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INFLUENCE OF DIFFERENT METHODS OF CONTROLLING MICROWAVE SINTERING
The characteristics of oxide ceramics

M. Yasuoka[*], *Y. Nishimura, T. Nagaoka and K. Watari*

National Institute of Advanced Industrial Science and Technology (AIST) 2266 Anagahora, Shimo-Shidami, Moriyama-ku Nagoya, Aichi 463-8560, Japan

The ceramic industry uses enormous amounts of energy to produce products at high temperatures. Energy-saving measures based on sintering process improvements are examined.

Oxide ceramics were irradiated by two different methods during microwave sintering: intermittent use of the same high power levels (time-control method) and continuous use of lower, increasing levels of power (power-control method). We found that: 1) the power consumption and efficiency of the time-control method were lower than those of the power-control method; 2) the power-control method gave more precise control of temperature under our experimental conditions; 3) sample densification did not differ greatly between the two methods; and 4) grain growth did not differ greatly between the two methods.

Keywords: *alumina, barium titanate, microstructure, microwave sintering, power-control method, time-control method, zinc oxide*

Introduction

To protect the Earth's environment, 21st Century manufacturing industries will have to reduce their consumption of energy. Because the ceramic industry uses sintering processes to make its products, the use of the high temperatures needed requires large quantities of energy. Ceramics are sintered by begin placed into a uniformly heated environment provided by a gas or an electric furnace. This method fits well for mass production, because the temperature of a relatively large volume can be uniformly controlled. However, a great portion of the energy is consumed in maintaining the temperature of the surrounding furnace material or container, rather than being used in the actual product manufacturing. If the energy could be used more efficiently, less energy would be consumed, resulting in energy savings and improvements in the sintering process. The microwave sintering process has attracted attention since the 1990s [1–6] as being energy efficient. The advantages of microwave sintering are: 1) selective heating can be performed; 2) homogeneous heating is achieved; and 3) the temperature can be raised or lowered rapidly. By employing these advantages, this process can be applied to the fabrication of products of various kinds in small quantities.

When samples are heated using power from an electric furnace or a microwave furnace, two kinds of methods can be used to control the temperature: 1) intermittent powering of the magnetron at a fixed high power output (time-control method); or 2) continuous powering of the magnetron with a variable power output (power-control method). In a conventional electric furnace that uses a heating element, the whole interior space is heated together with the sample and there is no big difference between the two above-mentioned methods in terms of the energy added to the sample. In contrast, because the microwave sintering process uses self-generation of heat by the sample when it absorbs the microwaves, it is thought that the heating state of the sample can be influenced by the microwave irradiation method. Moreover, differences in the pattern of energy absorption by the sample arise when the sample is direct-heated by different irradiation methods, so differences in energy efficiency could result.

We irradiated a sample by using two kinds of irradiation methods during microwave sintering: a high amount of electric power applied intermittently and a lower dose one applied continuously. We report here the energy efficiency produced by differences in the irradiation methods and the resulting microstructure development of the samples.

Experimental

Zinc oxide (Wako Pure Chemical Industry Co., Osaka Japan; 95.0% purity, 0.02 μm particle size), alumina (TM-DAR, Taimei Chemicals Co., Nagano Japan)

* Author for correspondence: yasuoka-m@aist.go.jp

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Akadémiai Kiadó, Budapest, Hungary
Springer, Dordrecht, The Netherlands

and barium titanate (BT-01, Sakai Chemical Industry Co., Japan) were used as raw materials. ZnO was suitable as a model material because it tends to absorb microwaves [7–10]. Alumina is typical oxide ceramics [11–14]. Barium titanate is a typical ceramic material for the electronic industry [15–17]. Green pellets with a diameter of 18 mm were formed using a uniaxial pressure of 17 MPa and a cold isostatic pressure of 98 MPa. A magnetron multimode microwave furnace (MW-Master, Mino Ceramic Co. Ltd., Mizunami Japan) was used for the sintering experiments at 2.45 GHz. Figure 1 is a schematic illustration of the system. The sample was placed into a thermally insulated box, to the inner surface of which SiC had been applied. The SiC acts as a susceptor. The generation source of the microwaves was a magnetron with a 1.5 kW maximum rating at 2.45 GHz. With the power-control method, temperature was controlled by adjusting the voltage and current to induce a variable load on the magnetron. With the time-control method, temperature was controlled by adjusting the oscillation time of the magnetron, to which a maximum load was applied. The sample temperature was measured with an optical radiation thermometer. Density was measured by the Archimedes method. Microstructural observations of the fracture surfaces were made under a scanning electron microscope (SEM, JSM-5600N, JEOL Ltd., Akishima Japan).

Results and discussion

Figure 2 shows the temperature change of a sample treated by the power-control method. The heating rate was 0.5°C s^{-1} and the sintering temperature was 900°C for 600 s. The temperature difference between the set and measured values was very small. The temperature variation during holding at 900°C was about 3°C. The advantages of the power-control method were: 1) temperature change could be performed smoothly; and 2) the magnetron could be used at a comparatively low output . Because the power supply needed to be controlled by changes in voltage and current, one disadvantage of this method is that more complicated equipment could be needed.

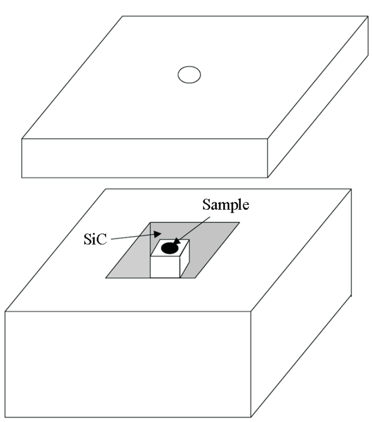

Fig. 1 Illustration of sample setting for microwave sintering

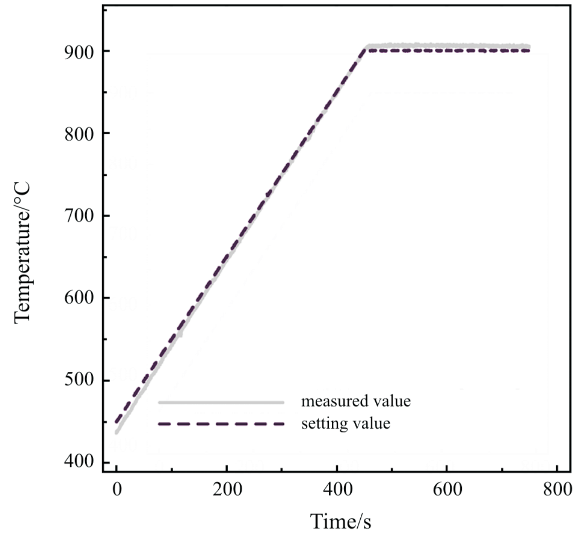

Fig. 2 Relationship between heat-treatment time and temperature using power controlling method

Figure 3 shows the temperature change of a sample treated by the time-control method. The heating conditions were the same as in the output-control method. Compared with Fig. 2, there was a greater change in temperature during temperature raising or holding. There was a 10°C change over 10 s at any temperature. Because the current and voltage of the power supply were fixed, one advantage is that the power supply equipment needed is simple. One disadvantage is that fine-tuning of temperature control is difficult and the magnetron would tend to break because of the large repeated bursts of electric power.

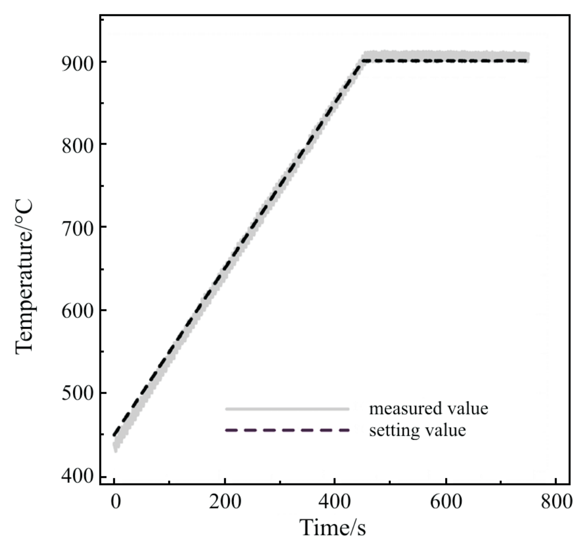

Fig. 3 Relationship between heat-treatment time and temperature using power controlling method

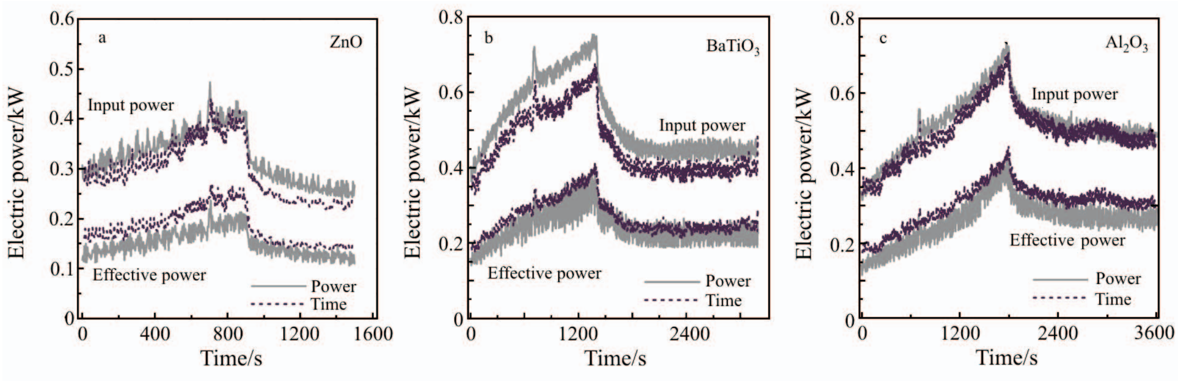

Fig. 4 Relationship between heat-treatment time and input and effective power using each controlling method, a – ZnO, b – $BaTiO_3$, c – Al_2O_3

Changes in the power output from the magnetron and the effective electric power during heating are shown in Fig. 4. The electric power output is the measured quantity of electric power emitted from the magnetron, and the effective electric power is the value achieved by subtracting the quantity of reflected electric power from the electric power output. The magnitude of reflected electric power is the dose of electric power that returns to the magnetron without being consumed by the microwave furnace. With the time-control method, on-off control was performed every 10 s as 1 cycle to protect the magnetron. The average of the quantity of power applied for 10 s on both sides of the time concerned was used to calculate the quantity of electric power at a certain time point. The magnitude of electric power output from the magnetron increased with temperature with both of the control methods. When the specimen reached a fixed temperature, the power output value decreased gently. The total quantity of electric power used was less with the time-control method than with the power-control method. On the other hand, the change in the effective electric power followed the same curve as the change in the electric power output. With the time-control method, the absolute effective electric power was greater than the electric power output. When a material absorbs microwaves efficiently the effective electric power is large. Vibration of molecules starts when enough energy is supplied. Comparison of high-output irradiation for a short time and low-output irradiation for a long time revealed that the amount of energy exceeding standard energy was greater with high-output irradiation for a short time. We think that this is caused by a difference in absorption efficiencies between the two methods. On the other hand, we consider that the energy required for a rise in temperature of the material was the same with both methods, because the same heating conditions were set. However, the absolute effective electric power was larger with the power-control method than with the time-control method. With the power-control method only the energy required for heating was supplied. In contrast, with the time-control method there were time gaps (no output), which resulted in cooling-off periods. Therefore, it was necessary to maintain the temperature by supplying a greater amount of energy.

We investigated the influence of heating method on the characteristics of sintered bodies. The influence of the irradiation method on density is shown in Fig. 5. The two curves show almost the same tendency at any samples. The change in the zinc oxide density is described in detail. When the sintering temperature exceeded 800°C, samples were equally dense by either method. However, when the sample was sintered at 700 or 750°C, the density using the

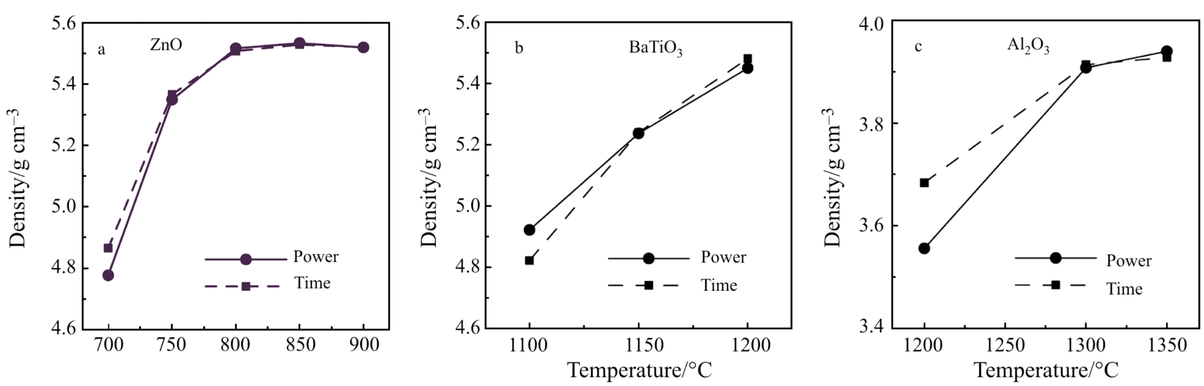

Fig. 5 Density of samples as a function of sintering temperature, a – ZnO, b – $BaTiO_3$, c – Al_2O_3

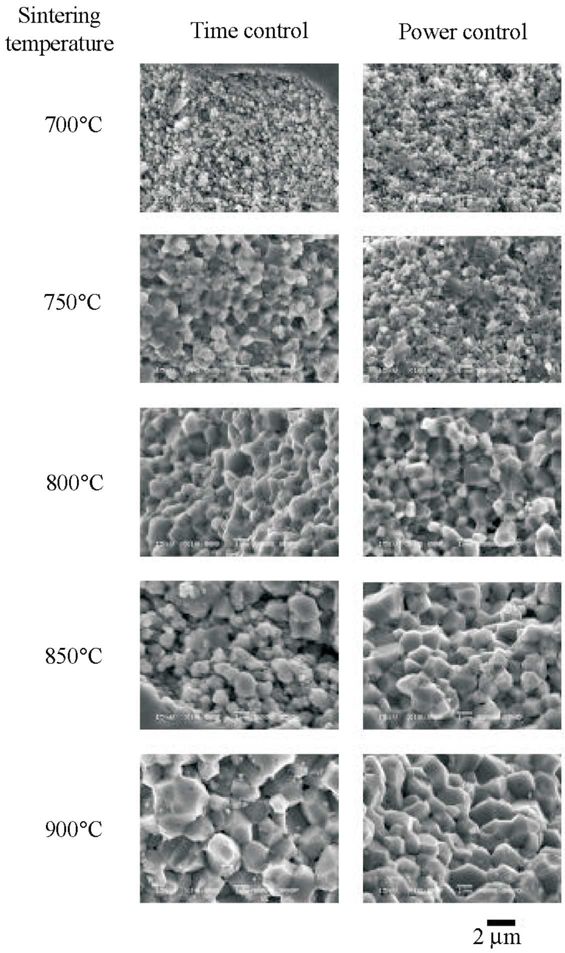

Fig. 6 SEM micrographs of sintered bodies

time-control method was higher than with the output-control method. When it is compared with retention temperature, the temperature of a sample becomes slightly high on using time controlling method. This is probably why the density was slightly higher using this method. This phenomenon was observed in sintering alumina too.

The microstructure of each sample was observed. Because any samples show the same tendency, the zinc oxide result is shown as a representative example. SEM micrographs of sintered bodies are shown in Fig. 6. The particle size of zinc oxide was about 0.2 μm when the sintering temperature was 700°C, indicating the start of sintering of the zinc oxide, and with increasing sintering temperature the particle size increased. The particle size was larger when the time-control method was used, probably for the same reason that the density was greater. When the sintering temperature was 800°C or more, the microstructures were the same with both methods. We consider that the microstructure was probably influenced by the energy supplied as temperature rather than by the magnitude of instantaneous energy.

Conclusions

We obtained the following results when oxide ceramics were sintered by two different microwave irradiation methods:

- Power consumption and efficiency were lower with the time-control method than with the power-control method.
- The power-control method gave more precise temperature control under our experimental conditions.
- Densification was influenced slightly by irradiation method.
- Grain growth was influenced slightly by irradiation method.

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