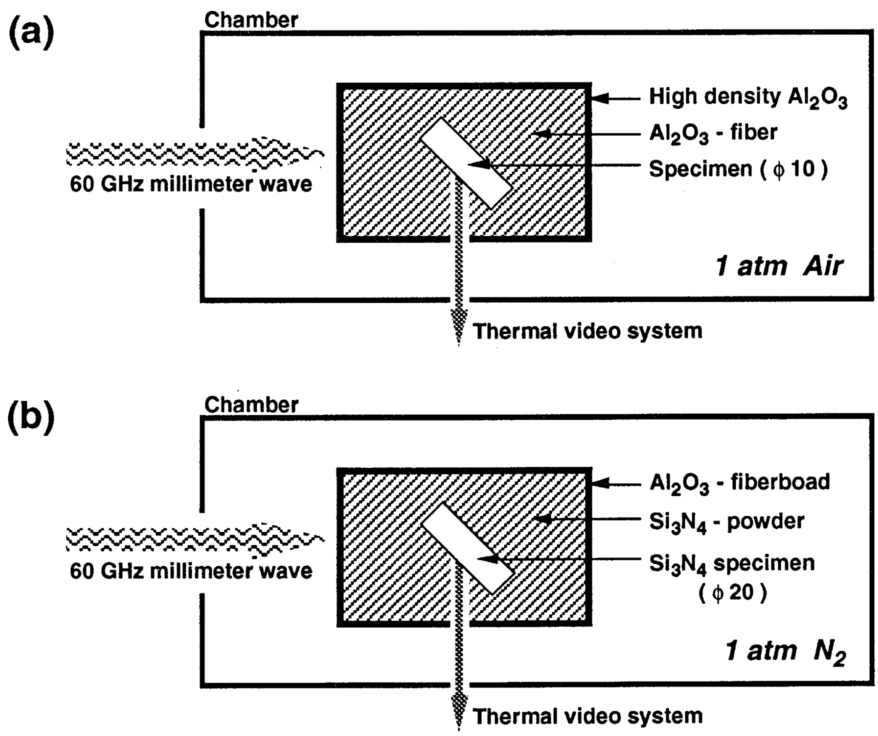


Fig. 5. Experimental setup used for thermal insulation of (a) yttria-stabilized zirconia samples and (b) silicon nitride samples.

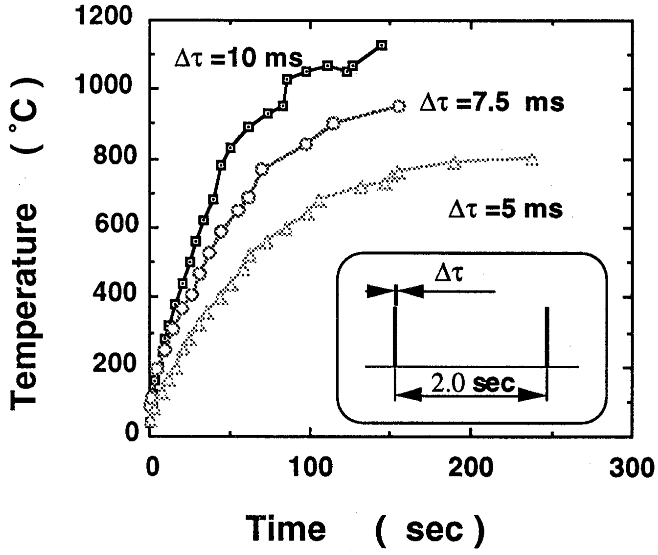


Fig. 6. Heating characteristics of alumina (5wt% SiC) samples fired using the focused 60-GHz beam with pulse width as a parameter.

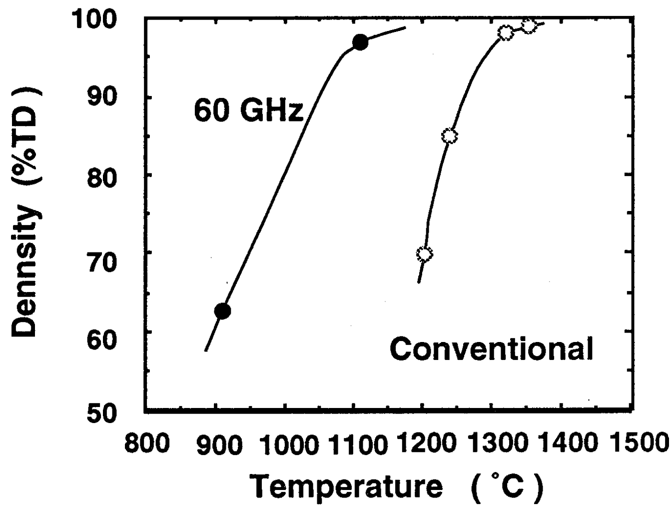


Fig. 7. Accelerated densification of yttria-stabilized zirconia was demonstrated with 60-GHz firing in pulse operation.

readings. Here the cavity was located so that the sample was positioned at the focal point of the beam, and the pulse irradiation was performed in air at a room temperature. The alumina powder used in this work was Sumitomo AKP-50 (α-Al_2O_3 > 99.995%) with a mean particle size of ~200 nm. The silicon carbide powder used in this work was Showa Densic Ultrafine (SiC > 98 %) with a mean particle size of ~370 nm. Green bodies of alumina and alumina-SiC mixture were formed by cold pressing to obtain the green density of 54 and 57 %TD, respectively.

Sintering of yttria-stabilized zirconia was carried out using a cylindrical applicator with a experimental setup for thermal insulation shown in **Fig.5 (a)**. The zirconia powder used in this experiment was grade TZ-8Y (Tosoh; ZrO_2-8mol% Y_2O_3) with an average particle size of ~200nm, which was formed to obtain green bodies by the slip casting method. The applicator used in the experiment had a dimension of 109 mm in diameter and 420 mm in length (L/λ~84; L-scale length of the applicator, λ-wavelength of radiation). The focused 60GHz beam was injected from the axial direction of the applicator through a 1-mm thick quartz window. The workpiece covered with alumina fiber for thermal insulation was fired with the focused beam in pulse operation with a 10-ms duration at a 0.5-Hz repetition rate. Heating and cooling rates of the samples were controlled to be 40-70°C/min and 60°C/min, respectively, by selecting appropriate pulse durations and the output powers from the 60GHz gyrotron.

Sintering of silicon nitride was also performed in the applicator with a thermal insulation configuration shown in **Fig. 5 (b)**. The silicon nitride used in this work was grade Ube-SN-COA (Ube Industries, Ltd.; E-10, with the inclusion of 3%Al_2O_3 and 5%Y_2O_3) with a mean particle size of ~200 nm. Green bodies of silicon nitride were prepared by the slip casting method.

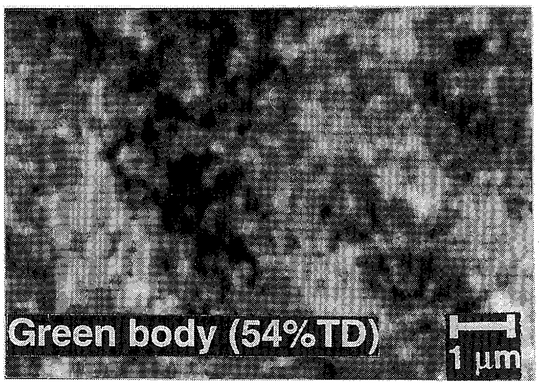
3. Results

The heating characteristics of alumina samples with 5wt% SiC were investigated using the finely focused 60GHz millimeter-wave radiation in pulse operation. The results summarized in **Fig. 6** show the temporal variations of the sample temperature with pulse width as a parameter. Here it should be pointed out that the horizontal axis denotes the real time instead of the sum of the irradiated pulse durations, which is 1/200 of the real time for the pulse width of 10 ms; i.e., the temporally averaged incident power for 60-kW operation is 300 W. Within the region where the temperature varies linearly with time, the heating rate has been shown to have a linear dependence on the pulse width. A temperature saturation is also observed, possibly due to the balance between the absorbed radiation power and the cooling rate (conduction loss and radiation cooling). Furthermore it has been observed that, even with an averaged power as small as 300 W, the sample can be heated quite rapidly (within 150 s) to ~1200 °C, which is typically required to sinter ceramics such as alumina and zirconia.

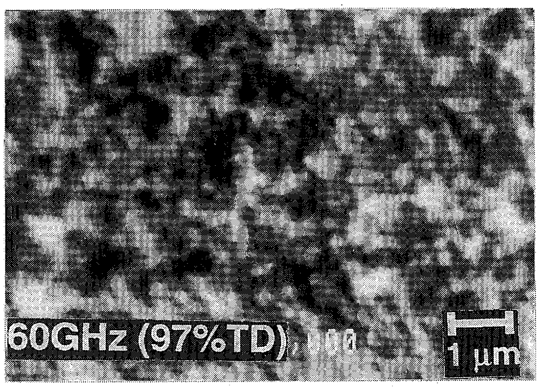
The results summarized in **Fig. 7** clearly demonstrate the accelerated densification of the yttria-stabilized zirconia samples at lower temperatures during the 60-GHz sintering for 60 min. The zirconia sample sintered with 60 GHz radiation at 1120°C for 60 min reached 97 % of the theoretical density (TD). In contrast, to achieve the same density conventionally required a temperature of ~ 1300°C.

The fracture surfaces of the green body and the sample densified to 97%TD are compared in **Fig. 8**. In this example the zirconia workpiece was kept at 1120°C for 60 min for sintering. These micrographs indicate that the densification of the yttria-stabilized zirconia has been achieved without suffering appreciable grain growth. The grain size of 60GHz sintered zirconia was approximately equal to that of the green body (~200nm), which is in contrast to the grain size of the conventionally sintered zirconia (~3000nm). At higher temperatures (1360°C), however, significant grain growth (~2000nm) was observed in the sample (96 %TD) sintered for 30 min using the focused 60 GHz radiation in pulse operation, but was still smaller than the grain size obtained from the conventionally sintered zirconia. It is remarkable that the 10-ms pulse irradiation by a 60GHz beam at a small repetition rate of 0.5 Hz has achieved densification of zirconia with a finer grain size at a lower temperature.

The results summarized in **Fig. 9** clearly demonstrate the accelerated densification of the silicon nitride samples at lower temperature in the 60-GHz sintering experiments performed using the configuration shown in Fig. 5 (b). Here it should be noted that the experiments with the 60 GHz beam were performed in an atmospheric pressure of nitrogen, while the results for the conventional method were obtained from the sintering experiments performed under nitrogen pressures as high as 10 kgf/cm^2. The silicon nitride



(A)



(B)

Fig. 8. Fracture surfaces of yttria-stabilized zirconia (ZrO2 - 8mol% Y2O3);(A) green body (54%TD) and (B) millimeter-wave firing at 60 GHz (97%TD).

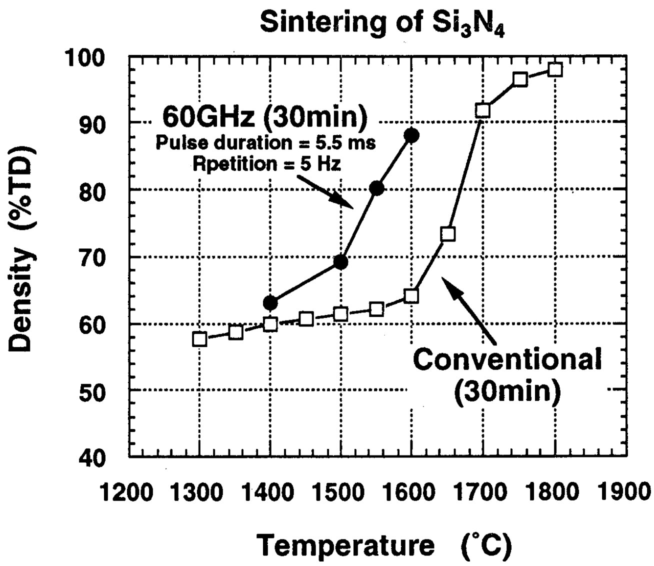


Fig. 9. Accelerated densification of silicon nitride was demonstrated with 60-GHz firing in pulse operation. Sintering experiments were performed in nitrogen pressures of 1 and 10 kgf/cm^2 for the 60GHz firing and the conventional method, respectively.

samples sintered at 1600°C for 30min reached nearly 90%TD. In contrast, to achieve the density as high as 90%TD conventionally required a temperature of 1700°C even with specimens sintered at nitrogen pressures as high as 10 kgf/cm^2.

4. Summary

The results of ceramic sintering experiments performed with a 100-kW 60-GHz gyrotron in pulse operation together with the fine focusing of high-power pulsed 60GHz radiation using quasi-optical nature of the millimeter-wave radiation have been presented.

Feasibility of the high energy density radiation (~100kW/cm^2) method was experimentally demonstrated. The heating characteristics of alumina using the focused beam in pulse operation were examined.

The efficient sintering of yttria-stabilized zirconia was demonstrated at about 200°C lower temperature than that conventionally required. Millimeter-wave firing in pulse operation resulted in the production of densified zirconia with finer grain size. Accelerated densification of the silicon nitride samples was also observed at lower temperature.

Acknowledgments

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