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Effects of Atmospheric Gas Pressure on Particle Size Distribution of Nanosized Powders Synthesized by Pulsed Wire Discharge

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In general, the particles of diameter less than 100nm are referred to as nanosize powders. The typical average particle size of the powders synthesized by pulsed wire discharge is several tens of nanometers. However, a large number of micrometer-sized particles over 100 nm in diameter have also been observed. In this research, copper nanosize powders were synthesized by pulsed wire discharge at 100 Torr(1.33×10^4 Pa) and 800 Torr (1.07×10^5 Pa)of nitrogen gas, and the particle size distributions of the powders were evaluated by SEM observations. The results showed that the particle diameters of peak counts were 30 nm and 45 nm at pressures of 100 Torr and 800 Torr, respectively. However, the number of micrometer-sized particles was fewer at 800 Torr than at 100 Torr. The energy deposited in the wire during ohmic heating process was 38 J at 100 Torr, and 52 J at 800 Torr. Even though discharge energy was 135 J that is two times larger energy to be required for vaporizing the wire, the energy deposited at both pressures were lower than vaporization energy. From these experimental results, we conclude that the micrometer-sized particles were generated due to the incomplete vaporization of the wire.

Key words: nanosize powder, pulsed wire discharge, particle size distribution, micrometer-sized particles, energy deposited

1. INTRODUCTION

Pulsed wire discharge (PWD) is a very simple method of producing various kinds of nanosize powders, where a thin wire is exploded by a high-voltage high-current pulse.[1] Many kinds of nanosize powders have been synthesized by the PWD.[1-6] It has been considered that nanosize powder synthesis by pulsed wire discharge is a promising and challenging method because of its high energy-conversion efficiency and simple process.

In general, it has been known that average particle sizes of the nanosize powders synthesized by PWD increased as the atmospheric gas pressure increased.[3-5] It has been considered that metallic vapor concentration is higher at higher pressure than that at lower pressure.

Particles with diameter between 100 nm to a few micrometers in the powders synthesized by PWD have been also observed.[1, 2, 4] We describe the particles over 100nm as micrometer-sized particles. The origin of the micrometer-sized particles may be attributed to the micrometer-sized liquid droplets that were generated due to the incomplete vaporization of the wire. Thus, it is expected that if the energy deposited into the wire increase, the number of micrometer-sized particles significantly decrease. Increase in the energy deposited is limited by plasma formation in surroundings of the wire through Paschen breakdown.[7] The Paschen breakdown depends on the atmospheric gas pressure. In general, the Paschen breakdown takes place more easily at lower pressure than at higher pressure.

In this research, we investigated the effects of the atmospheric gas pressure on the size distributions and on the number of micrometer-sized particles of the copper powders synthesized by PWD at 100 Torr and 800 Torr of nitrogen as an atmospheric gas.

2.EXPERIMENTAL SETUP AND CONDITIONS

Figure 1 shows the schematic of the experimental setup. Six pieces of copper wire are installed on the wire holder that was made with an insulator disc and stainless steel screws. Both ends of each wire are contacted with two copper electrodes that were connected to the capacitor bank through the spark gap switch and high voltage feed-through.

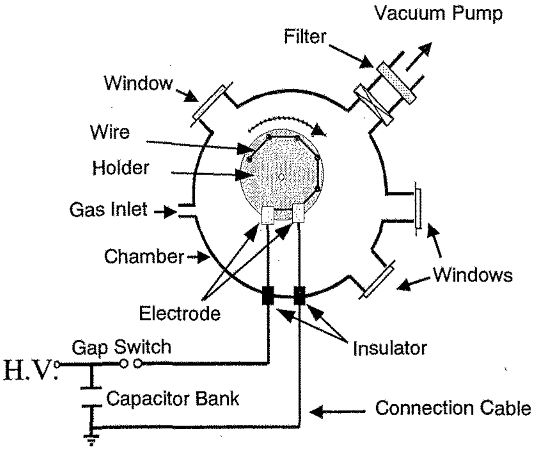

Fig. 1 Experimental setup.

This apparatus enables us to explode six pieces of wires by rotating the wire holder without opening the chamber. A membrane filter was installed between the chamber and a vacuum pump, and the chamber was vacuated by the rotary pump. After evacuation, the chamber was filled up with atmospheric gas of N_2.

Electrical energy was stored in capacitors and released through a gap switch to the wire installed between the electrodes. The electrical energy was deposited in the wire because of its relatively high resistance. Thus, only the wire between the electrodes was heated, vaporized, and turned into plasma. The powders were collected on the surface of the filter by evacuating the chamber through a filter.

Experimental conditions are shown in Table I. Copper wires 0.25mm in diameter, and 25mm in length were evaporated in the atmospheric gas of N_2. The capacitance and voltage were respectively 10μF, 5.2kV, which was two times larger energy stored for vaporizing the wire.

Discharge current and voltage drop across the wire were measured, and energy deposited into the wire was calculated by integrating the electrical power.

Phases in the synthesized powders were identified by X-ray diffraction using Cu-K_α radiation (0.15406 nm). Particle size distribution was evaluated by measuring the diameter and by counting the number of particles with a scanning electron microscopy (SEM).

Table I. Experimental conditions.

Wire	Copper ϕ0.25 ×25 mm
Atmospheric gas	N_2 : 100 Torr (1.33×10^4 Pa)
	800 Torr (1.07×10^5 Pa)
Capacitance	10μF
Inductance	0.8μH
Discharge energy	135 J

3. EXPERIMETAL RESULTS AND DISCUSSIONS

Figure 2 shows the X-ray diffraction patterns for the Cu powders synthesized at 800 Torr. We see that all the peaks are in good agreement with JCPDS data of Cu.

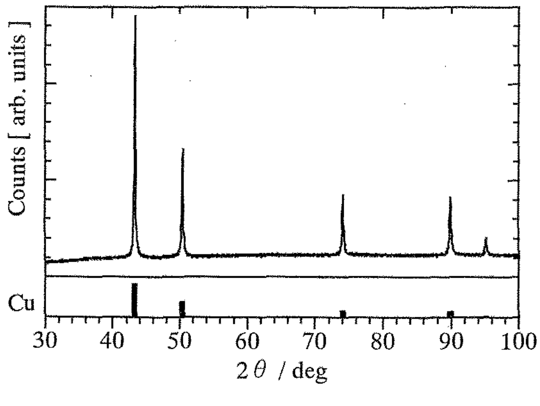

Fig. 2 X-ray diffraction pattern of the powder.

Figure 3 shows the SEM image of the powder synthesized at 800 Torr. Particles seem to be almost spherical. It is seen that there are a few particles of relatively larger diameter over 100 nm.

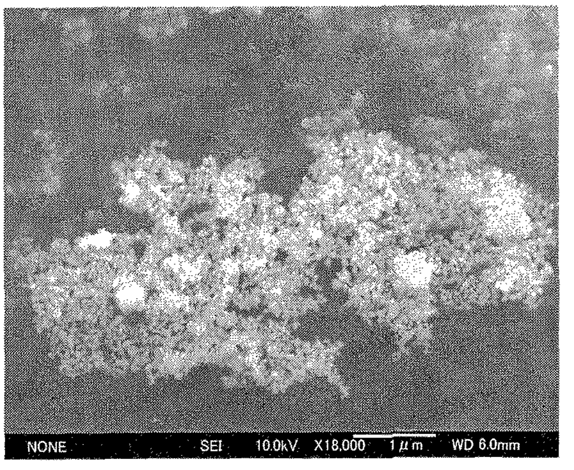

Fig. 3 SEM image of the powder (800 Torr).

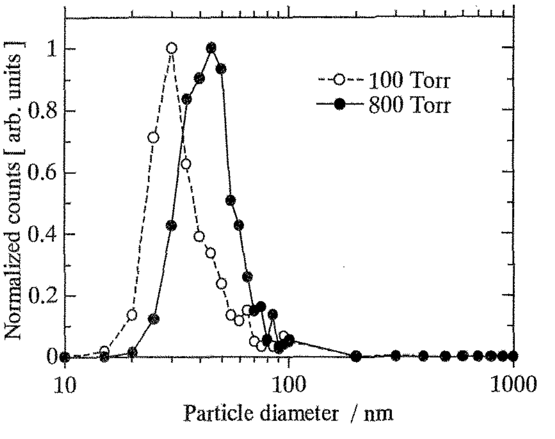

Fig. 4 Particle size distributions.

Figure 4 shows the particle size distributions of the powders synthesized at 100 Torr and 800 Torr. The counts were normalized by the total observed counts. Sizes of maximum count were 30 nm and 45 nm at 100 Torr and 800 Torr, respectively. This result indicates that the average particle size increased as atmospheric gas pressure increased, which is in good agreement with the results of the previous researches.[3-5]

The micrometer-sized particles were frequently observed as shown in Fig. 3, even though the observed counts were very small compared to the counts of nanosized particles. The number of micrometer-sized particles is shown in Fig. 5, where the number of micrometer-sized particles was normalized with the observation area. The number of micrometer-sized particles in the powder synthesized at 800 Torr significantly decreased compared to the synthesized at 100 Torr.

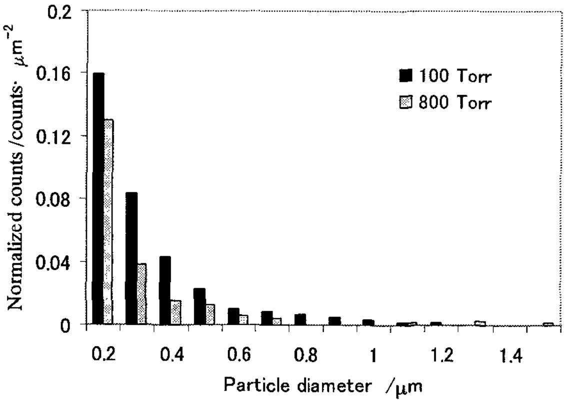

Fig. 5 Number micrometer-sized particles normalized by observation area.

Waveforms of discharge current and voltage drop across the wire are shown in Fig. 6 and Fig. 7 The most noticeable feature in the current and voltage waveforms is the sudden drop of current and sudden rise of voltage. The amplitudes of the dropping current and the rising voltage depended on the atmospheric gas pressure, and were higher at 800 Torr than at 100 Torr. The current drop arose from the increase in resistance of the wire due to the wire vaporization, and the voltage rise arose from current flow in the high resistive state of the wire. In this high resistive state, a driving force of current flowing is the inductive energy stored in the circuit inductance.

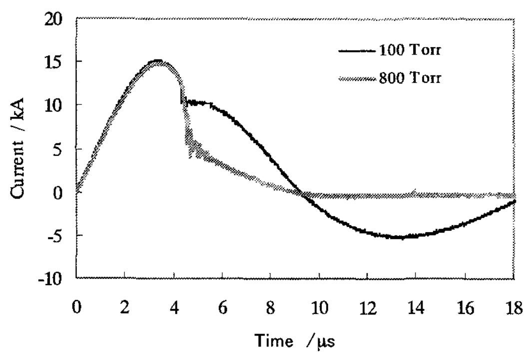

Fig. 6 Current waveforms.

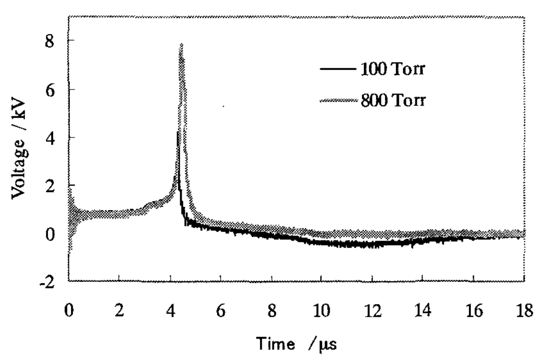

Fig. 7 Voltage waveforms.

It is considered that the Paschen breakdown occurred at the time of the peak voltage, and then plasma was formed in the surroundings of the wire material. After the breakdown, the current smoothly damped because of the low resistivity of the plasma. The waveforms of current and voltage at the different pressures were almost the same until the breakdown occurred. A noticeable feature in the voltage waveforms in Fig. 7 is the small difference of their peak times. The times of voltage peaks were 4.34 μsec and 4.46 μsec at 100 Torr and 800 Torr, respectively. The breakdown was delayed about 120 ns at 800 Torr compared to at 100 Torr. At low pressure, the plasma formation is easier than at high pressure because the Paschen breakdown voltage is lower than that of high pressure according to the law of Paschen breakdown.

The energy deposited into the wire can be calculated by time integration of the power dissipated in the wire using the following equation.

$$E_d(t) = \int_0^t v_w(\tau) \times i(\tau) d\tau , \quad (1)$$

where $E_d(t)$, $v_w(t)$ and $i(t)$ are respectively the energy deposited in the wire, voltage drop across the wire, and current through the wire at time t.

Figure 8 shows the energy deposited in time. It is clear that most energy has been deposited before the breakdown even though large amounts of current continuously flowed after the breakdown. Thus, it was confirmed that the energy deposited is limited by plasma formation in the surroundings of the wire material. The energy required to vaporize the whole wire is 67 J. However, the energy deposited, E_d, did not reach the vaporization energy in both pressures as shown in Fig. 8. The energy deposited before breakdown was higher at 800 Torr than at 100 Torr. The delay of the breakdown about 120 ns at 800 Torr resulted in a meaningful increase in the energy deposited. Thus, it is expected that the larger part of the wire turned into the vapor at 800 Torr than at 100 Torr, and the number of micrometer-sized particles decreased.

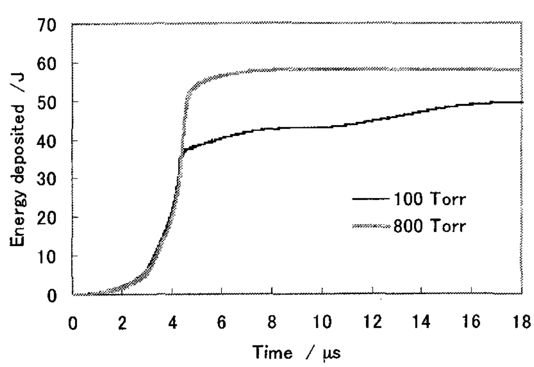

Fig. 8 Energy deposited into the wire.

4. CONCLUDING REMARKS

Copper nanosize powders were successfully synthesized in the 100 Torr and 800 Torr atmospheric gas pressure of nitrogen by pulsed wire discharge. The particle diameters of peak counts were 30 nm and 45 nm at the pressures of 100 Torr and 800 Torr, respectively. These results confirmed that average particle size increased as the atmospheric gas pressure increased. The micrometer-sized particles were frequently observed at both pressures. The amount of micrometer-sized particles was smaller at 800 Torr than that of 100 Torr.

Even though discharge energy was 135J that is twice large for vaporization of the whole wire, the energy deposited was lower than vaporization energy at both pressures. The energy deposited into the wire before the breakdown was 38 J at 100 Torr, and 52 J at 800 Torr. Thus, it is considered that the larger part of the wire turned into the vapor at higher pressure than at low pressure, and the number of micrometer-sized particles that originated from liquid droplets decreased.

For the purpose of decreasing the amounts of the micrometer-sized particles in the powders synthesized by PWD, increasing the atmospheric pressure is a useful method, although the average particle size increases.

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