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RECYCLING

# Recycling of waste printed circuit boards by mechanical milling

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Abstract. Waste printed circuit boards (WPCBs) – a common type of e-waste consisting of electronic components with a significant amount of rare and precious metals as well as glass fiber composites. The development of WPCB retreatment technology through mechanical methods, resulting in metallic and non-metallic waste fractions, can be efficiently classified as enriched concentrate. The research presented in this paper highlights the potential trends for recycling WPCBs. The integration of impact milling for WPCBs is a promising technology for the partitioning and fragmentation of different components and for the separation of fractions. The mechanical size-reduction and separation technology of pre-crushed WPCBs, using direct and separative disintegrator milling, has been compared with other technologies (hammer milling and high voltage fragmentation). The main parameters, such as the specific energy ( $E_S$ ) of WPCB treatment and the rate of separation of different phases, were studied. As preliminary results show the optimal parameters of partitioning of the metallic and non-metallic parts, fragmentation and delamination (specific energy of treatment and rate of separation) by disintegrator milling were proposed.

Keywords: WPCBs, impact milling, partitioning, fragmentation, separation, waste metal, glass fiber.

## **1. INTRODUCTION**

Mechanical recycling is a promising way to process printed circuit boards (PCBs) and is becoming increasingly popular because of its simplicity.

In most cases, the process of recycling PCBs begins with pretreatment, followed by separation processes and the creation of concentrates of metals and non-metals, and completed by the final chemical/mechanical cleaning of the obtained metallic part.

Pretreatment includes the following: a) disassembly and separation into parts, resulting in shredded pieces of PCBs suitable for further processing, b) delamination and fragmentation of board plates, which will result in the opening of the composite structure and the release of components suitable for follow-up separation, c) separation of the metallic part (MP) and non-metallic part (NMP). Traditional mechanical milling devices, such as shredders and hammer mills [1–4], are not very efficient because they do not selectively grind components. In direct fragmentation by these methods, the components of WPCBs will be partitioned but without delamination of board plates from the MP. However, high-quality release and separation of the components is possible only when the composite structure is delaminated, which requires an unreasonably high level of fragmentation. Therefore, traditional mechanical grinding methods have a relatively high energy consumption (Table 1). As a result, alternative ways of opening the structure are being searched.

For example, in [5–7], selective electrodynamic fragmentation (EDF) technology was used. The use of EDF enables selective fragmentation of materials, removal of electronic components (ECs) to open the structure, and size reduction through generating electrical discharges as a means of fracturing. EDF can be used as a pre-weakening tool for recycling materials such as WPCBs, carbon fibers, bottom ash, and siliceous rods. The ECs of WPCBs

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Method and stress type	Ground WPCB size, mm	Partitioning rate (%) at specified size	Cumulative ground product – yield, wt%	Energy of treatment <i>E</i> , kWh/t
Dry hammer mills; shearing [1–3]	2.8–2.0	66	≤84	~198
Wet hammer mills; shearing [2]	6-0.5	89	90	~2.6 times higher than dry milling
EDF; electric shock [1]	6–3	99	95	Partitioning 132 + Delamination 877 + Fragmentation 1485
Disintegrator mill; impact [10]	1.4-0.355	>90	95	129

Table 1. Comparison of different technologies for WPCB retreatment

are partitioned along the material interfaces instead of being comminuted. The form of the components is largely preserved, and they are separated from other surrounding elements or contaminants. The benefits of EDF are coarse partitioning of different elements (i.e., energy savings), recovery of original-size elements (aggregates), and selective partitioning of valuable metals. With Selfrag technology [5], WPCB components can be efficiently separated without the fine comminution of the material often achieved with mechanical crushing. The valuable materials can be recovered as these are partitioned during the process.

In EDF processes, minimal size reduction results in the partitioning of ECs and the delamination and fragmentation of the board. EDF technology delaminates the boards and opens their structure, exposing Cu foils to leaching solutions [7]. In EDF technology, there are three stages: partitioning, delamination, and complete fragmentation.

Partitioning-oriented leaching results indicated that EDF treatment should be used as a pretreatment stage for the recycling of WPCBs. Delamination and fragmentation stages consume more energy. Thus, EDF-partitioned WPCBs can be further size-reduced to render Cu available for hydrometallurgical downstream processing.

High Intensity Impact Milling (HIIM) using disintegrator technology (promoted by TalTech) is relatively little known. In many cases, this technology has a number of advantages over traditional methods. First and foremost, it is the high selectivity of the impact and the low specific energy of grinding  $E_{\rm S}$ .

Papers [8,9] are devoted to the study of the recovery of used PCBs based on green pyrolysis. Derived from previous experience [10,11], this paper proposes to apply disintegration technology for PCB recycling as an alternative to traditional grinding methods.

The objects of the study are:

• determination of the rational energy of delamination and fragmentation,

- grinding dynamics (accumulation of size fractions),
- distribution of the metal component in the fractions of the milled material, which is important for the sub-sequent optimization of separation processes.

## 2. EXPERIMENTAL PART

#### 2.1. Initial materials

The initial material selection for the experiments is justified with a recreation of the industrial conditions for the trials. The following materials were used:

- manually dismantled WPCBs (Fig. 1a) better preserved PCBs with higher gross value of rare precious metals (RPMs) due to the attached electrical and electronic components. This test group is to be subjected to the "multi-step" simulation trials,
- pre-crushed (shredded) WPCBs (Fig. 1b) the most common stream as the automated disassembly has higher production rates. This test group is to be utilized in the separative milling mode.

Particle-size distribution of the ground product was determined by sieving. The screening rate  $k_Q$  was determined as the ratio of the mass of the stated powder fraction to the total mass of the ground product, where the fraction from 1.4 to 0.355 mm was assigned a stated granulometry.

## 2.2. Technology and used devices

For mechanical treatment of WPCBs, the following devices were used:

- one-rotor moderate velocity disintegrator mill DSA operating in direct mode,
- two-rotor high velocity semi-industrial disintegrator DSL-115 operating in direct or separative modes.

Milling experiments were carried out at different energies (velocities) of milling: direct multi-step milling



Fig. 1. WPCBs: (a) – manually disassembled, (b) – pre-crushed by shredder.

was carried out by disintegrator DSA (up to 4 times) followed by disintegrator DSL (up to 16 times). Parameters of the used disintegrators are given in Table 2.

For the classification of the ground product and the determination of granulometry, Fritsch Analysette vibration sieves were used. According to our estimates, the uncertainty of the obtained results was less than 2%. Chemical composition of the ground product (disintegrator milling by DSL-115 in separative mode) was determined, using X-ray fluorescence (XRF) and atomic absorption spectroscopy (AAS) methods.

## 2.3. Content of metals

Depending on the type of PCBs, the content of metals in them may differ significantly. However, the rate of accumulation of size fractions and the proportional content of the MP in them with the same technological parameters should remain approximately the same since this is determined by the mechanism of impact on the material.

PCBs based on FR-4 fiberglass composite, the most common in the production of PCBs, were pre-crushed by

Type of disintegrator mill	Mode of operating	Energy of treatment $E_{\rm S}$ , kWh/t				
		Nominal (regime 1)	Increased (regime 2)			
DSA	direct	2.4	4.06			
DSL-115	direct/separative	4.3	7.27			

Table 2. Parameters of used mills



Fig. 2. Schemes of: (a) - disintegrator DSA and (b) - disintegrator milling system DSL-115.

shredder mills to a piece size less than 20 mm (Fig. 1b) and finally ground using the disintegrator milling system DSL-115 in separative mode to obtain a fraction of the material less than 1.4 mm in size. The amount of the MP was determined both in the total ground product as well as in the stated fraction.

## **3. RESULTS AND DISCUSSION**

The results of multistep milling using various disintegrators are given in Table 3. According to Table 3, the amount of the desired ground product granulometry (1.4– 0.355 mm) after pre-crushing grinding in DSA was about 21–25%, reaching up to 41% during milling by DSL-115 at the nominal treatment velocity of 58.3 kWh/t and 30.5% at 30.8 kWh/t (increased velocity). The amount of dust after pre-crushing grinding was 17–28%. With subsequent milling, the amount of the desired fraction began to decrease. Simultaneously, the amount of fine fraction – dust (<0.355 mm) – increased up to 56–86% at both treatment velocities.

The influence of the specific energy of treatment at subsequent separative milling mode on the ground product particle size and yield of the desired fraction are given in Fig. 3. The threshold for mean particle size was decreased monotonically from 14 mm to less than 0.090 mm.

According to Fig. 3, the optimal specific energy of treatment to obtain a more suitable fraction for the future reuse of MP and NMP is about 35–45 kWh/t. Increasing the specific energy of treatment results in an increase in the amount of dust.

The specific energy of treatment  $E_{\rm S}$  (dependent on impact velocity) plays an important role in the yield of the stated fraction. By increasing  $E_{\rm S}$ , the maximal yield increased up to 48% at nominal velocity (Fig. 3). At increased velocity, the maximal yield of the stated fraction is achieved by increasing  $E_{\rm S}$  up to 37 kWh/t, reaching up to 31%. At subsequent milling, the amount of fraction of 1.4–0.355 mm starts to decrease, while the amount of fine fraction (dust) increases up to 56–86%.

Based on the results of disintegrator milling experiments, it is important to highlight the following findings in the retreatment of WPCBs:

- The effect of velocity (specific energy of treatment *E*<sub>S</sub>) is important both at the preliminary as well as at the final milling, and the specific energy of treatment plays the main role in the range of studied energies.
- The optimal amount of the stated fraction was obtained at specific energies of 35–45 kWh/t (42 at lower speed, about 37 at higher speed); the cumulative ground product amounts (screening rates) were about 48% and 31%, correspondingly. Simultaneously, the amount of dust was 37% and 52% at nominal and increased energies of treatment.
- The dynamics for size reduction of the ground product  $(d_m)$  was similar at different milling velocities: at nominal and increased velocities, the output of the stated size (1.4–0.355 mm) was about 1.5 mm (nominal) and 0.5 mm (high). To achieve higher treatment efficiency (higher output) and reduce the amount of fine fraction (dust), lower velocity is preferable.

The composition of the metallic part of the milled PCBs is presented in Table 4 and summarized in Fig. 4.

- It can be observed that the milled PCBs contain approximately 31% of various metals, with about half of the total composition being copper (16%) and aluminum (10%), which account for 52% and 32% of the total metal content, respectively.
- The ground material contains precious elements, such as gold and silver, at approximately 265 ppm (Au) and 210 ppm (Ag).

Milling steps	-	energy of	Screening rate $k_Q$ , %						
	treatment	t Es, kWh/t	1.4-	-0.355 mm	Dust (<0.355 mm)				
	nominal	increased	nominal	increased	nominal	increased			
Pre-crushing by DSA	2.4	4.1	8	11.6	7	10.3			
	4.8	8.1	14	16.1	12	15.5			
	7.2	12.2	17	22.5	15	23.0			
	9.6	16.2	21	25.2	17	27.9			
Milling by DSL-115	13.7	23.5	24	27.3	21	40.0			
	17.7	30.7	32	30.5	24	45.9			
	42.1	74.4	48	21.6	37	75.9			
	58.3	103.4	41	13.8	47	85.9			
	74.6	132.5	38	_	56	_			

Table 3. Disintegrator treatment at nominal and increased milling energies



Fig. 3. Dependence of changes in the granularity (mean particle size  $d_m$ ) and yield (screening rates  $k_Q$ ) on the nominal (1) and increased (2) specific energies of treatment  $E_S$ .



Fig. 4. Content of metals in the metallic part (MP) and in the milled PCB.

Fraction, mm		Metallic component, % of total milled PCBs											
	Fe	Си	Al	Sn	Bi	Zn	Pb	Sr	Ni	Ag (ppm)	Au (ppm)	Other metals	Σ%
1.4-0.71	0.10	10.7	3.14	0.42	0.22	0.42	0.04	0.03	0.04	111	34	0.54	15.64
0.71-0.355	0.07	3.72	2.59	0.14	0.14	0.02	0.03	0.01	0.10	54	39	0.23	7.05
0.355-0.180	0.08	1.36	2.40	0.08	0.14	0.01	0.02	0.01	0.07	24	40	0.41	4.58
0.180-0.09	0.04	0.30	0.65	0.05	0.04	0.00	0.01	0.01	0.02	15	9	0.20	1.32
<0.09	0.19	0.27	1.26	0.27	0.05	0.01	0.08	0.02	0.02	59	87	0.43	2.60
Σ%	0.47	16.34	10.04	0.96	0.59	0.46	0.19	0.08	0.25	265	210	1.81	31.25

Table 4. Composition of the MP in different fractions



■ 1.4–0.71 mm ■ 0.71–0.355 mm ■ 0.355–0.180 mm ■ 0.180–0.090 mm ■ less than 0.090 mm

Fig. 5. Distribution of the MP in different fractions.

- The distribution of the MP in different fractions is shown in Table 4. Figure 5 illustrates the proportionality of the distribution of metallic components in various fractions. The fraction sized 1.4–0.71 mm consists mainly of the MP suitable for subsequent processing, containing most of the non-ferrous metals. The amount of middle and lower fractions (<0.710 mm) was 68%, but the main MP was predominantly present in the coarse fraction (51% of the total metal content).
- Lower fraction dust (<0.355 mm) is mainly an NMP (glass fiber and epoxy) and will be re-treated by pyrolytic processes. In the fine fraction (<0.090 mm), the mode of accumulation of the metal component is clearly traced, which confirms the effect of increased grinding energy on the process efficiency.

## 4. CONCLUSIONS

- High Intensity Impact Milling (HIIM) technology and disintegrator mills can be used for the recycling of WPCBs, which encompasses the fragmentation of boards as well as partitioning of electronic components and the separation of metallic and non-metallic parts.
- To achieve a higher output (screening rate) of the stated fractions, a lower specific energy (impact velocity) is recommended. At the optimal specific energy of treatment, the output of fraction 1.4–0.355 mm is about 50%, consisting mostly of the MP of the ground product.
- 3. The obtained data on the composition of the MP and the proportional distribution of the metallic content in various fractions are important as a guideline for the development of optimal grinding–separation parameters, which should consider the requirements of end-users when calculating the cost of the method and the rationality of its use.

 Retreatment of the fine fraction (<0.09 mm), mainly glass fiber and epoxy, will be realized by thermal processes.

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# E-jäätmete ümbertöötlus mehaanilise jahvatuse teel

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Uurimistöös tuuakse välja e-jäätmete – kasutatud trükiplaatide (WPCB) – ümbertöötlemise võimalikud suundumused. Ümbertöötlus löökjahvatamise teel on WPCB-de puhul paljulubav tehnoloogia nende erinevate komponentide purustamiseks, eri materjalide peenendamiseks ja erinevate osiste eraldamiseks. Eelpurustatud WCPB-de edasiseks peenendamiseks ja osiste eraldamiseks kasutati desintegraatorjahvatust otse- ja separatsioonjahvatuse meetodil ning võrreldi seda teiste jahvatustehnoloogiatega (vasarveskis ja kõrgpingelahenduse teel jahvatus). Uuriti peamisi parameetreid, nagu töötluse erienergia ja eraldamise määr jahvatuse eri etappides. Esialgsete tulemustena pakuti välja optimaalsed parameetrid elektroonikakomponentide ja metalsete ning mittemetalsete osade eraldamiseks, peenendamiseks ja ka WCPBde laminaatplaadi delamineerimiseks desintegraatorseadmete abil.

Saadud andmed metalse osa koostise ja metallisisalduse kohta erinevates fraktsioonides on olulised optimaalsete jahvatus- ja eraldusparameetrite väljatöötamiseks, mis peaksid olema aluseks pakutud meetodi maksumuse arvutamisel ja otstarbekuse üle otsustamisel, võttes arvesse WCPB-de ümbertöötluse tulemusena saadud materjalide lõppkasutajate nõudeid.