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INNOVATIONS IN WEEE RECYCLING

Novel Pretreatment Process of Critical Metals Bearing E-Scrap By Using Electric Pulse Disintegration

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Abstract
Recovering various metals, involving minor rare metals, from electronic devices is important for conserving resources and establishing a sustainable society. This paper describes an economically feasible and radical process for such recovery, especially, for Ta concentration from printed circuit boards (PCBs) which is the two-stage electric pulse disintegration (ED) process. Ta capacitors are detached from the PCBs in the first stage, and the sintered Ta contained within the capacitors is liberated from the capacitor in the second stage. It has been indicated that as much as 100% of Ta capacitors was detached from PCBs in nondestructive form in the first stage of ED and that 95% of sintered Ta and MnO_2 phases was liberated from covered plastics, which is much higher than the 46% detachment achieved when using mechanical roll crusher. The detailed detachment and liberation mechanisms of both stages were also described, and an upper limit of the solution's conductivity in the second-stage ED was indicated.

Keywords Electric pulse disintegration · Printed circuit board · Ta capacitor · Detachment · Liberation

Introduction

Considerable amounts of minor rare metals are contained in the WEEEs especially in printed circuit board (PCB) and are required to be recovered by some physical concentration method before being fed into the Cu smelting process, because most of these metals are more reactive with oxygen than Cu and thus could not be recovered by the smelting processes. Since these metals are usually present in some specific devices in the PCB, detachment of devices from the PCB followed by the mutual separation is one of the best methods of concentration [1–4]. Electric pulse disintegration (ED) has been known for its strong ability to liberate compositional elements from ores [5–12] and solid waste materials (urban mines) [13–15], and more specifically, to detach the devices from PCB in nondestructive form and/or liberate compositional metal elements from the other insulating materials depending on the size of the devices [16]. The paper mainly describes the detachment and liberation processes of the devices and metal phases, especially, Ta capacitors and the sintered Ta, present within the Ta capacitor, from the PCBs, which must be an economically feasible and efficient method for their concentration.

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Sample and Experimental

Samples used in this research were the printed circuit boards (PCBs) of communication equipment as shown in Fig. 1. All the devices on the board were classified into 56 types, and each type of devices was crushed, finely ground, and then analyzed using x-ray fluorescence (XRF) for the metal elements except precious metals, and using inductively coupled plasma mass spectrometry for precious metals in order to identify their characteristics in each process. The Mineral Liberation Analyzer (MLA) is also

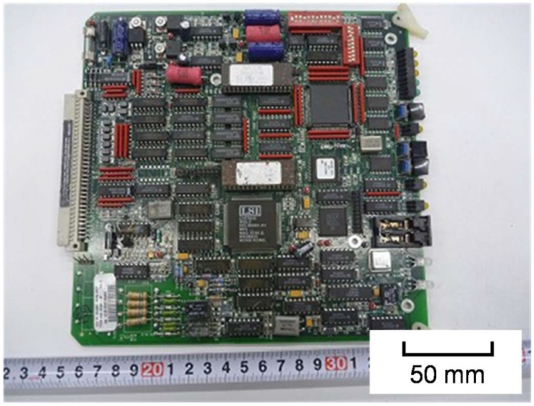

Fig. 1 Communication PCB used in this research (Color figure online)

used for more detailed analyses to clarify the liberation process of the sintered Ta from the Ta capacitors.

Electric pulse disintegration (ED) equipment used was from SELFRAG LAB and is shown in Fig. 2 (left). We applied two-stage ED process to the PCBs, objectives of which were the detachment of Ta capacitors from PCBs in nondestructive form in the first stage and the liberation of the sintered Ta from the detached Ta capacitors in the second stage. The voltage and the number of pulses applied in the ED were 160 kV and 200 in the first stage and those applied were 100–160 kV and 50–200, respectively, in the second stage in order to perform the liberation process of Ta capacitors. The PCB was first cut into several parts for being fed into the first-stage ED because the cell size was limited. We used a high-speed camera to capture the images of the ED phenomena of Ta capacitors and IC chips. Roll crusher, shown in Fig. 2 (right), was also used in the second-stage comminution in order to compare the liberation behaviors of the sintered Ta from Ta capacitors.

ED experiments of Ta capacitors were also carried out in the aqueous solutions of various electric conductivities in order to determine the effect of solution conductivity to the ED performance.

Composition of Minor Rare Metals in PCBs

We calculated the number of device types to be collected for obtaining 90% recovery of each metal element from the results of the chemical analyses of all the 56 types of the device. The results are shown in Fig. 3. Most of the minor rare metals indicate the small amounts, which shows that these metals are concentrated in some specific devices and that the other common and precious metals which could be recovered in nonferrous metals smelters are distributed into various types of devices. Then, minor rare metals could be classified into “Concentrated-type elements” and the common and precious metals into “Dispersed-type elements.” It must be mentioned that device separation is one of the best methods to obtain the concentrations of the minor rare metals. Here, we classified Ta capacitors into two types, yellow and black colored, and then Ta, the number of which was 2.5, is almost present in only two-colored Ta capacitors.

Two-Stage Electric Pulse Disintegration

First-Stage ED

Typical images of the ED products are shown in Figs. 4 and 5, which indicate that most of the metal phases were liberated from the other components such as insulating plastics almost in nondestructive form, especially Cu wire which was completely liberated from covered plastics (Fig. 4), and that ceramic capacitors, with sizes being less than several millimeters, were all detached from the PCBs also in nondestructive from PCBs (Fig. 5 left). Ta capacitors showed completely the same behavior as the ceramic

Fig. 2 ED equipment, SELFRAG LAB, (left), and roll crusher (right) (Color figure online)

Nd	Dy	Ta	W	La	Ce	Zn	Cr	Pd	Mn	Sr	Fe	Co	Au	Pb	Cu	Zr	Ni	Ag	Pt
0.9	1.6	2.5	2.5	2.7	3.4	3.7	4.3	4.4	4.7	6.1	6.9	8.2	9.9	10.7	12.1	12.2	14.4	14.6	17.8

“Concentrated type elements” ⟷ “Dispersed type elements”

Fig. 3 The number of device types to be collected for obtaining 90% recovery of each metal (Color figure online)

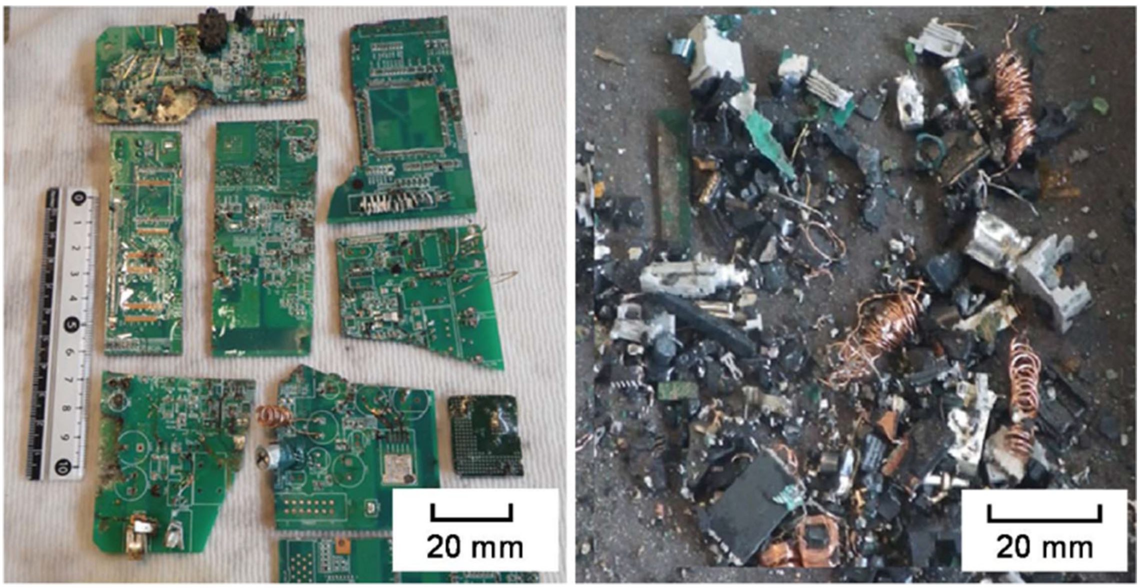

Fig. 4 ED products of PCB (left) and detached materials (right) after application of 100 pulses at 160 kV (Color figure online)

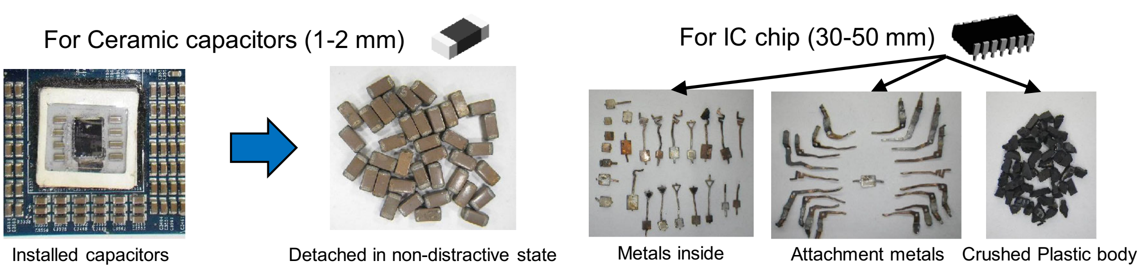

Fig. 5 ED products of detached ceramic capacitors (left) and IC chips (right) after application of 100 pulses at 160 kV (Color figure online)

capacitors. We also recognized that IC tips, with their sizes being of the order of several centimeters, were destroyed, but the metal phases, such as those inside electrodes and connectors to the PCB, were completely liberated in non-destructive form (Fig. 5 right) from the insulating covered plastics. These results must indicate that the minor rare metals could be concentrated by the subsequent device separation and that liberated metal phases could be separated by subsequent conventional physical separation technologies such as magnetic separation, eddy current separation, and/or electrostatic separation.

Detachment Process of Ta Capacitor from PCBs

Ta capacitors could be detached from PCBs in nondestructive form by applying proper operating conditions for ED. A finite element method simulation, involving electrostatic analysis, using EStat program (Field Precision LLC) showed four kinds of positions in a Ta capacitor to which the high-voltage pulse reaches, as shown in Fig. 6, and the high-speed camera images proved the simulation result to be reliable (Fig. 7). According the above observed phenomena, all the Ta capacitors could be detached from PCBs completely in nondestructive form in these ED experiments.

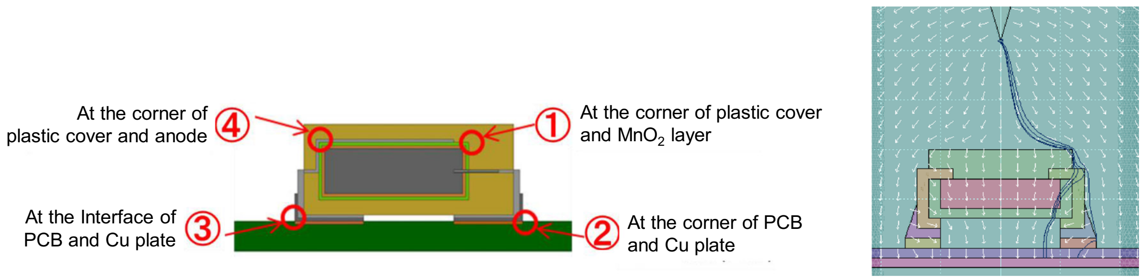

Fig. 6 Specific four positions at which high voltage pulse reaches (left) and the calculated electric flux lines from the upper electrode in the ED (right) (Color figure online)

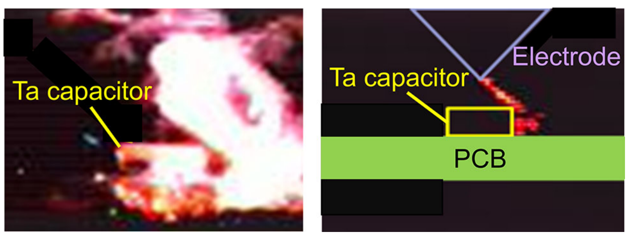

Fig. 7 Photograph showing ED behaviors of Ta capacitor and IC chip taken by high speed camera (Color figure online)

Second-Stage ED

In the second stage of the ED, the objective is the liberation of the sintered Ta from the plastic cover. We could clearly identify the liberation process of the sintered Ta by performing many experiments and by the observation of the products using x-ray diffraction, XRF, and MLA. Figure 8 shows the investigated process, in which the electrode in Ta capacitor was first liberated from the body and then the sintered Ta; furthermore, Mn oxides which cover the sintered Ta could be also gradually liberated from the sintered Ta.

Comparison of Liberation Behaviors Between ED and Mechanical Crushing

We compared the liberation behaviors of the ED and a mechanical roll crusher (RC). The results are shown in Fig. 9 which shows the relationship between the particle size of the ED product and the degree of liberation of each component in the Ta capacitor. From the figure, we can recognize that the ED greatly enhances the liberation of Ta-bearing components (Ta + MnO_2 and Ta itself) and that the differences between recovery rates can be as much as 46 wt% (95 wt% by ED and 48 wt% by RC) for Ta + MnO_2 and 40 wt% (47 wt% by ED and 7 wt % by RC) for Ta alone, on the assumption that only the free Ta-bearing particles could be recovered in the subsequent

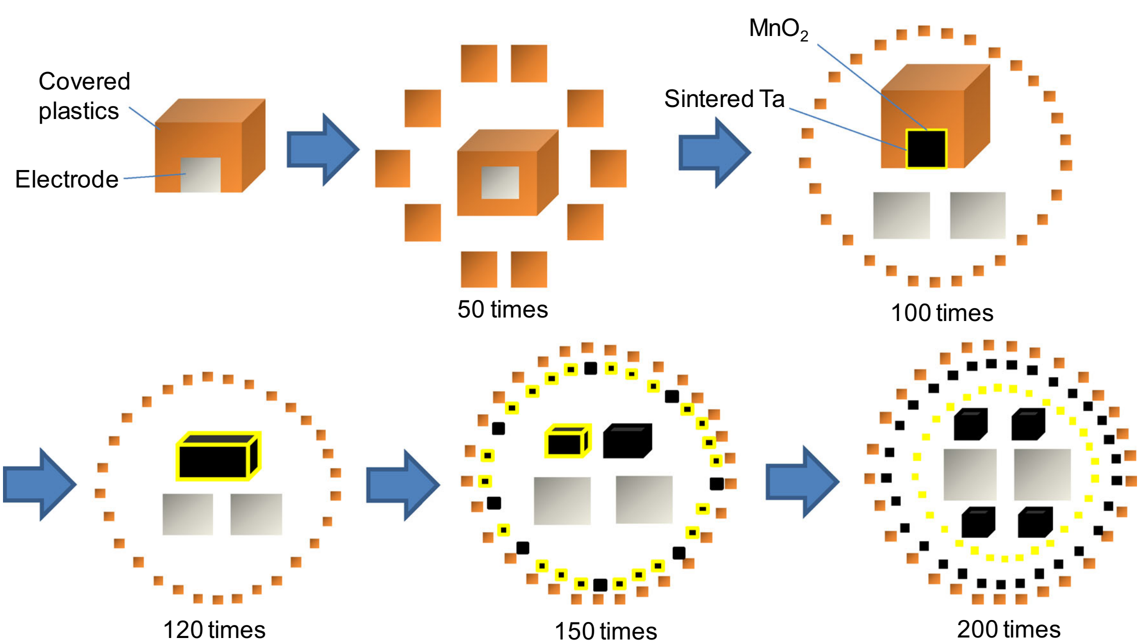

Fig. 8 Liberation process of Ta capacitors in the second stage ED, 160 kV (Color figure online)

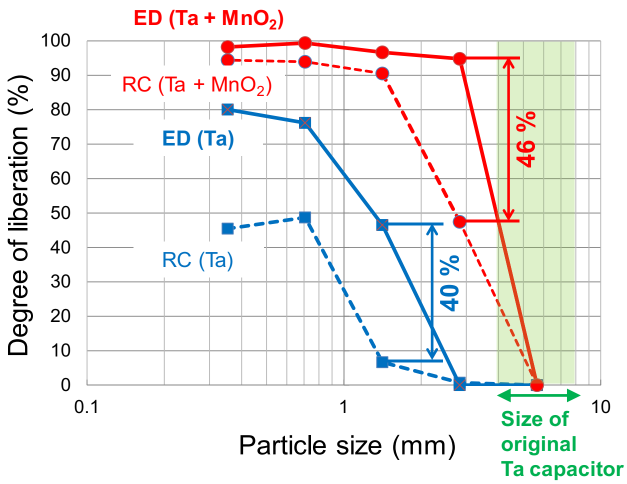

Fig. 9 Degrees of liberation of various components of the comminuted products after ED (160 kV) and RC (1.5 mm of rolls gap) (Color figure online)

physical separation processes such as gravity separation, etc. It was found that the ED clearly contributed toward improving the Ta recovery.

Effect of Aqueous Solution Conductivity

We carried out ED experiments of Ta capacitors obtained from the PCB in the aqueous solutions of various electric conductivities. Figure 10 (left) shows the relationship between solution conductivity and "non-ED ratio" which was defined as the ratio that electric pulse did not reach the solid sample but the surrounding water, under the condition of. Figure 10 (right) shows the same relationship but with respect to the 50% of average size of ED products. These figures illustrate that the solution conductivity, 400 mS/m, was roughly determined as the upper limit to make an appropriate ED of solid samples on the PCBs. Then, the conductivity of aqueous solution in a continuous ED process should be monitored, and the ED equipment should have a cleaning process to remove electrolyte from the solution.

Conclusion

We applied a two-stage electric pulse disintegration (ED) process for concentrating the sintered Ta which was present in Ta capacitors contained in the PCBs. In the first stage, the detachment of 100% Ta capacitors from PCBs was achieved in nondestructive form, and in the second stage, 95% of the sintered Ta was liberated from covered plastics which was 46% higher than the recovery achieved when using a mechanical roll crusher, which could indicate that much higher recovery of Ta components could be expected in the subsequent physical separation processes such as gravity separation, etc. We also demonstrated the detachment and liberation mechanisms of both the ED stages and the higher limit of electric conductivity of aqueous solution, 400 mS/m, in the second stage ED.

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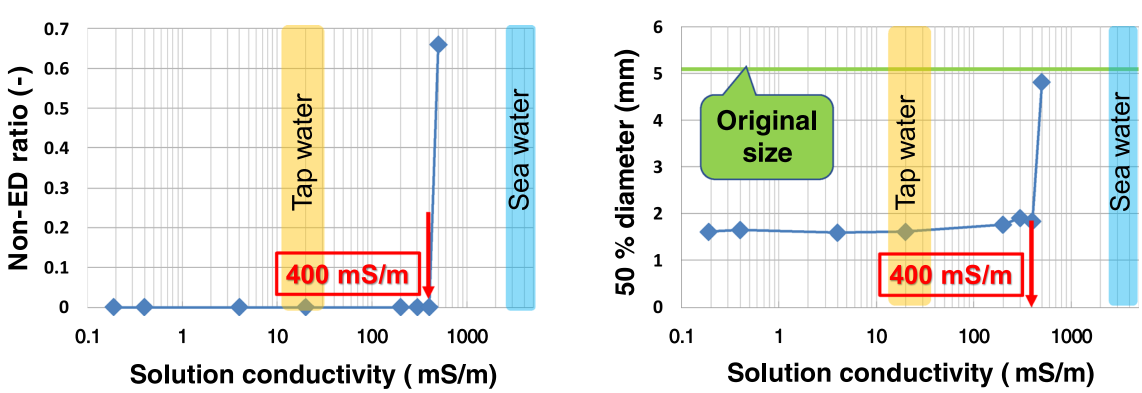

Fig. 10 The effect of solution conductivity on the ED performance under the condition of 160 kV and 200 times, showing the relationship between solution conductivity and non-ED ratio to all the pulses applied (left) and 50% diameter of the product (right) (Color figure online)

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