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Effects of Particle Diameter and Compacted Mass on Microwave Heating of Copper Powder

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Cu powders with different diameter and mass were compacted on quartz substrates, followed by microwave (2.45 GHz) irradiation in air. Microwave heating of Cu powder was quite anomalous. Both peak and steady-state temperatures appeared during microwave irradiation. In small Cu powder particles, the whole of Cu particle was heated, while in large Cu powder particles microwave heating occurred locally on the surface of the Cu powder particles. The minimum mass over which the temperature rise was observed decreased with increasing mean particle diameter of Cu powder. Thus, the microwave heating of the Cu powder varied depending on both the mean particle diameter and the compacted mass of Cu powder.
Keywords: copper powder, microwave heating, particle diameter, compacted mass

1. INTRODUCTION

Metal bulk is an excellent reflector of microwave energy and, in general, is not heated significantly by microwaves. Therefore, few applications have been conducted so far on heating metal bulk [1]. On the other hand, recently, there has been much interest in microwave heating of metal powders [2-7] and thin metal films, [8-10] which have large specific surface areas compared with metal bulk.

Roy *et al.* [2,3] reported that the microwave heating of a Cu powder-compact sample is quite anomalous although the sample heats up very fast in both electric and magnetic fields. On the other hand, Yoshikawa *et al.*[7] reported that Cu particles having diameters >2 mm are not heated well at the position of maximum magnetic field, and are only heated by the occurrence of electric discharge at the position of maximum electric field. The authors [10] previously reported that microwave heating of thin Cu film shows anomalous behavior in which an abrupt temperature rise and drop occur in the early stage of microwave irradiation, followed by a continuous temperature rise. However, there have been few reports regarding microwave heating of Cu powder, and also its heating behavior is not well understood.

In the present study, the microwave irradiation was carried out using Cu powder-compacts having different particle diameters and mass and the effects of these factors on microwave heating masses were examined.

2. EXPERIMENTAL

A 2.45 GHz, 1.8 kW microwave generator was used as the microwave source. Figure 1 shows a schematic diagram of the microwave irradiation test. The distribution of a microwave field was complex because of a multi-mode microwave cavity (section: 164×295 mm, height: 335 mm). A Cu powder-compact specimen was placed at the height of 90 mm from the bottom where it received the maximum incident flux of microwaves.

Four types of high purity Cu powders (Cu: 99.8 mass%; symbol 5Cu: maximum particle diameter of 5 μm, mean particle diameter of 2.7 μm; symbol 10Cu: maximum particle diameter of 10 μm, mean particle diameter of 6.5 μm; symbol 45Cu: maximum particle diameter of 45 μm, mean particle diameter of 18.3 μm; and symbol 100Cu: maximum particle diameter of 100 μm, mean particle diameter of 72.8 μm; Fukuda Metal Foil & Powder Co. Ltd.) were used.

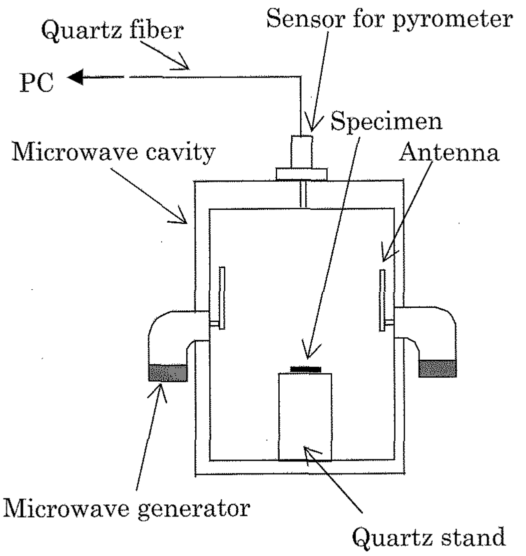

Fig. 1 Schematic diagram of the microwave irradiation test.

Figure 2 shows secondary electron (SE) images of the Cu powders used. All the powders are almost spherical.

Cu powders were compacted on quartz substrates (10×13×1 mm) having a low permittivity, followed by microwave irradiation at 1 kW for 1.2 ks in air. The net incident flux of microwaves to the Cu powder-

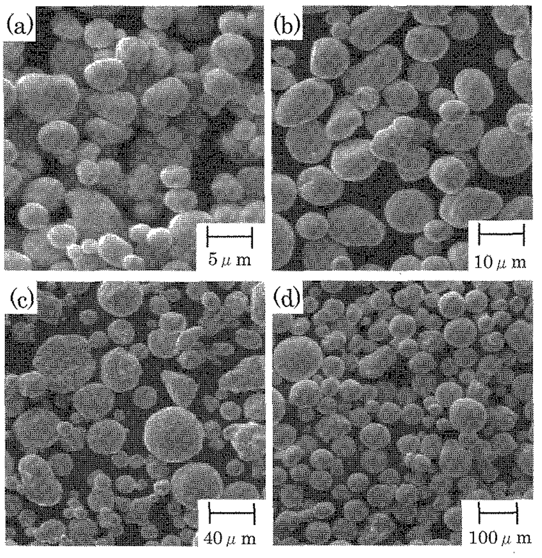

Fig. 2 SE images of the Cu powders used.
(a) 5Cu, (b) 10Cu, (c) 45Cu and (d) 100Cu.

compact specimens was 563 W. The temperature of compacted Cu powder was measured using a glass-fiber-type radiation pyrometer (IR-FL3, Chino Co. Ltd.). The microwave cavity was shielded to prevent the entrance of light. After microwave irradiation morphology of compacted Cu powder was examined by scanning electron microscopy (XL-30 ESEM, FEI Co. Ltd.).

3. RESULTS

Microwave heating behavior varied depending on both the particle diameter and the compacted mass of Cu powders. Figure 3 shows the change in temperature of 10Cu powders (compacted mass: 0.8 g) during microwave irradiation. The radiation pyrometer used in the present study was unable to measure temperatures lower than 100 °C because of the detection limit of the sensor. This is the reason behind the lack of temperature measurements in the early irradiation stage. Because a quartz substrate having a low dielectric property is heated little by microwave irradiation, the temperature rise is caused by the heating of Cu powder. As shown in Fig.3, the temperature change is quite anomalous. In the early irradiation stage, the temperature increased gradually

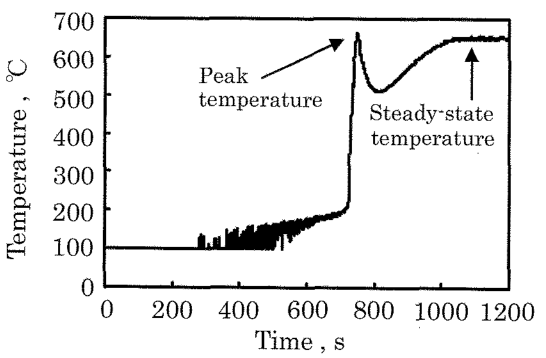

Fig. 3 Change in temperature of 10Cu powders (compacted mass: 0.8 g) during microwave irradiation.

with irradiation time. After reaching about 200 °C, abrupt temperature rise and drop occurred. Then the temperature increased again and remained almost unchanged in the late stage of microwave irradiation. Thus, both peak and steady-state temperatures appeared during microwave irradiation.

Figure 4 shows SE images of 10Cu powder (compacted mass: 0.8 g) after microwave irradiation. As shown in Fig.4(a), the sintering of Cu powder was achieved by microwave heating. This suggests that Cu powder is heated up to the temperature at which the sintering of Cu powder occurs. As shown in the high-magnification image (Fig.4(b)), irregularity of the surface of Cu powder particles was observed. Because no irregularity was observed on the surfaces of as-received Cu powder particles or the Cu powder particles heated using a conventional furnace, the irregularity may be due to a nonthermal effect of microwaves, such as ponderomotive force [11,12].

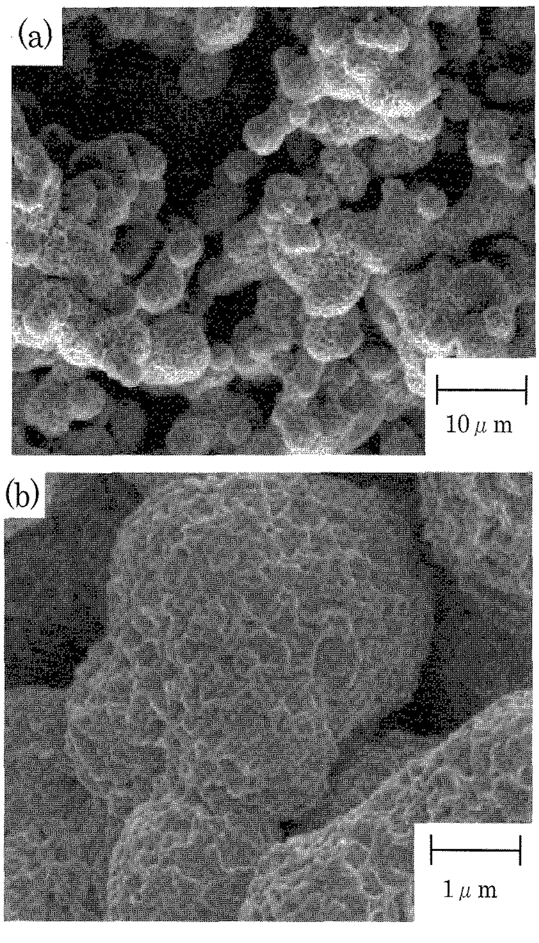

Fig. 4 SE images of 10Cu powder (compacted mass: 0.8 g) after microwave irradiation.

Figure 5 shows SE images of 100Cu powder (compacted mass: 0.4 g) after microwave irradiation. As shown in Fig.5(a), little sintering of Cu powder was achieved though Cu powder was heated by microwave irradiation. This is because the surface energy of large Cu powder particles is very small. In the heating temperature profile, the temperature drop after the peak temperature was lower than that of 10Cu

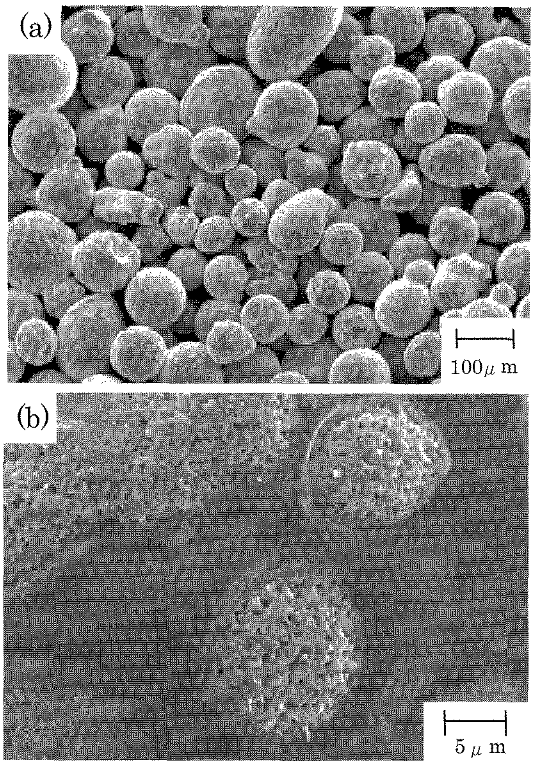

Fig. 5 SE images of 100Cu powder (compacted mass: 0.4 g) after microwave irradiation.

powder. Thus, the temperature drop seems to be pertinent to the sintering of Cu powder. Many white spots were observed on the surface of Cu powder particles (Fig.5(a)). In the high-magnification image (Fig.5(b)), irregularity similar to Fig.4(b), was observed in the white spots having a diameter of several μm. This suggests that microwave heating occurs locally in the case of large Cu powder particles.

As shown in Fig.3, both peak and steady-state temperatures appeared. Cheng *et al.*[3] reported that the temperature rapidly rises up to about 700 °C, then quickly drops to about 500 °C and increases again. However the reason was not explained. The authors [10] have reported similar behavior in the microwave heating of thin Cu film. According to the results, the temperature change is caused by various combinations of the change in the rate of temperature rise (ΔT) depending on the ratio of thickness to resistivity of thin Cu film, the increase in ΔT owing to Cu oxide and resistivity rise at elevated temperature, and the decrease in ΔT caused by Cu particle growth during microwave irradiation and thermal emission from the surface of thin Cu film. Thus, in the case of the microwave heating of Cu powder, the abrupt temperature rise may be caused by the increase in ΔT owing to Cu oxide on the surface of Cu powder particles and resistivity rise at elevated temperature, while the subsequent rapid temperature drop may be due to the sintering of Cu powder. The steady-state temperature in the late stage of microwave irradiation is caused by the establishment of a heating/thermal emission equilibrium.

Figure 6 shows the relationship between peak temperature, T_{peak}, and compacted mass of Cu powders with different diameters. The radiation pyrometer used

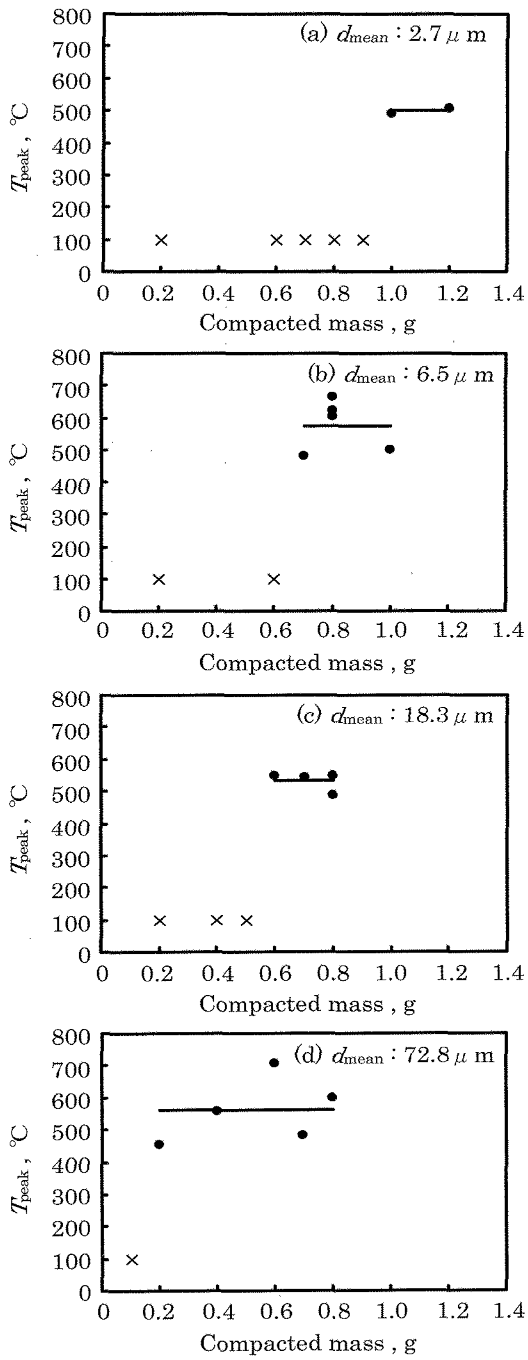

Fig. 6 Relationship between peak temperature, T_{peak}, and compacted mass of Cu powders with different diameters.

in the present study was unable to measure temperatures lower than 100 °C. The symbol × in Fig.6 shows that no temperature measurement was observed during microwave irradiation. The line in Fig.6 shows the average value of peak temperatures. Thus, microwave heating behavior varied depending on both particle diameter and compacted mass of Cu powder.

Figure 7 shows the relationship between the peak temperature, T_{peak}, and the mean particle diameter of Cu powder. The peak temperature was independent of the mean particle diameter of Cu powder and was almost constant. The scatter increased as the mean particle diameter of Cu powder became larger.

Figure 8 shows the relationship between the minimum mass over which temperatures higher than 100 °C were observed (Fig.6), m_{min}, and the mean particle diameter of Cu powder. The minimum mass decreased with increasing mean particle diameter of Cu powder.

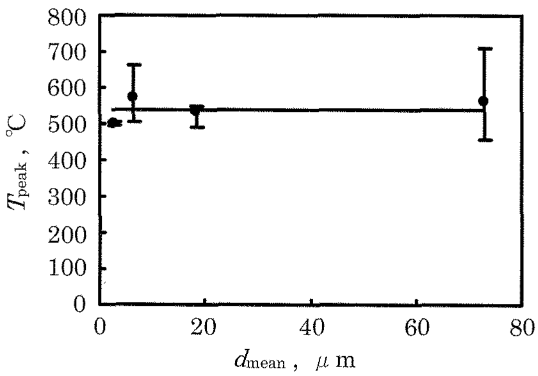

Fig. 7 Relationship between the peak temperature, T_{peak}, and the mean particle diameter of Cu powder.

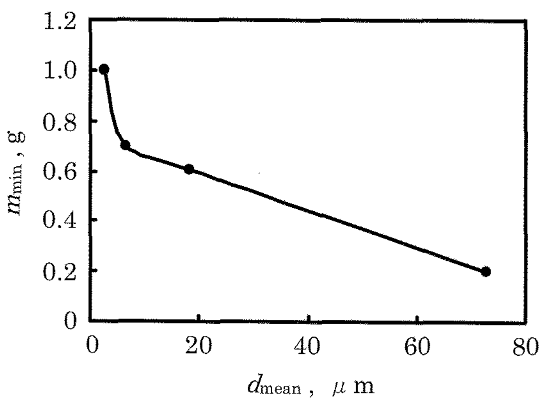

Fig. 8 Relationship between the minimum mass over which temperatures higher than 100 °C were observed (Fig.6), m_{min}, and the mean particle diameter of Cu powder.

4. DISCUSSION

Figure 9 shows a schematic illustration of microwave heating. Although Cu powder reflects most of the incident microwaves, some incident microwaves penetrate into the interior of Cu powder. The depth of penetration, that is, skin depth (defined as the distance from the surface into the material at which the power drops to e^{-1} of the original value) is given by [13]

$$\delta = (2/\omega\mu_0\sigma)^{1/2} \tag{1}$$

where δ is the skin depth, ω the angular frequency ($\omega = 2\pi f$, where f is the frequency of the microwave), μ_0 the free space permeability, and σ the conductivity. Substituting the value of σ for Cu in bulk ($\sigma = 59.5\times10^6$ $\Omega^{-1}m^{-1}$) [14] into eq.(1), we obtain $\delta = 1.32$ μm. This value is almost equal to the mean radius of 5Cu powder particles. As a result, in the case of small Cu powder particles, such as 5Cu powder, all Cu particles located at the surface of the compacted Cu powder layer are heated by microwave irradiation, as shown in Fig.9(a). However, in the case of large Cu powder particles, such as 100Cu powder, microwave heating occurs locally on the surface of Cu powder particles as shown in Fig.9(b) because of the large radius of curvature. This is the cause of the white spots on the surface of Cu powder (Fig.5). Thus, heat generation due to microwave irradiation decreases with the increase in particle diameter of Cu powder. Heat transfer from the surface to the interior of the compacted Cu powder layer occurs in addition to thermal emission into surrounding air. In the case of small Cu powder particles, heat conductivity is low because heat transfer occurs mainly at the contacts between Cu particles, which is a small fraction of the total volume of the compacted Cu powder layer as shown in Fig.9(a). As a result, the compaction of large amounts of Cu powder is required for heating up to a high temperature (Fig.8). On the other hand, in the case of large Cu powder particles heat conductivity is high because heat is conducted into the interior of Cu powder over the whole surface of the heating region (Fig.9(b)), resulting in high heat accumulation. Contrary to heat generation mentioned above, the heat accumulation increases with increasing particle diameter of Cu powder. The result (Fig.8) that even a small amount of Cu powder enables heating up to a high temperature is caused by this effect. The result (Fig.7) that the scatter for the peak temperature increases as the mean particle diameter of Cu powder becomes larger is due to the diameter distribution of Cu powder. Thus, the microwave heating of the compacted Cu powder vary depending on both the mean particle diameter and the compacted mass of Cu powder.

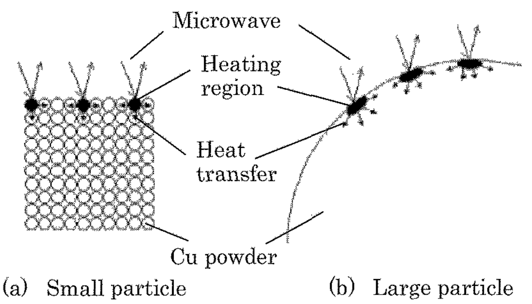

Fig. 9 Schematic illustration of microwave heating.

5. CONCLUSIONS

Cu powders with different diameter and mass were compacted on quartz substrates, followed by microwave irradiation in air (frequency of microwaves: 2.45 GHz, incident flux of microwaves:

563 W, irradiation time: 1.2 ks). The following conclusions were obtained.

(1) Microwave heating of Cu powder is quite anomalous. In the early stage of microwave irradiation, the temperature increases gradually with irradiation time. After reaching about 200 °C, abrupt temperature rise and drop occur. Then the temperature increases again and becomes almost constant in the late stage of microwave irradiation.

(2) In the case of small Cu powder particles, all Cu particles located at the surface of the compacted Cu powder layer are heated, while in the case of large Cu powder particles microwave heating occurs locally on the surface of Cu powder. The minimum mass over which the temperature rise is observed decreases with an increase in the mean particle diameter of Cu powder.

(3)The microwave heating behavior mentioned above is because practical heating area on the surface of Cu powder, the thermal emission into surrounding air and the heat transfer from the surface to the interior of the compacted Cu powder layer vary depending on both the mean particle diameter and the compacted mass of Cu powder.

REFERENCES

1)D. E. Clark and W. H. Sutton: *Mater. Sci.*, **26,** 299-331 (1996).

2)R. Roy, D. Agrawal, J. Cheng and S. Genevanishvilli: *Nature*, **399,** 668-670 (1999).

3)J. Cheng, R. Roy and D. Agrawal: *J. Mater. Sci. Lett.*, **20,** 1561-1563 (2001).

4)T. Hayashi and Y. Asano: *Report of Industrial Technology Center Gifu Prefectural Government*, No.6, 1-6 (2005).

5)S. Takayama, Y. Saito, M. Sato, T. Nagasaki, T. Muroga and Y. Ninomiya: "Proc. 9th Inter. Conf. on Microwave & RF Heating", (2003) 064m.

6)S. Yanagita: "New Application Technology of Microwave", NTS ,Tokyo (2003) pp.1-32.

7)N.Yoshikawa, E.Ishizuka and S.Taniguchi: *Mater. Trans.*, **47,** 898-902 (2006).

8) H. Sueyoshi and S. Kakiuchi: *KINZOKU*, **76,** 894-898 (2006).

9)H. Sueyoshi and S. Kakiuch : *Mater. Trans.*, **48,** 531-537 (2007).

10)S. Kakiuchi, Y. Suda, K. Obara and H. Sueyoshi: *Trans. Mater. Res. Soc. Japan*, **32,** 603-608 (2007).

11)K. I. Rybakov and V. E. Semenov: *Phys. Rev.*, **B49,** 64-68 (1994).

12) K. I. Rybakov and V. E. Semenov: *Phys. Rev.*, **B52,** 3030-3033 (1995).

13) H. Bosman, Y. Y. Lau and R. M. Gilgenbach : *Appl. Phys. Lett.*, **82,** 1353-1355 (2002).

14) Japan Inst. Metals: "A Data Book on Metals", Maruzen ,Tokyo (2007) pp.13-15.

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