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Evaluation of a recycling process for printed circuit board by physical separation and heat treatment

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ABSTRACT

Printed circuit boards (PCBs) from discarded personal computer (PC) and hard disk drive were crushed by explosion in water or mechanical comminution in order to disintegrate the attached parts. More parts were stripped from PCB of PC, composed of epoxy resin; than from PCB of household appliance, composed of phenol resin.

In an attempt to raise the copper grade of PCB by removing other components, a carbonization treatment was investigated. The crushed PCB without surface-mounted parts was carbonized under a nitrogen atmosphere at 873–1073 K. After screening, the char was classified by size into oversized pieces, undersized pieces and powder. The copper foil and glass fiber pieces were liberated and collected in undersized fraction. The copper foil was liberated easily from glass fiber by stamping treatment.

As one of the mounted parts, the multi-layered ceramic capacitors (MLCCs), which contain nickel, were carbonized at 873 K. The magnetic separation is carried out at a lower magnetic field strength of 0.1 T and then at 0.8 T. In the +0.5 mm size fraction the nickel grade in magnetic product was increased from 0.16% to 6.7% and the nickel recovery is 74%.

The other useful mounted parts are tantalum capacitors. The tantalum capacitors were collected from mounted parts. The tantalum-sintered bodies were separated from molded resins by heat treatment at 723–773 K in air atmosphere and screening of 0.5 mm. Silica was removed and 70% of tantalum grade was obtained after more than 823 K heating and separation.

Next, the evaluation of Cu recycling in PCB is estimated. Energy consumption of new process increased and the treatment cost becomes 3 times higher comparing the conventional process, while the environmental burden of new process decreased comparing conventional process.

The nickel recovery process in fine ground particles increased energy and energy cost comparing those of the conventional process. However, the environmental burden decreased than the conventional one.

The process for recovering tantalum used more heat for the treatment and therefore the energy consumption increased by 50%, when comparing with conventional process. However, the market price for tantalum is very large; the profit for tantalum recovery is added. Also the environmental burden decreased by the recycling of tantalum recovery. Therefore, the tantalum recovery is very important step in the PCB recycling. If there is no tantalum, the consumed energy and treatment cost increase in the new process, though the environmental burden decreases.

1. Introduction

Waste Electrical and Electronic Equipments (WEEEs) should be recycled because they contain many kinds of valuable materials. Most of WEEEs include printed circuit boards (PCBs). In Japan, 20–30 km^2, that is about 50 thousand tons, and approximately 10 billion USD per year of printed circuit board (PCB) has been produced as shown in Fig. 1 (METI, 2011). PCBs are normally separated from WEEEs by handpicking and sold to non-ferrous metal suppliers because PCBs include high-grade copper and other precious metal such as gold, silver and palladium (Cui and Zhang, 2008). The typical composition of PCB from a personal computer is listed in Table 1 (MOE & METI, 2010). Plastic content is about 30% that may include flame retardant. Copper content is large (i.e. about 20%), and several precious metals are also present, for

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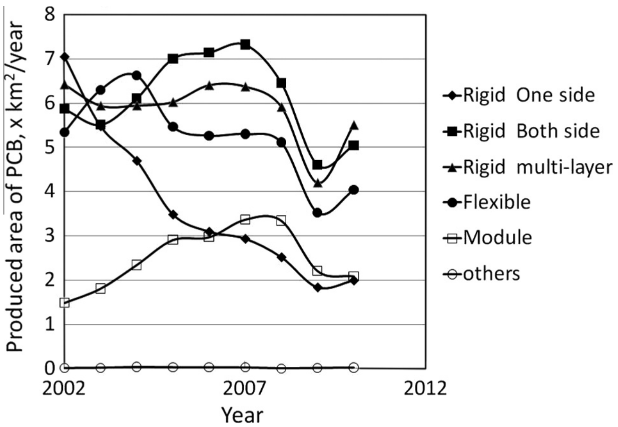

Fig. 1. Production of printed circuit board in Japan (METI, 2011).

Table 1
Typical composition of a printed circuit board (PCB) for personal computer (MOE & METI, 2010) (Important parts are shown in bold.).

Organic components	**30%**	Cr	760 ppm
Br in organic components	**3%**	Mn	614 ppm
Sb	0.42%	Sr	407 ppm
		Au	**320 ppm**
Si	10%	Zr	190 ppm
Ca	5%	Nd	158 ppm
		Pd	**90 ppm**
Cu metal	**15%**	Nb	66 ppm
Al metal	4%	In	64 ppm
Fe	2.50%	Co	62 ppm
Pb	2.00%	Mo	47 ppm
Ni	**0.46%**	W	40 ppm
Ag	**0.12%**	La	31 ppm
Ta	**0.10%**	Ce	29 ppm

example, Au is used for plating whereas Ag and Pd are used as electrodes. The precious metals and copper are recovered by smelting. However, PCB also contains impurities such as silica, alumina and calcium oxide, which are slag materials in non-ferrous smelting process, and other harmful elements such as lead, bromine and antimony. Low grade PCBs are sold after the removal of valuable parts or sometimes land filled without any treatment. On the other hand, the electric parts mounted on PCB contain several kinds of rare metals such as Ta and Ni. However, they are difficult to be recovered in the conventional copper smelter.

Recovery of metal from PCB has been carried out using several methods, such as crushing, separation and leaching (Eswaraiah et al., 2008; Wu et al., 2008; Yoo et al., 2009; Fouad and Abdel Basir, 2005). The recovery of glass fiber has also been studied.

In this paper, at first, the mechanical crushing and water explosion method are employed to remove the mounted parts on PCBs. The underwater explosion is very convenient to liberate the different density of materials (Fujita et al., 2009).

After the removal of mounted parts, copper and nickel recovery is subject of the carbonization treatment. Pyrolysis of PCB has been studied but these studies were mainly focused on the pyrolysis gas and char constituent (Hall and Williams, 2007; Hung-Lunga et al., 2007; Grause et al., 2008; Jie et al., 2008). As the PCB includes bromine as flame retardant, bromine should be removed in the combustion process of PCB. The time to reach thermodynamic equilibrium is within 0.25 s and at 1073 K, Br_2 is still main component gas in the 4.4% partial pressure of O_2 and the ratio Br_2/HBr decreases with rising temperature as shown in the following formula (Ni et al., 2012).

$$2Br_2\ (g) + 2H_2O\ (g) = 4HBr\ (g) + O_2\ (g)$$

The incinerated gas is treated with water, since the exhausted gas containing Br_2 and HBr is passed through the activated carbon and the bromine compounds are adsorbed on the carbon. In the carbonization of this experiment the muffle furnace of 2.5% of O_2 atmosphere is used by purging N_2 gas. Resin in PCB is pyrolyzed and metal is not oxidized by carbonization. It is, therefore, expected to recover copper and nickel as metals efficiently in the post process.

On the other hand, the tantalum capacitor, as one of the valuable mounted electric parts in the removed mounted parts, is collected by the eddy current separation. In this study, the tantalum capacitor is collected from mounted parts on the hard disk drive. The recovery method from tantalum capacitor is studied by the heating method. Tantalum recovery is carried out in the production process of tantalum capacitor; however, the tantalum is not recycled from the used capacitor or PCBs (JOGMEC, 2008). Regarding the tantalum recovery, several reports suggest that metal tantalum is recovered from capacitor by electronically mediated reaction (Mineta and Okabe, 2005), and the parts of tantalum capacitor is then recovered after crushing (Kageyama et al., 2009; Ooki et al., 2009). Here, the recovered tantalum will enter the tantalum smelting process.

2. Experimental and discussion

2.1. Liberation of mounted parts and bare board

In the copper smelter, PCB is usually chopped the handling its size for the furnace by mechanical crushing and then heated. It is difficult to recover low grade of rare metals in fine crushed parts. Also aluminum metal complicates slag handling. Many kinds of mechanical crushing methods are developed to liberate the mounted parts from the PCB (Yokoyama and Suzuki, 1999; Ueno et al., 2000; Hisakado et al., 2001, 2003; Yamaguchi et al., 2009). In this experiment, the mounted parts are liberated from the board by underwater explosion and mechanical crushing.

2.1.1. Underwater crushing

The underwater explosion (Fujita and Shibayama, 2002; Fujita et al., 2008) is utilized, because it is very effective in separating composites of different densities. A gel type explosive is used for PCB in personal computer and household electrical appliance. Several PCBs with mounted parts are immersed in the water and the explosive is placed above the board to be detonated. The photographic results are show in Fig. 2. The circles are the explosion area in (a) and (b) of Fig. 2. In the near explosion area the parts are well liberated. The phenol resin board was easily disintegrated. The stripped mounted parts from PCB of personal computer are shown in (c) and the range is from several mm to 5 cm. The effective emulsion explosive that is set on the hanged PCB in water and about 1 wt% of PCB in the water is important for underwater explosion.

2.1.2. Mechanical crushing

There are several crushers being used in order to liberate the mounted parts on PCB such as the inclined crusher (Japanese patent, 2009) that fixes several protruding portions, the cross flow shredder (Sato Tekko, 2012) that rotates several chains and so on. The as-received PCB or the crushed PCB by underwater explosion is fed into the inclined crusher and the cross flow shredder.

2.1.3. Physical separation

Next, the liberated mounted parts are sieved in 4 mm and the +4 mm size fraction of mounted parts is released from the PCB bare boards. The metal parts such as tantalum capacitor and aluminum