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Paper:

Copper/Silver Recovery from Photovoltaic Panel Sheet by Electrical Dismantling Method

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The volume of spent photovoltaic (PV) panels is expected to grow exponentially in future decades. Substantial material resources such as silver (Ag), copper (Cu), aluminum (Al), silicon (Si), and glass can potentially be recovered from silicon-based PV panels. In this paper, we targeted the recovery of Cu and Ag from a cell sheet separated to a glass panel from a spent PV panel. The technical feasibility of a novel electrical dismantling method was experimentally studied. This method employed a pulsed power technology that releases high energy in a short time. It allowed a selective separation of the Cu/Ag wires from the sheet once per discharge in water. The experimental results indicated that 95.6% of the total Cu and 17.2% of the total Ag in the sample were successfully separated from the cell sheet using a 3.5-kJ capacitor bank circuit. Moreover, 3.66% of the total Si in the sample was contaminated by the separated Cu/Ag particles from the cell sheet, mainly by shockwaves generated by plasma expansion, and some of them formed a compound with Cu and Ag by eutectic melting, resulting in low liberation. At the lower energy of 3.5 kJ, eutectic melting of Cu and Ag with Si was more suppressed than 4.6 kJ, and 94.3% of Cu and 77.5% of Ag in the separated particles were liberated, which would be acceptable for further wet gravity and/or shape separation of Cu and Ag.

Keywords: recycling, physical separation, metal recovery, pulsed discharge, solar power

1. Introduction

Photovoltaic (PV) technology is strongly expected to provide a fundamental contribution to the transition from conventional fossil fuels to renewable energy-based economies [1, 2]. In the last 20 years, the application of PV has rapidly increased [3]. A dramatic increase in the annual flux of end-of-life PV panels could be expected by 2025 because PV installations started in the late 1990s, and the average lifetime of a PV panel is approximately 25 years [4].

Recycling technologies involve physical separation followed by chemical separation. In general, physical separation is in low energy but its separation efficiency is not very high. The opposite is true in chemical separation. Therefore, the best combination of the two is necessary to achieve a low-energy recycling process with high separation efficiency [5–7]. Additionally, technological innovation is required to improve physical separation efficiency [8–11] and reduce cost/energy in chemical separation [12–15].

Based on this background, several recycling processes have been proposed for recycling PV panels. PV recycling technologies can be divided into two types. After initial mechanical separation via aluminum flame, one type comprises shredders and physical separations for mass processing, and the other carefully separates glass panels and cell sheets mechanically or thermally for advanced reuse/recycling of glass [16]. As approximately 60% of silicon-based PV panels are glass, it is desirable to establish the latter recycling technology to reuse/recycle materials at a high rate. To accomplish advanced recycling, the construction of the glass supply chain and cost/energy-effective recycling technology are essential. Separation of metals and resin in the cell sheet after removal from the glass panel is especially a technical issue to recover copper (Cu) and silver (Ag) in the advanced recycling process.

The hot-knife method is a representative method to separate the glass panel and the cell sheet. The cell sheet comprises Cu and Ag wires and silicon (Si) cells embedded in the ethylene-vinyl acetate (EVA) copolymer resin sheet (**Fig. 1**). As the resin causes coloring of sulfuric acid as a by-product, EVA should be separated before recovering Cu and Ag in copper smelting. However, conventional physical separation methods such as grinding are difficult owing to the hard wire shape of Cu.

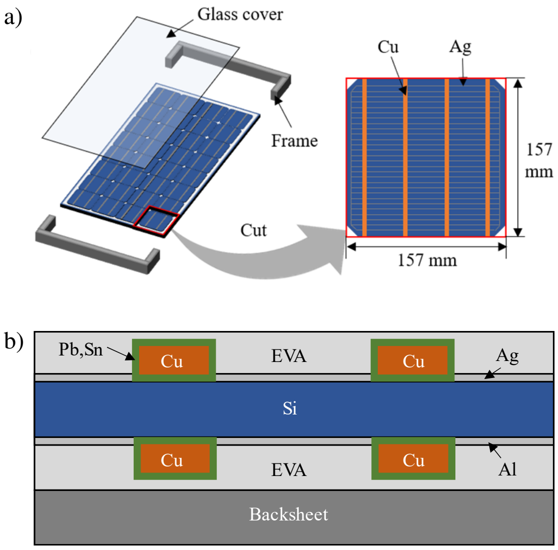

Fig. 1. Schematic of the PV panel. (a) Preparation of the sample; (b) cross-section of the cell sheet.

Table 1. Chemical composition of the sample [wt%].

Cu	Ag	Si	Al	Sn	Pb	etc.	I.L.
3.45	0.12	18.5	2.26	0.30	0.25	3.62	71.5

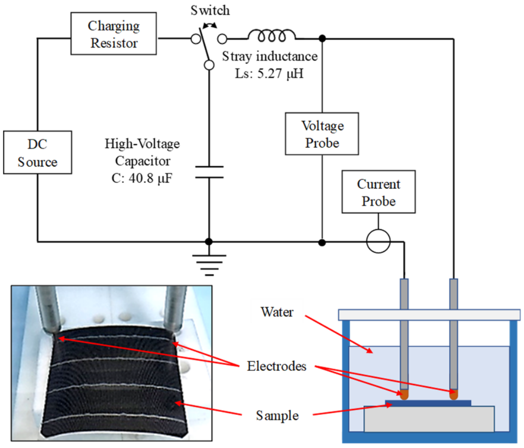

Fig. 2. Schematic of the experimental setup of electrical dismantlement.

Recently, as a new fragmenting technology, electrohydraulic fragmentation (EHF) has emerged, offering new liberation possibilities for higher separation efficiency than conventional mechanical methods [17, 18]. EHF uses intensive shockwave impulses of discharge plasma generated along with the interface of different materials. However, because these EHF methods use non-contact underwater discharge, the shockwave due to plasma generation in water contributes more to grinding than in target materials. As a result, it is difficult to achieve high selective separation with a small number of discharges by EHF.

In this paper, the technical feasibility of a novel electrical dismantling method using electrical wire explosion by contacting the electrode directly with cell sheet is experimentally studied. This method uses pulsed power technology, which emits high energy in a short time and can selectively separate the Cu wire with Ag from the cell sheet once per discharge in water. Unlike EHF, the direct installation of the electrode on the target materials is expected to allow highly selective liberation by using the electrical characteristics of the target directly. Separated Cu and Ag particles were quantitatively evaluated in terms of recovery and liberation and discussed for use as raw materials for copper smelting and/or Cu wire directly.

2. Materials and Methods

2.1. PV Cell Sheet Sample

Experiments were performed with a crystalline silicon PV module (Shanghai JA Solar Technology, JAM6(K)-60-290/PR). Samples of cell sheets were obtained after the hot-knife separation of external aluminum frames and glass (**Fig. 1(a)**). Each sample was taken by cutting a piece of approximately 157 × 157 mm, which was equivalent to one cell module. A cross-sectional view of the sample is shown in **Fig. 1(b)**.

The chemical composition of the sample was semi-quantitatively analyzed by X-ray fluorescence (XRF, Rigaku Corporation ZSX PrimusIII+) analysis (**Table 1**). In XRF analysis, the sample was sufficiently ground to fine particles by agate mortar, placed into the 10 mm of the sample holder, and measured by a Rh 4.0 kW X-ray tube in a vacuum. All sampling and analysis were conducted in triplicate and confirmed within a 5% error. The sample contained 3.45% of Cu, 0.12% of Ag, and 71.5% of ignition loss (I.L.) owing to EVA and backsheet which is a multi-layer film comprising polyvinyl fluoride (PVF), polyethylene terephthalate (PET), polyethylene (PE), and so on.

2.2. Experimental Methods

The schematic of the experimental setup is shown in **Fig. 2**. The copper-tungsten electrodes were placed on both ends of the Cu wire of the sample. The distance between the electrodes was 140 mm. The electrode was placed in direct contact with the Cu wire by fixing the electrode and the sample firmly with a jig. The volume of the water tank was 200 dm^3, and 140 dm^3 was filled with tap water to sufficiently submerge the sample. As the pulsed power source, a simple capacitor bank circuit was used. A mechanical relay switch was used to discharge the stored capacitive energy. Once the switch was changed over from the charging side to the load side, the stored energy was discharged into the Cu wire. The pulsed discharge current caused the explosion of the electrical Cu wire.

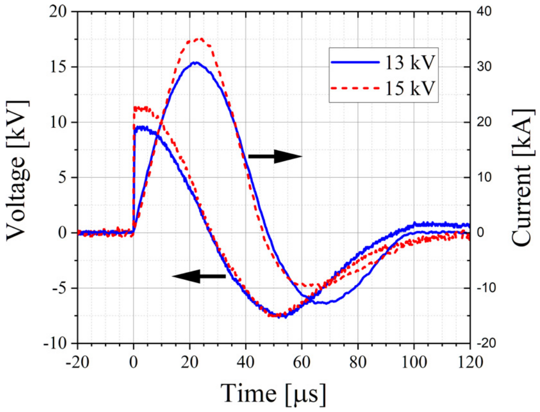

Fig. 3. Voltage and current waveforms of pulsed discharge for the cell sheet.

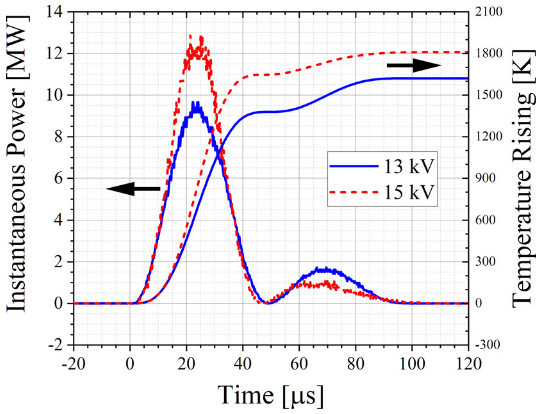

Fig. 4. Instantaneous power and temperature rising during pulsed discharge.

For the simple RLC circuit, appropriate voltage V, charged to capacitance C (40.8 μF), was given by

$$V = \frac{2^{\frac{3}{4}} L_s{}^{\frac{1}{4}} K^{\frac{1}{2}} A}{C^{\frac{3}{4}}} \quad . \; . \; . \; . \; . \; . \; . \; . \; . \; . \; . \; . \; . \quad (1)$$

where L_s is stray inductance (5.27 μH), A is a cross-section of Cu wire (0.34 mm^2), and K is a constant describing the mass density scaled integral of the resistivity of the material from the initial temperature to the onset of vaporization (5.9×10^{16}) [19]. For this condition, the specific energy accumulated at the time of the peak of the sinusoidal circuit current was equal to that required to bring the wire to the vaporization temperature. According to Eq. (1), the theoretical optimum voltage for the Cu wire explosion was calculated to be 13 kV. Therefore, the experiments were carried out at voltages of 13, and 15 kV in this study. These conditions correspond to 3.5 and 4.6 kJ, respectively.

Four pulsed discharge experiments were conducted on each sample, i.e., one for each Cu wires, as shown in **Fig. 1(a)**. After the pulsed discharge experiment, the water in the tank was filtered and filtrates were analyzed as separated particles and sheets. The filtrates were sieved, and the Cu, Ag, and Si concentration in each size fraction was analyzed by ICP-OES (Thermo Fisher Scientific iCAP6000) after aqua regia leaching. The liberation of Cu and Ag in each size fraction was also observed by scanning electron microscopy (SEM, Hitachi S-4500S) and analyzed by a mineral liberation analyzer (MLA, FEI Quanta Feg). For the MLA, the samples were set into a mold with epoxy resin to form a hardened block. Then the block was ground down to expose a representative cross-section of particles which was subsequently polished before the measurement.

3. Results and Discussion

3.1. Observation of Electrical Wire Explosion

The discharge current peak values were 30.8 kA and 35.2 kA at 13 kV and 15 kV, respectively (**Fig. 3**). There was no sudden current drop during the current rise as found in a typical electrical explosion [20–22]. The cause of the current drop was the increase in the resistance between the electrodes due to the vaporization of the wire. However, in this study, because the current rise time was relatively long, plasma may have been partially generated, and the current drop did not occur. However, after approximately 47 μs when the current passed through zero, the voltage-current waveform changed significantly, possibly because the applied reverse voltage (approximately -7.5 kV) when the current to the Cu wire reached zero was not enough to maintain the wire explosion. Therefore, the total circuit resistance increased by the narrow plasma channel.

The electrical power calculated from the measured Cu wire resistance and measured current and the temperature change in the Cu wire by Joule heating (Eq. (2)) are shown in **Fig. 4**.

$$T = \frac{\int_0^t I^2 R dt}{CM} \quad . \; . \; . \; . \; . \; . \; . \; . \; . \; . \; . \; . \; . \quad (2)$$

where T is the temperature rise, I is the current, R is the resistance of the Cu wire, C is the heat capacity of the Cu wire (3.9×10^{-4} J kg^{-1} K^{-1}), M is the mass of the Cu wire (4.1×10^{-4} kg) and t is time. The resistance R measured by the multimeter (PSM1735, Newtons4th Ltd.) was 9.9 mΩ at 10 kHz, which was slightly higher than the ideal Cu wire resistance of 8.5 mΩ, which should have increased by bending and lifting.

Neglecting heat loss by the surrounding water, the melting point of Cu (1358 K) was attained at about 31 μs and 27 μs at 13 kV and 15 kV, respectively. The total temperature did not exceed the boiling point of Cu (2835 K) when the local plasma generation was ignored.

Photographs of the sample after the pulsed discharge are shown in **Fig. 5**. The amount of Cu wire remaining was 4% and 0% of the original sample at 13 kV and 15 kV, respectively. Higher charging energy resulted in a higher Cu separation rate and more damage to both sides of the sample. Because the damage was observed mainly around

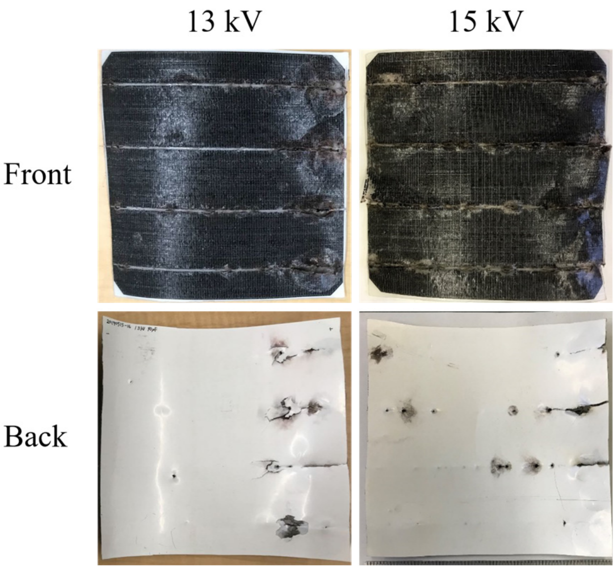

Fig. 5. Photographs of the sample cell sheet after pulsed discharge.

Table 2. Chemical composition of recovered particles [wt%].

	Cu	Ag	Si	etc.	I.L.
13 kV	52.1	1.59	39.4	7.55	N.D.
15 kV	49.0	1.26	43.5	6.21	N.D.

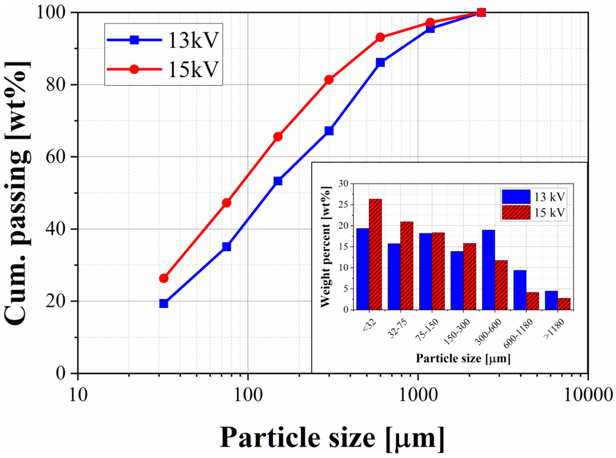

Fig. 6. Particle distribution of recovered particles.

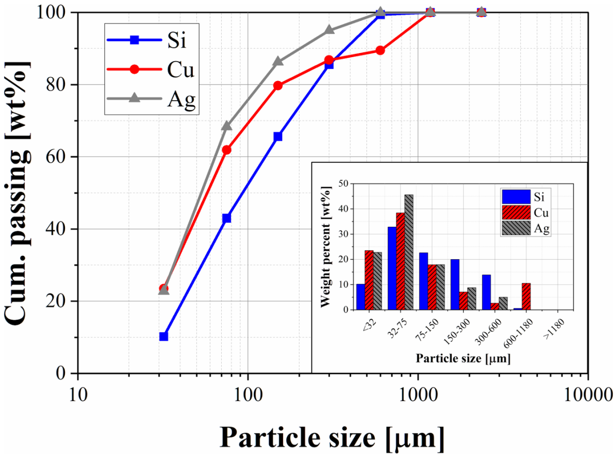

Fig. 7. Size distribution of Cu, Ag, and Si at 13 kV.

the electrodes and along the Cu wire, it appeared to be due to the generation of hot plasma and subsequent shock-waves. During discharge, dielectric breakdown occurred at the EVA layer between the electrode and Cu wire, and hot plasma should have been generated there. The temperature increased explosively and eventually produced hot plasma at the bent part of the Cu wire. Typically, the bent part of metallic wire has a higher resistance than the straight part owing to microcracks. Because the metals had inherent properties in which resistance increased with increasing temperature, the slightly higher resistance of the bent part spatially concentrated the energy consumption.

3.2. Composition and Particle Distribution of Recovered Particles

After the pulsed discharge, the particles and sheet could be easily separated depending on the difference in size. The chemical composition of the recovered particles is shown in **Table 2**.

A total of 95.6% of Cu and 17.2% of Ag with 3.66% of Si were separated at 13 kV, and 96.9% of Cu and 20.3% of Ag with 7.02% Si were separated at 15 kV. In these recovery calculations, each concentration of Cu, Ag, and Si in the original sample was used as a basis. As shown in **Table 2**, the Cu concentration of the recovered particles was sufficient to serve as a raw material for a copper smelter. Good Cu separation was achieved, but further recovery of Ag was a challenge for the near future. For example, conventional grinding and physical separation may be feasible for further Ag recovery because no more Cu wire exists in the cell sheet after the pulsed discharge.

To discuss the further separation between Cu/Ag and Si from the recovered particles, the particle distribution (**Fig. 6**) and concentration of Cu, Ag, and Si in each size fraction at 13 kV (**Fig. 7**) were analyzed.

By comparing the results of 13 kV to 15 kV in **Fig. 6**, 80% particle size was 137 μm at 13 kV and 118 μm at 15 kV. In the case of 15 kV, the weight percent tended to decrease in larger particle groups, but in the case of 13 kV, it was relatively evenly up to 600 μm. Typically, a wet physical separation is feasible for particles of this size.

As shown in **Fig. 7**, Ag-containing particles were mostly fine in texture, whereas Si-containing particles were larger. However, Cu-containing particles were distributed in both smaller and larger size fractions than Ag and Si. However, because no remarkable size difference was observed among Cu, Ag, and Si, a mutual separation between them just by classification was impossible, and gravity and/or shape separation may be necessary.

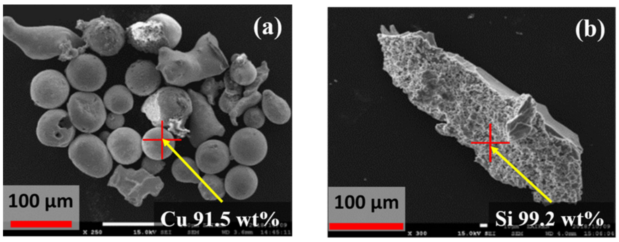

Fig. 8. SEM images of (a) Cu and (b) Si particles obtained at 13 kV.

3.3. Liberation of Recovered Particles

SEM images for small particles obtained at 13 kV are shown in **Fig. 8**. The Cu particles were spherical, and the Si particles were irregularly shaped. The generation of microparticles by electrical explosion appeared to be caused by the droplets. Under certain discharge energy, the Cu wire transformed to liquid state. At this time, during the heat expansion, the Cu droplets were divided into small pieces and simultaneously cooled by the surrounding water and formed spherical microparticles. However, the Si particles appeared to be produced by shockwaves generated by plasma expansion [23]. The SEM results indicated that physical separation was possible for small particle size fractions using the difference in the shapes of Cu and Si particles.

The samples were analyzed via MLA, an image analysis software application, using SEM-EDS. The results of the MLA measurement of Cu and Ag particles are shown in **Figs. 9** and **10**, respectively. The particles were sorted by their high Cu/Ag content with a high degree of separation; the others are not shown in **Figs. 9** and **10**.

Among the Cu particles shown in **Fig. 9**, those larger than 1180 μm seem to have been separated by the shockwave after the Cu wire explosion and disconnected by the plasma without the temperature rising above the melting point. Because the large particles had almost the same composition as the original Cu ribbon, these would be reusable without further treatment. Particles below 300 μm were mostly circular, whereas 300–1180 μm of the particles had an arbitrary shape. This may be because of the temperature rise of the particles; if the liquid Cu droplets cooled after division into small droplets, they would have a small spherical shape, and if not, an arbitrary shape.

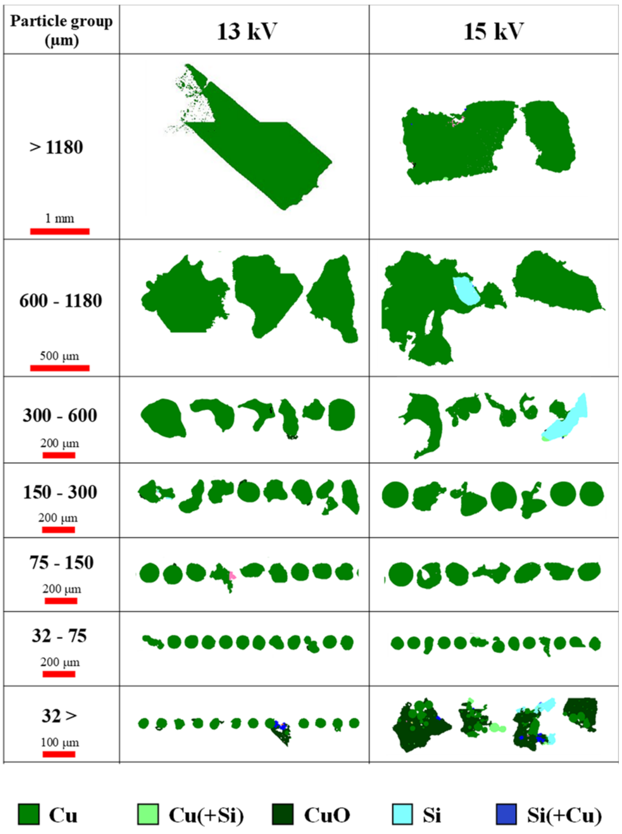

Fig. 9. MLA results of Cu particles.

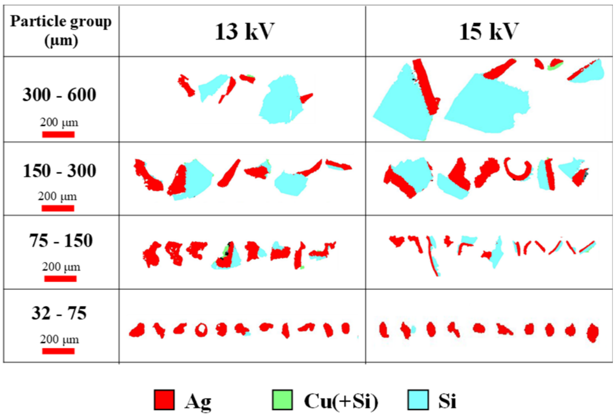

Fig. 10. MLA results of Ag particles.

Among the Ag particles shown in **Fig. 10,** smaller particles of 32–75 μm were circular, whereas larger particles than 75 μm had an arbitrary shape. Particles smaller than 75 μm and larger than 300 μm were liberated, whereas almost all medium-sized particles of 75–300 μm were rocked with Si particles. From the comparison between **Figs. 9** and **10**, Ag particles had fewer circular particles than Cu particles. This means that more Cu particles were melted by the explosion, whereas more Ag particles separated by shockwaves, although the melting point of Ag (1235 K) is lower than that of Cu. Because electrodes were connected at both ends of the Cu wire, the current mainly flowed through the Cu wire, so the direct Joule heat generation in the Ag wire was limited to the direct contact with the Cu wire. The results suggested that the parts of Ag wires in direct contact with the Cu wire exploded with the Cu wire, resulting in a small spherical shape. However, the other Ag wires were separated from the cell sheet by the shockwave of the explosion and then melted by the radiant heat of the plasma.

Both Cu and Ag are simple eutectic alloys with Si, and the lowest melting eutectic temperature is 1073 K for the Cu-Si system and 1103 K for the Ag-Si system [24]. Therefore, it seems that some Si melted at the lower

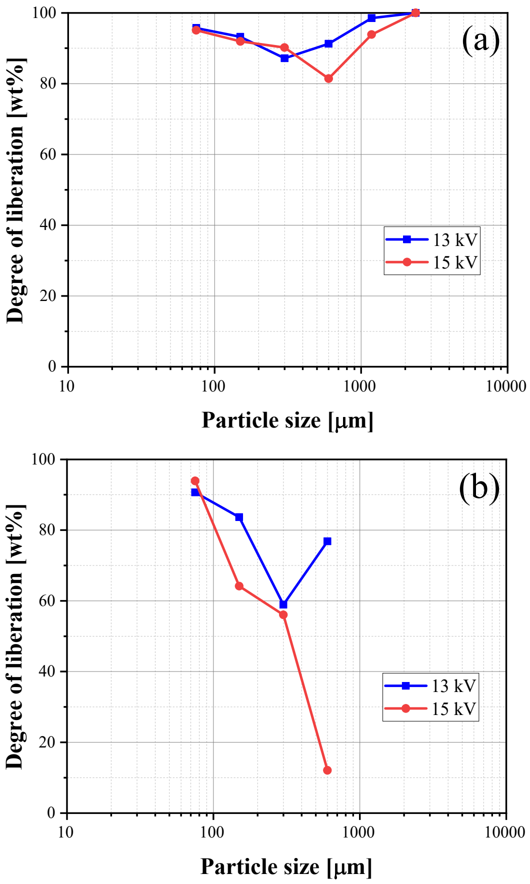

Fig. 11. Liberation of (a) Cu particles and (b) Ag particles.

temperature than the melting point of Si (1687 K), was cooled by the surrounding water, and formed silicide of Cu and Ag.

The liberation of Cu and Ag particles is shown in **Figs. 11(a)** and **(b)**, respectively. The particles containing Cu or Ag of more than 80% were regarded as in liberation. The liberations of Cu particles were particularly low at particle sizes below 32 μm at 15 kV, as shown in **Fig. 11(a)**, but the Cu contents were high, as shown in **Table 2**. The difference in liberation between 13 kV and 15 kV was more pronounced for smaller Ag particles as shown in **Fig. 11(b)**. The liberation of Ag particles was generally lower than that of Cu particles, implying that a significant amount of Cu and Ag reacted with other surrounding materials such as Si to form compounds by eutectic melting, and/or generated nano-sized Cu particles were attached to the other materials at 15 kV. The liberation of 15 kV was lower than that of 13 kV because higher discharge energy made it easier to produce a hot dense plasma that could react with surrounding materials frequently. A large number of Cu and Ag compounds at 15 kV are undesirable because they require high-load chemical treatment for further concentration. As a result, if a further separation of Cu and Ag is necessary for a higher level of reuse/recycling, pulsed discharge at 13 kV should be more desirable than at 15 kV and liberated Cu and Ag particles can be separated by wet gravity and/or shape separation from Si particles.

In the previously reported EHF methods, a non-contact electrode with the sample generates a shockwave by plasma generation of water between the electrodes and the sample, and the shockwave indirectly grinds the sample. However, in this study, the contact between the electrodes and the Cu wires enables the plasma generation of Cu and Ag wires directly, and then selective separation and grinding of Cu and Ag wires is achieved.

4. Conclusions

A novel electrical dismantling method was experimentally studied to selectively separate Cu and Ag from the cell sheets of spent PV panels. The electrode was directly applied to the Cu wire of the cell sheet, and Cu and Ag were simultaneously separated as particles from the resin sheet by electrical wire explosion using pulsed power. Si also accompanied the particles owing to shockwaves generated by plasma expansion and some of them formed compounds with Cu and Ag by eutectic melting. Excessively high energy discharge of 15 kV in the experiment partially generated hot plasma, resulting in a low liberation and increased Cu and Ag compound for small particles below 600 μm. In the case of a 13-kV experiment, the total average liberation of the Cu and Ag particle was 94.3% and 77.5% which are acceptable for wet gravity and/or shape separation for a higher level of reuse/recycling. Moreover, particles in the coarse grain group larger than 1180 μm were peeled from the Cu wire by shockwaves and had the potential to be reused as-is.

Improvement in the Ag recovery rate remains an issue of this method and it attempted to be addressed by changing the position of electrical pulse irradiation. A comparison of energy consumption, environmental load, and cost between this method and the pyrometallurgical process including thermal liberation and the hydrometallurgical process should be conducted for further industrial implementation. We will report the results in subsequent papers.

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Main Works:

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- "Modeling the Special Dynamics of Robotic Manipulators with Flexible Links and Joint Clearances," ASME J. of Mechanical Design, Vol.115, Issue 4, pp. 839-847, 1993.

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- "Effect of rice bran as a nitrogen and carbohydrate source on fed-batch simultaneous saccharification and fermentation for the production of bioethanol from rice straw," J. of the Japan Institute of Energy, Vol.94, No.1, pp. 151-158, 2015.
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- "Experimental study of the water jet induced by underwater electrical discharge in a narrow rectangular tube," Shock Waves, Vol.27, No.2, pp. 257-270, 2017.
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