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# Microwave Application in the Heating of Low-Loss Ceramic Materials

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***Abstract*** The zirconia-alumina composite, a low loss material, was sucessfully sintered using a 2.45 GHz microwave radiation. The dense zirconia was used as a microwave coupling aid. The effect of microwave power level on the heating rates of samples and the feasibility of microwave energy use in processing ceramic materials were obtained. It was also obtained how to accurately measure the temperature. According to the microwave heating theory, heating mechanisms were discussed.

## 1. INTRODUCTION

Material processing using microwave energy is a rapidly expanding technology. In particular, the processing of ceramics, glasses, and composite materials through this technology offers the potential for increased productivity, reduced cost, and improved material performance. The microwave energy has been used in sintering, joining, and melting of ceramic materials by a number of researchers. In the late 1940's, Von Hippel[1] began investigating how oxide materials interacted with a microwave radiation and how the loss characteristics of these materials varied as a function of microwave frequency. In the mid 1960's, Tinga et al.[2,3] investigated how certain oxides were heated in a microwave field and they developed microwave cavities that caused materials to be uniformly heated using 915 MHz and 2.45 GHz microwave radiation. Recently, Meek et al.[4-9] reported that microwave heating at 2.45 and 60 GHz obtained enhanced material transport, rapid sintering, and different microstructures for various multi-component oxide, composite, and glassy materials. Especially, to sinter low loss ceramics at a 2.45 GHz, they suggested the use of various materials to be acted as both a microwave coupler and a crack inhibitor upon microwave heating.

According to the concept of microwave heating theory,[1,10-12] when electric field vector is normal to the planar surface of matter, electric field intensity(Volt/cm) is given by;

$$E_2 = E_1(\kappa_1/\kappa_2) \qquad (1)$$

where $E_1$ is the applied electric field intensity, $E_2$ is the internal electric field intensity, and $\kappa_1$ and $\kappa_2$ are the relative dielectric constant of region 1 and 2, respectively.

Power density (Watt/cm$^3$), the power absorbed per unit volume, is given by;

$$P = K f E \kappa \tan \delta \qquad (2)$$

where P is the power absorbed per unit volume, K is the constant, E is the electric field intensity, f is the microwave frequency, $\kappa$ is the relative dielectric constant, and tan $\delta$ is the dielectric loss tangent. Equation (1) and (2) describe how an electromagnetic field behaves at an interface between two dielectric media: since the electric field intensity and the power density are greater in the porous regions between grains within a material, the microwave radiation couples more strongly to the regions of low density and low dielectric loss. It is ben-

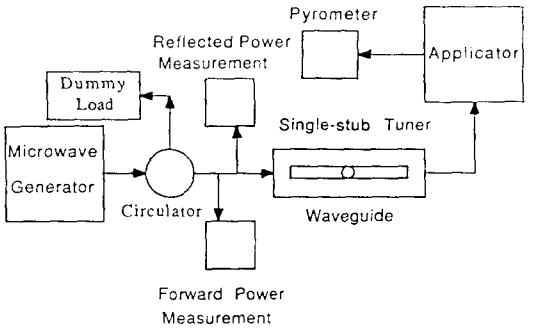


Fig. 1. The microwave heating system.

eficial to characterize how ceramic materials are heated in a microwave field.

In the present study, zirconia-alumina composite(a low-loss material) was heated using a 2.45 GHz microwave radiation. This paper discusses microstructural analysis and sintering data of samples. Also, the results of the present study explain how ceramic materials interacted with a microwave field.

## 2. EXPERIMENTAL PROCEDURE

Microwave heating was done in a Cober model S6F microwave heating system. The block diagram of this system is shown in Fig. 1. In this system, microwave power is supplied to an applicator through a waveguide by a 6kW variable-power microwave generator operating at a 2.45 GHz. The components in the microwave generator are a 3 port circulator(tube protective device) and a dummy load (water). They are directly water cooled. A pair of meters are used to indicate the forward and reflected power flow into and out of a waveguide. A multi-mode resonant cavity serves as an applicator that is coupled to power source using a WR-284 waveguide with a single-stub tuner which matches impedance between source(microwaves) and load(samples). The applicator($61\times65\times69$cm) is a box made with the stainless steel which is seam welded to prevent arcing. A reaction cavity ($24\times24\times25$cm) inside the applicator is located at the region of highest electric field intensity. The reaction cavity is covered with a Babcock-Wilcox 17C alumina insulator. The alumina insulator, transparent to microwaves but opaque to infrared radiations, forms an infrared cavity to trap heat generated within the load from microwave irradiation.

The composite powder 80 wt% $ZrO_2$(+3mol % $Y_2O_3$) 20 wt% $Al_2O_3$ was used as a starting material. Pellets($\phi\approx1.28$cm, h≈0.71cm, w≈2.4-2.5g) were prepared by cold pressing at 230MPa. Small size pellets were used to reduce density variations caused by steep thermal gradients during microwave heating. Green densities of samples were about 50% of theoretical density. The samples were always located at the center of a reaction cavity, which was in the region of highest electric field intensity, in all experiments. To maximize microwave heating, a single-stub tuner was controlled to match the impedance between microwaves and samples during the tests. For any given forward power setting, the optimum heating condition was obtained when reflected power was at its minimum. The relative occurrence in various power levels and temperature of the samples were investigated at 0.5kW increments ranging from 0.5 to 2.0kW. The heating of the samples was stopped when coupling and burning of an insulator, arcing between microwaves and surroundings, or stabilizing and decreasing of temperature occur. Temperature of the samples was measured using an optical pyrometer and a modified R-type thermocouple. The modified R-type thermocouple was made as follows: after a commercial Pt-13 % Rh thermocouple was inserted into an alumina tube, a shield wire was wrapped around the alumina tube with the pitch of a shield wire, which was less than a quarter wavelength of microwave, to prevent any interaction with a microwave radiation. It was placed near the top of samples. The optical pyrometer was focused on the front edge of samples with a small hole in the front part of reaction cavity wall. Microstructure analy-

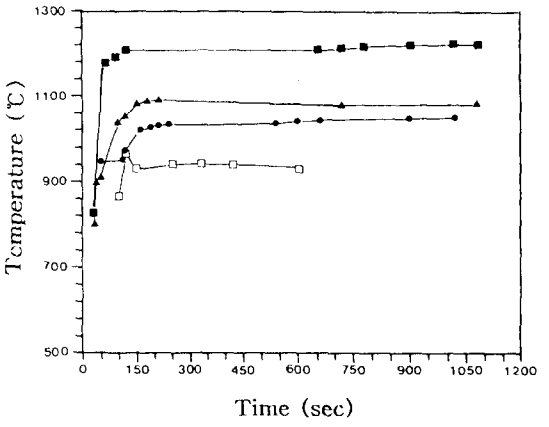


Fig. 2. Temperature profile of microwave heated samples on a susceptor at various applied power with adjustment of a tuner.(Input power:□-0.5kW, ●-1.0kW, ▲-1.5kW, ■-2.0kW)

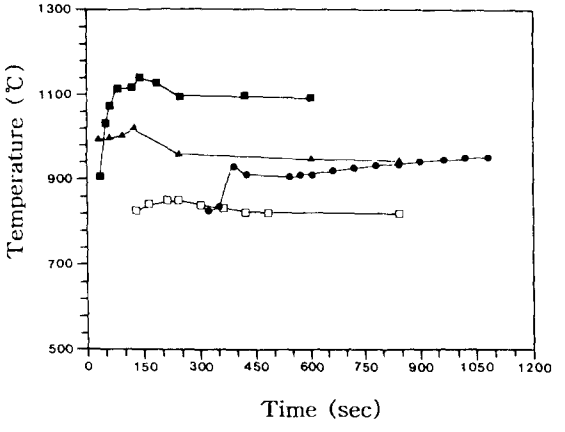


Fig. 3. Temperature profile of microwave heated samples on a susceptor at various applied power without adjustment of a tuner.(Input power:□-0.5kW, ●-1.0kW, ▲-1.5kW, ■-2.0kW)

sis of samples was done using an AMR 900 Scanning Electron Microscope(SEM). The composition of sample contents was obtained using Energy Dispersive X-ray Analysis (EDXA).

## 3. RESULTS AND DISCUSSION

Fig. 2 and 3 are the plots of temperature versus heating time at various input power with and without adjustment of a tuner, respectively. All samples showed an increase in heating rates when input power increased.

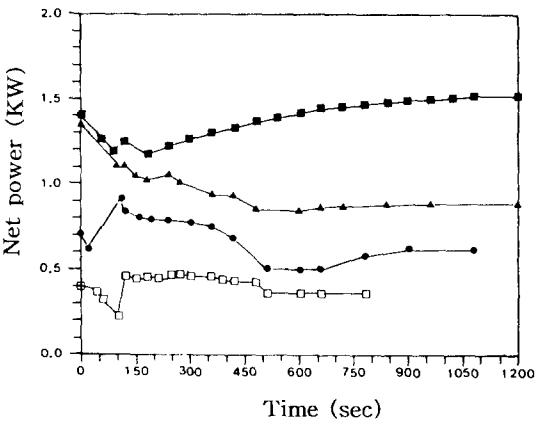


Fig. 4. Net power profile at various constant input power with adjustment of a tuner.(Input power : :□-0.5kW, ●-1.0kW, ▲-1.5kW, ■-2.0kW)

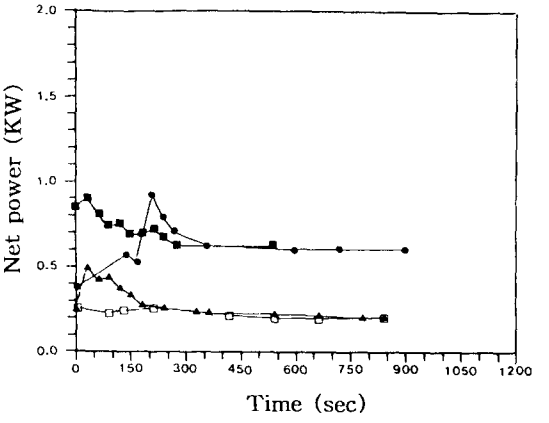


Fig. 5. Net power profile at various constant input power without adjustment of a tuner.(Input power : :□-0.5kW, ●-1.0kW, ▲-1.5kW, ■-2.0kW)

They coupled easily to microwaves and reached temperatures in excess of 800 ℃ in a few minutes. From equation (2), it was expected that temperature increase was due to the increase of power density and loss tangent within samples. The temperature with adjustment of a tuner was much higher than that of without adjustment of a tuner. Also, the coupling time with adjustment of a tuner was shorter than that of without adjustment of a tuner. The net microwave power(input power minus reflected power) with time at various input power increased with adjustment of a tuner and decreased without adjustment of a tuner

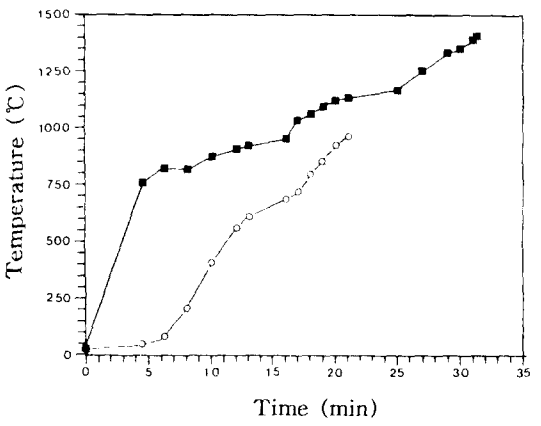


Fig. 6. The measured temperature profile of samples by using IR meter and thermocouple.( ○-Thermocouple, ■-IR meter)

as shown in Fig. 4 and 5. It meant that reflected input power increased with temperature increase without additional tuning. According to microwave heating mechanism,[1] once ceramic materials are microwave heated, the dielectric constant and the loss tangent of them increases. After that, resonant frequency changes to low frequency, and the degree of coupling decreases. As a result, the reflected power increases. In this investigation, it was necessary to adjust a tuner so that a reaction cavity came into resonance and the degree of coupling approached maximum coupling. Therefore, adjustment of a tuner caused samples to be matched better to microwaves, which resulted in optimum heating. This type of heating information may be used to select the optimum condition of microwave processing.

Fig. 6 shows temperature profile data taken using a modified R-type thermocouple and an optical pyrometer. In this study, the constant input power of 2.0kW was applied with adjustment of a tuner. The response of the modified thermocouple was much slower than that of the optical pyrometer. The determination of temperature using the optical pyrometer could be an adequate way of obtaining temperature data in samples heated with rapid rate. During the test, due to the concentration of an electric field on the metallic structure the modified thermocouple was arced, and then melted at about 1000℃; unwanted hot spots were produced as reported by Johnson et al.[13] Therefore, the direct temperature measurement using an optical pyrometer could be useful during microwave heating because it did not produce hot spots and resulted in more accurate temperature data. Since data were not available concerning the profile of a microwave field, it was impossible to explain more detail.

The microwave heating of samples with and without a susceptor was carefully done by controlling input power until sintering temperature was achieved. As shown in Table 1, 22 runs were made without a susceptor. The first 8 runs were conservative attempts and half of runs failed in reaching sintering temperature due to inadequate matching, insulation red-heating, and arcing. Nonetheless, the densities of samples were about 97-99% of theoretical. In the run 9-22, the constant input power of 0.5 kW was applied for the first five minutes and 0.5kW increments of input power were additionally applied for every 5 min. up to the input power of 3.5kW until samples coupled. After coupling, the input power decreased instantaneously at 2.0-2.5 kW to control desired heating rate and to prevent thermal run-away. In these runs, it was observed that some samples were cracked by ultra-high heating rate as a result of control difficulty and that the bottom of some samples was partially melted by hot spots due to thermal run-away. Runs 23-30 were made with a dense zirconia susceptor as a coupling aid because it had much higher loss characteristic than samples. The constant input power of 0.5kW was applied for first several minutes until samples coupled. After coupling, the input power increased gradually to reach sintering temperature. Just below the sintering temperature, the input power decreased rapidly at 1.0-1.5kW to prevent thermal run-away, and then was con-

Table 1. Data on microwave sintering of zirconia-alumina composites derived from 30 runs.

| Run Number | Quantity of Sample | Susceptor | Coupling Time (min) | Sintering Temperature (℃) | Sintering Time (min) | Input Power (kW) | Heating Rate (℃/min) | Density (%) |
|---|---|---|---|---|---|---|---|---|
| 1 | 3 | w/o | 83 | 1800 | 45 | 3 | 30 | 99 |
| 2 | 5 | w/o | 105 | 1376 | – | 4 | – | 97 |
| 3 | 3 | w/o | 21 | 1800 | – | 3.5 | 52 | 99 |
| 4 | 4 | w/o | 89 | 1800 | 45 | 4 | 114 | (severe crack) |
| 5 | 6 | w/o | 36 | 1306 | – | 4.2 | 237 | (severe crack) |
| 6 | 3 | w/o | 92 | 1800 | – | 4.5 | – | 97(crack) |
| 7 | 3 | w/o | 17 | 1800 | 45 | 2 | 107 | 97 |
| 8 | 3 | w/o | 20 | 1800 | – | 3 | – | – |
| 9 | 3 | w/o | 95 | 1800 | 45 | 3 | 121 | 96(crack) |
| 10 | 3 | w/o | 17 | 1800 | 45 | 3 | 52 | 97(crack) |
| 11 | 3 | w/o | 109 | 1800 | 45 | 4 | 37 | 96(crack) |
| 12 | 3 | w/o | 41 | 1800 | 45 | 3.5 | 50 | 97(crack) |
| 13 | 3 | w/o | 33 | 1800 | 45 | 3.5 | 80 | 97 |
| 14 | 3 | w/o | 20 | 1800 | 45 | 3.5 | 48 | 97 |
| 15 | 3 | w/o | 20 | 1800 | 45 | 3 | 81 | 97(crack) |
| 16 | 3 | w/o | 20 | 1800 | 45 | 3 | 74 | 97 |
| 17 | 5 | w/o | 41 | 1800 | 45 | 3.2 | 48 | 99 |
| 18 | 3 | w/o | 18 | 1800 | 45 | 3 | 109 | 98 |
| 19 | 41 | w/o | 17 | 1800 | 45 | 3.8 | 30 | 100 |
| 20 | 6 | w/o | 19 | 1800 | 45 | 3 | 62 | 100 |
| 21 | 3 | w/o | 40 | 1800 | 45 | 3.8 | 75 | 98(crack) |
| 22 | 3 | w/o | 32 | 1800 | 45 | 3.5 | 151 | 98(carck) |
| 23 | 3 | w | 4.5 | 1400 | 30 | 2 | 23 | 99 |
| 24 | 12 | w | 4.8 | 1400 | 30 | 3 | 19 | 100 |
| 25 | 21 | w | 3.3 | 1400 | 30 | 2.2 | 24 | 100 |
| 26 | 21 | w | 2.7 | 1400 | 30 | 2.4 | 19 | 100 |
| 27 | 21 | w | 2.8 | 1400 | 30 | 1.6 | 22 | 100 |
| 28 | 8 | w | 15 | 1400 | 30 | 3.4 | 28 | 99 |
| 29 | 6 | w | 13.7 | 1400 | 30 | 1.8 | 62 | (severe crack) |
| 30 | 6 | w | 18.9 | 1400 | 30 | 1.3 | 44 | (severe crack) |

trolled to reach the sintering temperature. These results showed that coupling time decreased with the increase of load mass. Also, samples coupled more easily at lower input power than those of without a susceptor due to greater mass(samples plus a susceptor). In the run 28-30, coupling time was delayed because samples were stacked at 90° to an electric field. It indicated that stacking samples might be placed parallel to the electric field, thus af-

Table 2. The densities of microwave sintered sample.

| Sintering Temperature(℃) | Sintering Time (Min.) | Initial Density (%) | Final Sintered Density (%) |
|---|---|---|---|
| 1200 | 5 | 50 | 94.99 |
| | 10 | 50 | 95.25 |
| | 30 | 50 | 95.92 |
| | 60 | 50 | 98.54 |
| 1300 | 5 | 50 | 94.72 |
| | 10 | 50 | 95.38 |
| | 30 | 50 | 96.88 |
| | 60 | 50 | 98.88 |
| 1400 | 2.5 | 50 | 98.50 |
| | 5 | 50 | 98.29 |
| | 10 | 50 | 98.63 |
| | 30 | 50 | 99.33 |
| 1500 | 2.5 | 50 | 97.94 |
| | 5 | 50 | 98.35 |
| | 10 | 50 | 100.0 |
| | 30 | 50 | 100.0 |

fecting their position relative to maximum field intensity. Thermal run-away would occur unless careful control of input power was exercised. It could be accomplished by manually decreasing input power level; decreasing input power gradually prevented thermal run-away within samples as reported by Roussy.[14] In the stable sintering temperature range up to 1800 ℃, thermal run-away became much easier to control because uniformly heated samples were formed. The stable sintering temperature could be maintained by applying constant input power at 2.0-2.5kW.

The microwave heated samples of zirconia-alumina composites listed in Table 1 were divided into two groups: one was heated without a susceptor; the other was heated with a susceptor. During microwave heating without a susceptor, a significant fraction of total heat generated in a reaction cavity was actually generated in an insulator. While samples were able to achieve sintering temperature, the external portion of an applicator was hot to touch. However, during microwave heating with a susceptor, most of heat was generated within samples. When temperature of samples was already up to 1400℃, the external portion of an applicator was relatively cooler to touch than that of the former case. Coupling mechanism in the initial heating step may be explained as follows: without a susceptor, initially a 2.45GHz microwave radiation couples with an insulator until samples reach at 700-1100℃ because of characteristic of low-dielectric loss in this temperature range. On the other hand, with a susceptor, the difference is initially coupling with a susceptor(dense zirconia) instead of an insulator. After samples are heated to 700-1100℃, they begin to couple efficiently because the loss tangent of the samples increases exponentially with temperature increase. Therefore, the samples are heated to sintering temperature and the insulator couples much less and efficiently insulates the

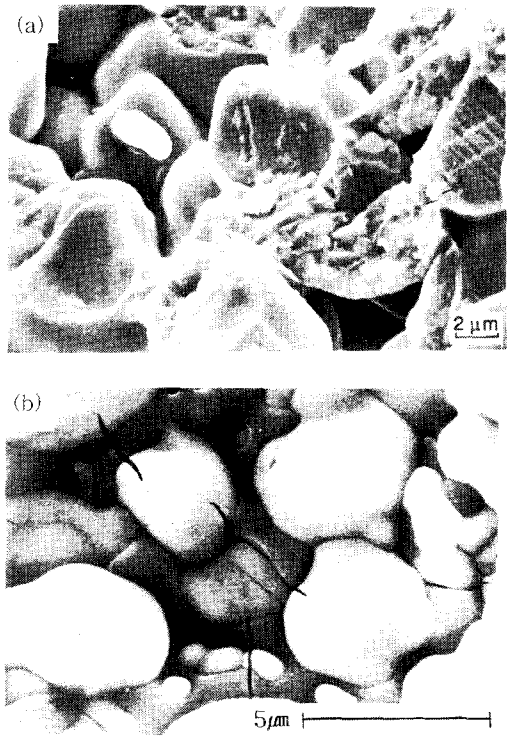


Fig. 7. Scanning electron micrographs of (a) fracture surface and (b) melted region in samples sintered at 1400℃ and 30min with a susceptor.

heated samples as reported by Meek.[9]

The bulk densities of microwave sintered samples are listed in Table 2. With optimum heating condition such as adjustment of a tuner and use of susceptor, samples were successfully sintered to 97–100% of theoretical density regardless of sintering temperature. Comparing data reported by Rajendran et al.[15] or Wakai et al.,[16] the densification rates of microwave sintered samples were much higher than those of conventionally sintered samples (i.e., HIPed samples) at comparable temperatures. It meant that microwave sintered samples were densified with fast heating rate, shorter sintering time, and lower sintering temperature. The fracture morphology of microwave sintered samples was mostly transgranular as shown in Fig. 7. Strongly bonded grain boundaries by the spread of a liquid phase resulted in transgranular fracture. The composition of Fig. 7(b) was determined by EDXA as shown in Fig. 8. Melted regions were alumina and grains were zirconia. The melting was believed to be caused by the formation of a liquid phase alumina when real temperature at intergranular region exceeded the melting point of alumina. Therefore, real sintering temperature was thought to be much higher than measured temperature because the melting point of alumina was 2054℃ and sintering temperature was 1400℃. It was expected that enhanced densification behaviour was due to the formation of liquid phase alumina. If sintering of ceramic materials in an electromagnetic field occurs as described in an earlier paper by Meek[10], the following assumption may explain how the alumina phase was melted along grain boundaries using the equation (1) and (2). If initial electric field intensity is greater in lower dielectric constant region(intergranular), electric field intensity and power density in the intergranular region will be greater than those in the bulk of grains. Therefore, microwaves will concentrate at the intergranular region. After liquid phase alumina is formed instantaneously, it is spreaded along the grain boundaries. Once the liquid will preferentially flow into the porous region, the microwaves will couple more strongly to the other low density region. As sintering progresses, the low density regions between grains vanishes, thus causing the overall bulk temperature to become more uniform. With further densification, microwave energy partitioning ceases.

## 4. CONCLUSIONS

Zirconia-alumina composite, a low loss material, was sintered successfully using a 2.45GHz microwave radiation with and without a susceptor. The dense zirconia was used as a microwave coupler to achieve stable heating during microwave processing. The optimum heating condition was obtained by adjustment of a single-stub tuner due to optimum matching the impedance between microwaves and samples. Microwave process under optimum

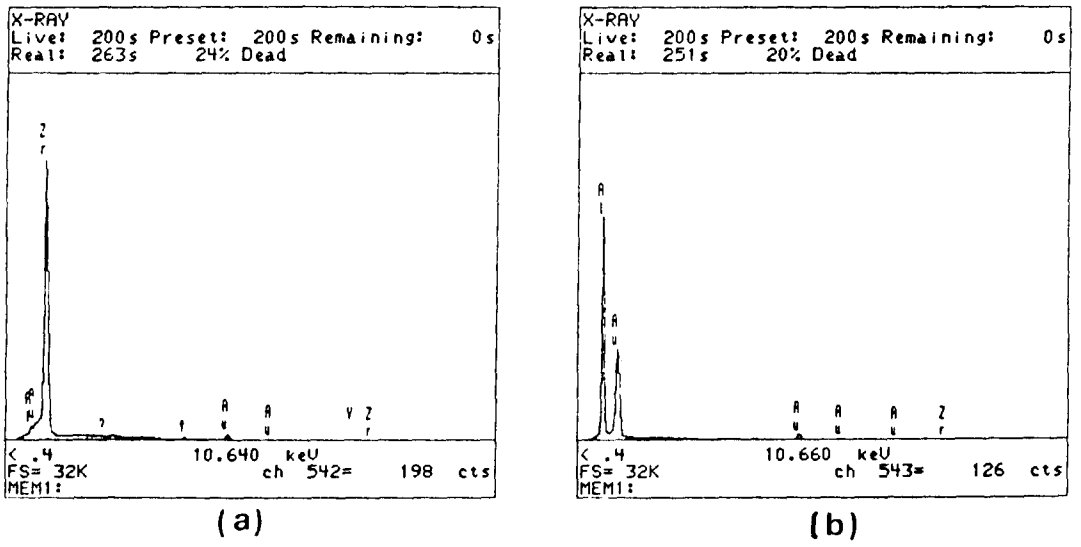


Fig. 8. EDXA of (a) grain and (b) melted region in Fig. 7(b).

heating condition maximized the temperature range that could be achieved with a given microwave heating system. Direct temperature measurement using an optical pyrometer was useful during the microwave heating. From the evidence of the formation of a liquid phase alumina, real microwave sintering temperature was much higher than measured temperature. It infered that electric field intensity and power density were greatly intensified in the porous region between grains. Microwave sintered samples were more rapidly densified than conventionally sintered samples for shorter sintering time, fast heating rate, and lower sintering temperature. Therefore, microwave process is very effective to sinter ceramic materials for its enhanced heating charateristics.

## REFERENCES

1. A.R. Von Hipple, in "Dielectric Materials and Applications", Technology Press of MIT and Wiley, New York, (1954).
2. W.R. Tinga and S.O. Nelson, J. Microwave Power, **8** (1), 23 (1973).
3. W.R. Tinga and W.A.G. Voss, in Microwave Power Engineering, 2, pp. 189-194, Academic Press, New York, (1968).
4. T.T. Meek, R.D. Blake, and T.G. Gregory, Ceram. Eng. Sci. Proc., **6**, 1161 (1985).
5. T.T. Meek and R.D. Blake, J. Mater. Sci. Lett., **5(3)**, 270-274 (1986).
6. T.T. Meek, M.H. Brooks, R.D. Blake, J.J. Petrovic, H. Jory, and J.V. Milewski, J. Microwave Power, **21(3)**, 193 (1986).
7. T.T. Meek, C.E. Holcombe, and N. Dykes, J. Mater. Sci. Lett., **6(9)**, 1060 (1987).
8. T.T. Meek, R.D. Blake, and J.J. Petrovic, Ceram. Eng. Sci. Proc., **8(7-8)**, 861 (1987).
9. T.T. Meek, R.D. Blake, J.D. Katz, J.R. Bradberry, and M.H. Brooks, J. Mater. Sci. Lett., **7(1)**, 28 (1988).
10. T.T. Meek, J. Mater. Sci. Lett., **6**, 638 (1987).
11. W.R. Tinga, in Microwave Processing of Materials, 124, pp. 33-44, Materials Research Society, Pittsburgh, (1988).
12. V.K. Varadar, Y. Ma, A. Lakhtakia, and V.V. Varadan, in Microwave Processing of Materials, 124, pp. 45-47, Materials Research Society, Pittsburgh, (1988).
13. C.C. Johnson and A.W. Guy, Proc. of the IEEE, **60(6)**, 692 (1972).
14. G. Roussy, A. Mercier, and J.P. Vaubourg,

J. Microwave Power, **20(1)**, 47 (1985).
15. S. Rajendran, M.V. Swain, and H.J. Rossell, J. Mater. Sci., **23(5)**, 1805 (1986).
16. F. Wakai, H. Kato, S. Sakaguchi, and N. Murayama, Yogyo-Kyokai-Shi, **94(9)**, 1017 (1986).