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Al, Cu and W Oxide Fine Particles Produced by Discharge Explosions

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放電爆発で作られるAl, Cu, Wの酸化物微粒子

佐橋稔雄　日置義明　山田諄

抄録　空気中に張られたAl, Cu, W線にコンデンサーの放電による大電流が流れると，各線は爆発的に蒸発し，空間に酸化物微粒子ができる．作られた各微粒子は，γAl_2O_3, Cu_2O+CuO, WO_3であり，微粒子の平均の大きさは約200, 200, 100 Aであった．放電爆発法と従来のガス蒸発法とを比べると，煙球の大きさでは，前者の大きさは後者のそれの10倍以上であり，微粒子の大きさでは，前者の大きさは後者のそれの1/10以下であった．

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When a large current produced by the discharge of a charged capacitor is introduced to Al, Cu and W wires in air, it evaporates the wires explosively, and metal oxide fine particles are thus produced. The resulting fine particles are γAl_2O_3, Cu_2O and WO_3, and their average diameters are 200, 200 and 100 Å, respectively. Comparison of smoke balls produced by the discharge-explosion method (DEM) and those produced by the conventional gas-evaporation method (CGEM) shows that the size of the smoke balls produced by the DEM was ten times or more greater than that of smoke balls produced by the CGEM. In terms of the diameters of particles, the size produced by the DEM was, at most, one-tenth that produced by the CGEM.

KEYWORDS: gas evaporation, discharge explosion, oxide fine particle, smoke ball

§1. Introduction

Metal wires explosively evaporate when a large current is introduced to the wires for a short period of time by means of the discharge of a capacitor.[1,2] The evaporated metals become fine particles in air. In such experiments, the diameters of these particles are usually less than 0.1 μm if the current is large, and the particles, when viewed by the naked eye, look like smoke. However, with a smaller current the diameters of particles are larger (3 ~ 300 μm) than those observed in the case with large currents.

In this investigation, we used an electron microscope to study the growth process of particles which had been observed as smoke. According to the observation, the average diameter ranged from 100 to 200 Å, and these fine particles grew in the flash (smoke ball) due to the explosion.

§2. Experimental

Figure 1 shows a schematic diagram of the experimental system. A wire e(cm) was installed between electrodes a and b, and the cross section of the wire was S(mm^2). In the experiment, various metal wires between the electrodes were evaporated and smokelike fine particles were produced. These particles were examined later by an electron microscope. C in the figure is a 4 μF capacitor. To collect samples, several pieces of mesh were placed at h(cm) above the electrodes a and b.

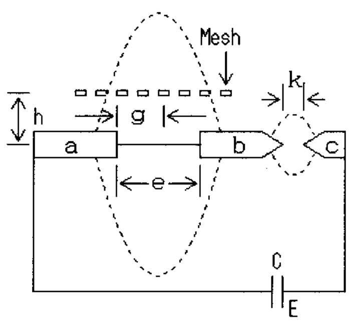

Fig. 1. Circuit diagram for discharge explosion. A metal wire of length e which will evaporate due to the explosion is stretched between a and b. The gap between b and c is a switch for this circuit, and the discharge voltage E is determined by the gap size k. C is a capacitor. Several pieces of mesh are aligned above the conducting wire at the height of h. The gap between the electrodes is denoted by e, and g represents the gap between one electrode and each mesh. The dotted lines indicate the flash region.

An arc discharge was initiated between the electrodes b and c at the voltage determined by the length k(cm) as the voltage E(kV) across the capacitor was increased (Fig. 1). When the discharge current was applied, the conducting wire between electrodes a and b melted and another arc discharge developed between a and b. The regions delineated by the dotted lines encompassing a and b, and also b and c, indicate the areas of flashes.

§3. Results

3.1 *Discharge current*

Discharge currents were measured using a Rogowshii coil. When a discharge occurred upon the use of an Al wire under the conditions $e=4$ cm, $S=0.14$ mm^2 and discharge voltage $E=12$ kV, an AC current with a period of 16 μs (63 kHz) passed through the wire. The maximum current was about 20 kA and the current gradually decreased and reached zero in 40 μs. The value of inductance was 1.6 μH and the DC resistance was 0.08 Ω.

3.2 *Melting voltage*

When the conducting wire in Fig. 1 melted, part of the wire between electrodes a and b broke and the other section remained when E was small. However, with a large E, the whole conducting wire explosively dispersed as particles. When E was increased further, a gaseous explosion took place. The minimum voltage required for melting the conducting wire is defined as the melting voltage E_0. Figure 2 shows the melting voltage for Al, Cu and W when $e=4$ cm. The abscissa represents the cross section S of the wire. From this figure, it is clear that the E_0 for each wire with the same cross section is nearly the same

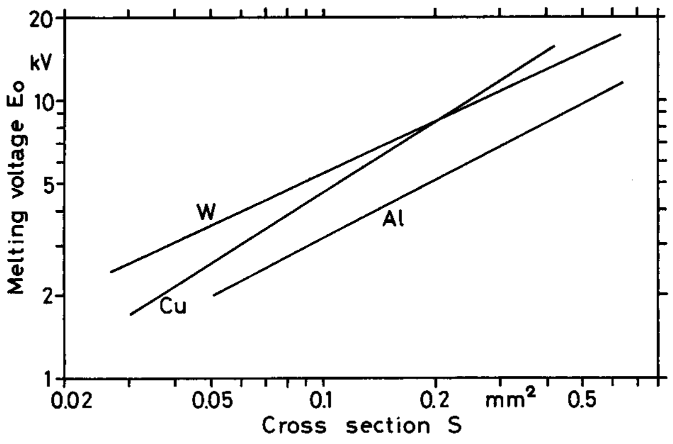

Fig. 2. Relationship between melting voltage E_0 and the cross section S of the Al, Cu and W wire. $C=4\ \mu F$ and $e=4$ cm.

for Al, Cu and W wires.

3.3 *Discharge explosion*

When the voltage E across the electrodes a and b exceeded E_0, an explosive discharge took place. Figure 3 shows the picture taken with an open shutter for the case of an Al wire. The conditions for picture (b) were $e=30$ cm, $S=0.14\ mm^2$, $E=7.5$ kV and $E_0=6.5$ kV; E was relatively close to E_0. The picture shows that metal particles spread from several locations between the electrodes. In the experiments, many Al particles about 3 μm to 300 μm in size were observed. Under (a) conditions, $e=4$ cm, $S=0.14\ mm^2$, $E=12$ kV and $E_0=4$ kV; E was considerably larger than E_0. In this experiment, the metal exploded in a gaslike manner. After the explosion, a thin smokelike region was observed above the area of the explosion.

In another experiment, several Al wires were stretched over electrodes a and b parallel to the current-conducting Al wire at varying heights from the current-conducting wire as shown in Fig. 4, and discharge was performed under the same conditions as in the experiment whose results are shown in Fig. 3(a). In this experiment, the region to which black soot adhered coincided with the region of flash discharge in Fig. 3(a).

3.4 *Aluminum oxide particles*

In this experiment, the discharge conditions were the same as those in the experiment in Fig. 3(a) and $h=4.5$ cm and $g=2$ cm. Again, the wire between a and b was Al. Figure 5(a) shows the magnified images of the particles and Fig. 5(b) shows the diffraction patterns of the particles.

According to Fig. 5(a), most of the fine particles are spherical, and Fig. 5(b) indicates that these fine particles consist of the γAl_2O_3[3] (cubic). The distribution of particle size was studied by varying h, and the results are shown in Fig. 6(a). The vertical axis represents the relative number of particles. If the mean diameter of particles D is defined as that producing the peak value in the distribution, the relationship between D and h can be shown by the graph (1) in Fig. 6(b). The flash region was $h=0\sim8$ cm, and according to Fig. 6, the particle growth stopped at about the center ($h=4$ cm). In the direction along a-b, there were no major changes in diffraction patterns. Although the distribution of the diameters of par-

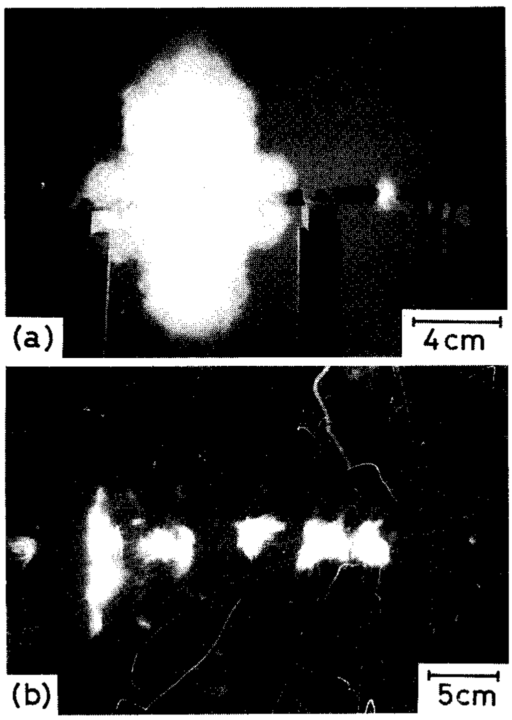

Fig. 3. Photographs taken with an open shutter at the time of explosion. Al wire was used. (a) $C=4\ \mu F$, $e=4$ cm, $S=0.14\ mm^2$, $E=12$ kV and $E_0=4$ kV. (b) $C=4\ \mu F$, $e=30$ cm, $S=0.14\ mm^2$, $E=7.5$ kV and $E_0=6.5$ kV.

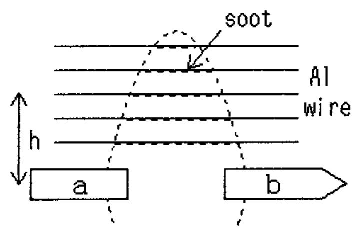

Fig. 4. Soot adhering to Al wires.

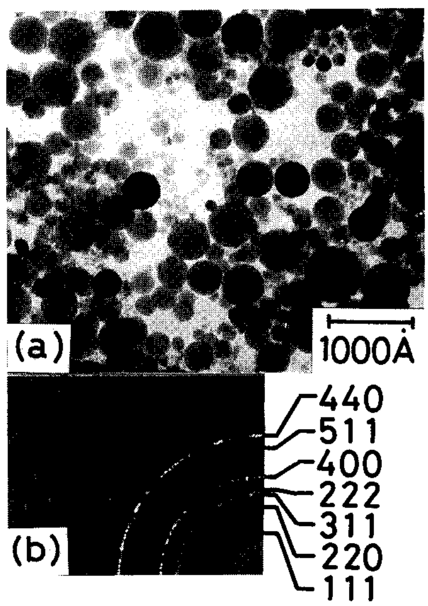

Fig. 5. (a) A magnified image of Al oxide fine particles and (b) diffraction pattern. $C=4\ \mu F$, $e=4$ cm, $S=0.14\ mm^2$, $E=12$ kV and $E_0=4$ kV.

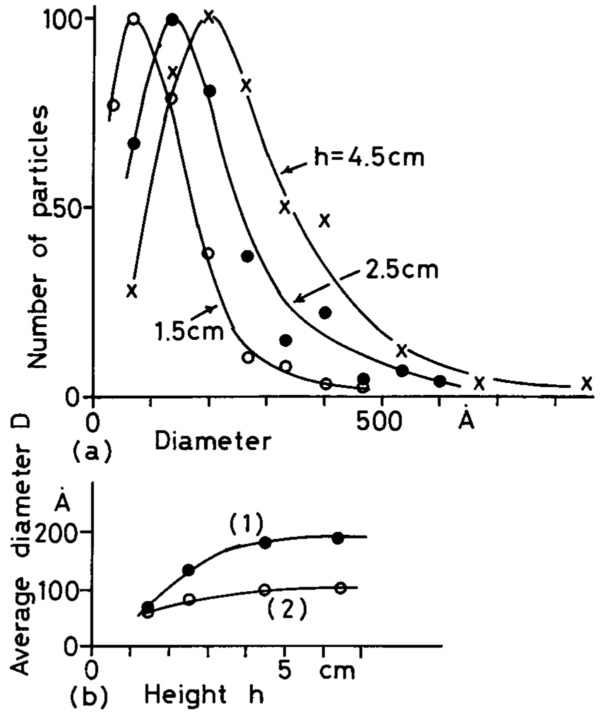

Fig. 6. (a) Distribution of Al oxide fine particles. The peak value is designated as the mean diameter D. (b) Relationship between D and h. (1): Al, $C=4\ \mu$F, $e=4$ cm, $S=0.14$ mm^2 and $E=12$ kV. (2): W, $C=4\ \mu$F, $e=4$ cm, $S=0.07$ mm^2 and $E=12$ kV.

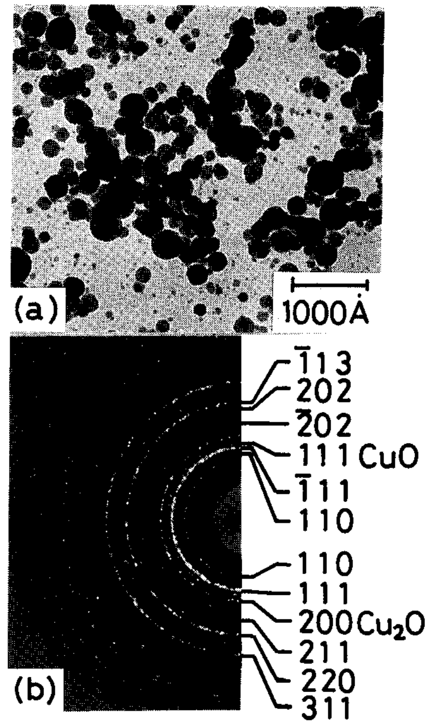

Fig. 7. (a) A magnified image of Cu oxide fine particles and (b) a diffraction pattern. $C=4\ \mu$F, $e=4$ cm, $S=0.08$ mm^2, $E=12$ kV and $E_0=3.5$ kV.

ticles did not change along the wire in terms of magnified images, the number of particles decreased toward the ends of the flash. Particles were scarcely observed outside the flash region. Since the region where particles were observed and the region of black soot mentioned in the preceding section coincided approximately, the soot is considered to consist of γAl_2O_3.

With conditions such that $e=4$ cm and $S=0.14$ mm^2 and discharge voltage $E=6$ kV, no fine particles were observed. At this time, the kind of discharge shown in Fig. 3(b) is considered to occur. In addition, for the discharge voltage range 9 ~ 15 kV, the magnified images and diffraction patterns are very similar to those results obtained from the experiment shown in Fig. 3(a).

3.5 *Oxide particles of Cu and W*

The experimental conditions for Cu experiments were $e=4$ cm, $S=0.08$ mm^2, $E=12$ kV and $E_0=3.5$ kV. In the experiment with $h=1.5$ cm, only diffraction rings of the Cu_2O (cubic) were observed.[4] However, as h increased, CuO (monoclinic) diffraction rings appeared and these rings due to the CuO increased.[5] Figures 7(a) and 7(b) show a magnification image and a diffraction pattern, respectively, for $h=4.5$ cm and $g=3$ cm. The exterior and the distribution are similar to those in Fig. 5(a), and the tendency of change of mean diameter with respect to h is similar to the tendency in Fig. 6(b-1). The color of soot resulting from the Cu wire evaporation was greenish black.

The experimental conditions for the experiment using W were $e=4$ cm, $S=0.07$ mm^2, $E=12$ kV and $E_0=4.5$ kV. As the diffraction patterns, diffraction rings of WO_3(cubic) were observed.[6] In terms of magnification images, the exterior was spherical, as in the case with Al and Cu; however, the size was approximately one half of those by Al and Cu. Figure 8 shows a magnified image and diffraction pattern for $h=2.5$ cm and $g=3$ cm. The change of D with respect to h is shown in Fig. 6(b-2). The color of the soot caused by the evaporation of the W wire was black.

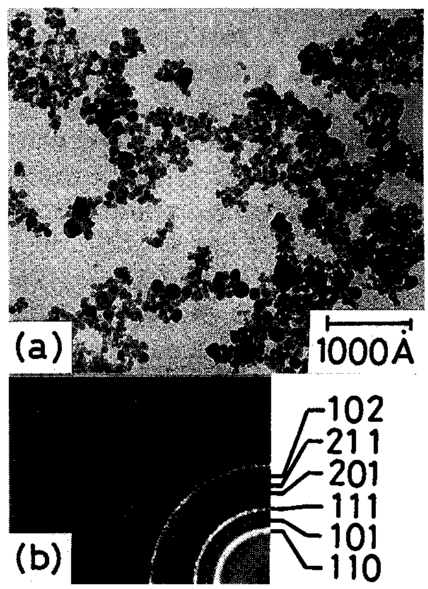

Fig. 8. (a) A magnified image of W oxide fine particles and (b) a diffractlon pattern. $C=4\ \mu$F, $e=4$ cm, $S=0.08$ mm^2, $E=12$ kV and $E_0=4.5$ kV.

§4. Discussion

4.1 *Evaporation energy of metal wires*

The weight of the Al wire in the experiment of Fig. 5 was 15 mg and the evaporation energy E_e was 200 J. The required electrostatic energy E_c was calculated to be $E_c=300$ J because $C=4\ \mu F$ and $E=12$ kV. This means that the major portion of electrostatic energy E_c was used for the evaporation of the wire.

4.2 *Smoke balls*

Wada[7] observed generation of smoke balls by means of the evaporation method. Yatsuya *et al.*[8] studied the internal structure of smoke balls which grew above the evaporation site. According to their study, there were two zones in smoke balls, an inner zone and an outer zone; particles grew in the upward-extending region of each zone. Furthermore, the exterior and the diameter of particles in each zone were different. Nishida and Kimoto[9] and Kaito[10] investigated the constituent of smoke balls in the experiments using Te, and Mo and W, respectively.

The flash region between electrodes a and b in Fig. 3(a) is considered to correspond to the smoke-ball region. In the sense that "fine particles are included in the smoke ball and grow in the smoke", the present results and those obtained by Yatsuya *et al.*,[8] Nishida and Kimoto[9] and Kaito[10] are in agreement. However, in their experiments the smoke balls were asymmetrical in the horizontal direction and extended upward from the evaporation source, unlike those shown in Fig. 3(a). In the current experiment, there were no differences in the inner and outer zones in the smoke; also, no differences in the particle features were observed.

4.3 *Size of the smoke ball*

Figure 9 shows an example of the relationship between the diameter of the smoke ball and Ar gas pressure in the bell jar when Cu was evaporated. The gas pressure ranged from 20 Torr to 400 Torr. The evaporation was carried out by flowing electric current through a W wire. The size of the ball depended on the type of the gas, the evaporation metal and the evaporation temperature. According to an estimation based on Fig. 9, the size of the smoke ball under 1 at. with Cu wire is less than 1 cm. The smoke ball in Fig. 3(a) was produced at 1 at. and the size was larger than 10 cm. The size of the smoke ball increased with the diameter of the wire and the discharge voltage. If the diameter and the discharge voltage were similar, the sizes of the smoke of Al, Cu and W were essentially the same.

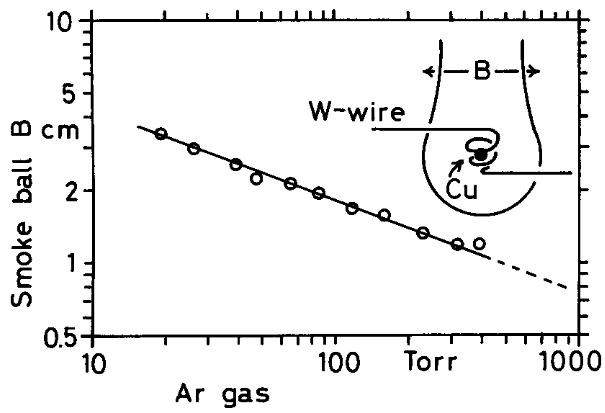

Fig. 9. Relationship between the size of the smoke ball B and Ar gas pressure.

The fact that the size of the smoke ball grew as described in this paper can be explained by the fact that the heat energy for explosion was generated within a short period of time and energy per unit time was great. In the experiment of Fig. 9, the energy provided to a W wire when a 28 mg Cu wire was evaporated was 300 W within 10 s. In the experiment of Fig. 7, the energy provided to the circuit by the capacitor was 300 J/30 μs. The energy per unit time was 300 J/s in the case of Fig. 9 and 1×10^7 J/s in the case of Fig. 7.

4.4 *Particle size*

The particle size decreased as discharge energy increased, and this tendency is in agreement with that of Suhara *et al.*[1] Suhara *et al.* measured the diameter of W by means of the precipitation method when the explosion energy was relatively small in comparison with the size of the metal wire used. The results show that there are two peaks, one at 3 μm and another 0.3 μm, in the distribution. We studied the distribution of particles for experiments with large energy using an electron microscope; the results indicated that a peak formed between 100 Å and 200 Å.

When a current flowed through a W wire in air at 1 atmosphere, the heated W wire became red, and white smoke was produced. The smoke consisted of particles of WO_3 crystals[6] with clear crystal surfaces, ranging in size from 0.1 μm to 0.3 μm. Explosion experiments were also conducted under 1 atmosphere. However, the size of WO_3 in this explosion was small, at most one-tenth of that in the above experiment.

The fact that the size of the particle becomes small can be explained by the fact that the energy per unit time is great, the smoke ball is big and the density of the evaporation metal in air is small.

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