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Posted Date: 6 January 2026

doi: 10.20944/preprints202601.0216.v1

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Eco-Efficient Recycling of Printed Circuit Boards

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Abstract

The article presents a technology for the physical recycling of printed circuit boards (PCBs) that is consistent with the principles of circular economy and sustainable production. A Life Cycle Assessment (LCA) was performed for PCB recycling using shredding, grinding and physical and physicochemical processes such as electrostatic separation, gravity separation and flotation for the separation of metals and plastics. On the basis of this assessment and the selectivity criterion, electrostatic separation was found to be the best separation method used after shredding and cryogenic grinding. Furthermore, the financial potential of recycling and other benefits that recycling can bring to the economics of the business and to the protection of the environment were presented. The possibility of using non-metallic fraction (plastic) generated during the recycling as an additive in the production of composite materials was assessed. The functional properties of the composite were assessed (static tensile, hardness, pin-on-disc, and Schopper-Schlobach abrasion tests), as well as the ecotoxicity of the powder added to polymeric materials such as polyester and epoxy resins, and silicone, used in the production of consumer goods.

Keywords: physical separation in recycling; printed circuit boards; LCA; circular economy; composite; phytotoxicity; economic analysis

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1. Introduction

The rapid growth of the world's economies has contributed to the overconsumption of natural resources, and with the development of new technologies, there is an increasing demand for certain critical raw materials whose deposits are limited and may be depleted in the near future. The reason for this was the economic model of the time, which consisted of extracting materials, producing goods, using them, and then, once they had lost their use value, disposing of them without considering further processing to obtain secondary raw materials. This has resulted in raw materials, even valuable ones, becoming waste [1]. The change in the old linear economic model started in the era of rising energy prices and the recognition of the problem of depleting deposits. A new business model began to be implemented in various industries with the aim of extending the life cycle of raw materials while maintaining their residual value [1,2]. This model has been referred to as a circular economy whose implementation is essential to ensure the security of raw materials. Furthermore, this new concept offers opportunities to reduce the environmental footprint of products therefore considered them compatible with sustainable production principles [3]. However, sustainable production is mainly about choosing technological processes to produce goods that have the least impact on the environment, thus life cycle assessment is a good tool to evaluate and compare them.

Waste Electrical and Electronic Equipment (WEEE) is currently one of the fastest growing waste streams. According to the United Nations Institute of Training and Research (UNITAR), 62.3Tg of WEEE was generated in 2022, while WEE generated and reported in 2019 was 53.6Tg valued at $ 57 billion [4]. This waste stream is estimated to grow to 74.7 Tg in 2030 and to 110 Tg in 2050 [5]. The rapidly growing volume of this waste is becoming a challenge for companies looking to recover valuable substances. A common element found in almost all WEEE is Printed Circuit Boards (PCBs). The most used are FR-4 type PCBs, which are constructed from a substrate made of glass fibre and epoxy resin. The FR-4 Waste Printed Circuit Boards (WPCB) composite contains embedded metallic pathways containing large amounts of metals such as Cu, Fe, Al, Sn, rare earth elements, Ta, Ga, and others from the lanthanide group, and the precious metals Au, Ag and Pt [6]. WPCB FR-4 also contains metals hazardous to human health and life and the natural environment, such as Cr, Pb, Be, Hg, Cd, Zn and Ni [7,8]. Therefore, to protect the environment, WPCB should be processed according to the principles of circular economy and sustainable production. It is also worth noting that the concentrations of metals contained in PCBs are tens or even hundreds of times higher than in ores [9,10]. The average metal content in PCB is: Cu - 20%, Al - 2%, Pb - 2%, Zn - 1%, Ni - 2%, Sn - 4%, Ag - 0.20%, Au - 0.10%, Pd - 0.005%, Pt - 0.0015% [7,11]. The most valuable metals in WPCBs are gold, palladium, silver and copper, and the recovery of these metals can bring significant economic benefits to the recycling entity [12]. The value of metals per tonne of PCB is: Cu - $1,770, Al - $51, Pb - $39, Zn - $30, Ni - $305, Sn - $1,139, Ag - $1,928, Au - $84,556, Pd - $1,532, Pt - $454 [13]. Therefore, WPCB recycling is necessary for reasons of resource conservation and raw material safety. This is also confirmed by the relative supply risk index developed by the Royal Society of Chemistry, which assesses future raw material availability problems. In determining the index, factors such as the abundance of the earth's crust, the distribution and quantity of reserves, production concentration, substitutability, recycling rates, and political stability are analyzed. Most of the metals of which PCBs are composed have an index greater than six on a 10-point scale, where 1 means very low risk and 10 means very high risk. These are mainly Pd, Pt, Sn, Ag, Pb and Ni, for which the relative supply risk is 7.6, 7.6, 6.7, 6.2, 6.2 and 6.2, respectively [14].

It should be noted that the environmental impact of inadequate WPCB recycling may be greater than that of extracting metals from ore deposits. Therefore, to protect the natural environment, WPCB should be recycled according to the principles of a closed-loop economy and sustainable production. Therefore, it should be remembered that it is necessary that residues resulting from the recovery of valuable substances can be reused, preferably for the production of other consumer goods. These principles should already be implemented in the design stage of recycling technology, which should be efficient, have a low environmental impact, and do not contribute to the depletion of natural resources. A technology that takes these criteria into account can be described as eco-efficient. Implementing eco-efficiency principles in the design of WPCB recycling technologies can contribute, among other things, to lower material consumption, reduced waste and reduced emissions, and ultimately increase the competitiveness and image of companies through sustainable solutions and environmental progress. Therefore, the development of an eco-efficient WPCB recycling technology represents a huge potential for e-waste recyclers. It is also an opportunity to reduce the environmental impact of low-carbon recycling technologies and source metals from secondary sources, thus reducing their extraction from natural resources.

Because of the complex structure of WPCBs, their recycling is a complex process that requires preparatory measures. The most important of these is the dismantling, which involves removing reusable components and/or decontaminating by removing toxic substances from the surface of the WPCB. Depending on the recycling method adopted, components that could interfere with further processes are also removed from the WPCB. The WPCB prepared in this way can become a suitable feedstock for the recovery processes, which can be carried out either by chemical methods and/or combination of both, chemical and physical methods which are based solely on mechanical separation. The most common recycling methods for WPCBs are chemical treatment, such as hydrometallurgy [15] or thermal process such as pyrometallurgy including plasma incineration. The common hydrometallurgical processing involves metal dissolution in acids while pyrometallurgy uses high temperature thermal process to produce metals. In addition to the high complexity of these methods and the high level of emissions to the environment, there is also a loss of non-metallic part that could be used for manufacturing of other products., which are otherwise lost into the residue after processing. The disadvantage of the hydrometallurgical method include water pollution by

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hazardous substances, while the pyrometallurgical method involves the emission of greenhouse gases and flue gases into ambient air. Both methods generate hazardous waste. Alternative ways of processing WPCBs are using mineral processing principles, starting with the size reduction done by crushing and grinding for liberation of valuable components which is necessary for the efficient separation. The physical separation methods can include electrostatic separation, gravity separation, magnetic separation, and any combination of above mentioned methods [16]. These physical separation techniques have a much lower environmental impact, but because of the difficulty of releasing metals (liberation) from the WPCB composite materials, which requires several technological processes, they are less commonly used. When using physical separation methods, three or more products are generally produced: a mixture of metals with a high financial value, plastics with a small amount of glass fibres, and an intermediate, middling's type of material i.e., grain containing plastics and metals. The key issue is to minimize middling's product while producing the cleanest possible mixture of metals and plastics.

The aim of this work was to introduce eco-effective WPCB recycling technology that allows to generate profits by selling precious metals and using plastics for the production of other consumer goods. In particular, the best option for separating metals from plastics from ground WPCBs was determined considering their economic and practical application in industry while leaving a minimal environmental footprint. A preliminary economic analysis was also conducted to obtain information on the process costs of physical recycling. Furthermore, the monetary value of recovered metals calculated per 1 million inhabitants of southern Poland was presented, as well as the possibility of using recovered non-metallic powders for the production of other consumer goods. This assessment took into account type of polymer matrix such as silicones, epoxy and polyester resins. The effect of powder addition on their functional properties of the composite was investigated, such as hardness, abrasion resistance, and tensile strength. Because consumer products are intended for use in the human environment, an additional phytotoxicity test of the non-metallic powder was conducted to assess its impact on the environment.

Products made of polymer-based composite materials, because of their properties, such as resistance to hydrolytic degradation and UV degradation, have found wide application, among others, as protective coatings in various engineering construction materials. The production of composite materials consists of the combination of at least two components with different physicochemical properties. Epoxy and polyester resins and silicones, which make up the matrix of the composite material, are used most frequently for the production of composite materials. Composite materials are obtained by using various processing techniques, the most common of which are extrusion, casting, and lamination [17–19] and 3D printing.

In the context of WPCB recycling, LCA provides a comprehensive framework to assess the environmental impacts of different recycling processes, including the steps of separating metals from plastics. By applying LCA, it is possible to select the process with the least environmental impact and identify hotspots where environmental impacts are most significant, as well as to investigate ways to mitigate these impacts. By conducting the LCA at the laboratory scale, the insight is gained into the potential environmental benefits and drawbacks of the processing before scaling up to an industrial level. This approach enables the identification of sustainable processes for the optimization of resource use, energy consumption, and waste generation.

2. Materials and Methods

2.1. Methods

2.1.1. WPCB Recycling Technology

The WPCB recycling technology using physical methods has been divided into 5 stages (Figure 1a). The goal was to separate metals from plastics, while the first stage of the research was to remove components from the surface of the PCBs that may interfere with subsequent processes, i.e., sockets, resistors, transistors, processors, RAM disc, and others. Disassembly was carried out using handheld workshop tools [20]. A preliminary selection of components with high reuse potential was made, such as processors and their cooling systems, various types of connectors, and RAM memory. Removal of these components is also possible using mechanical stripping methods. Dismantled WPCBs were crushed in a shredder to obtain pieces not larger than 1 x 1 cm [20]. Reducing the size of the WPCB was necessary to prepare material for the milling. The proper release of the metal grains from the rest of the WPCB composite matrix is one of the most critical factors for successful recycling by physical methods to achieve efficient separation. In the previous work by the authors [21], the use of liquid nitrogen was observed to have a positive effect on the WPCB grinding process. However, the cryocooling of the WPCB pieces could not take place directly in the mill's working compartment, so this stage was divided into two parts. First, the WPCB cut pieces were cooled to below -150°C using liquid nitrogen in a special liquid nitrogen tank. The frozen WPCB pieces were ground in LMN-100 laboratory knife mill (Testchem LLC, Poland) equipped with a 1 mm sieve. The result was generation of a WPCB powder with a maximum particle size of 1.5 mm.

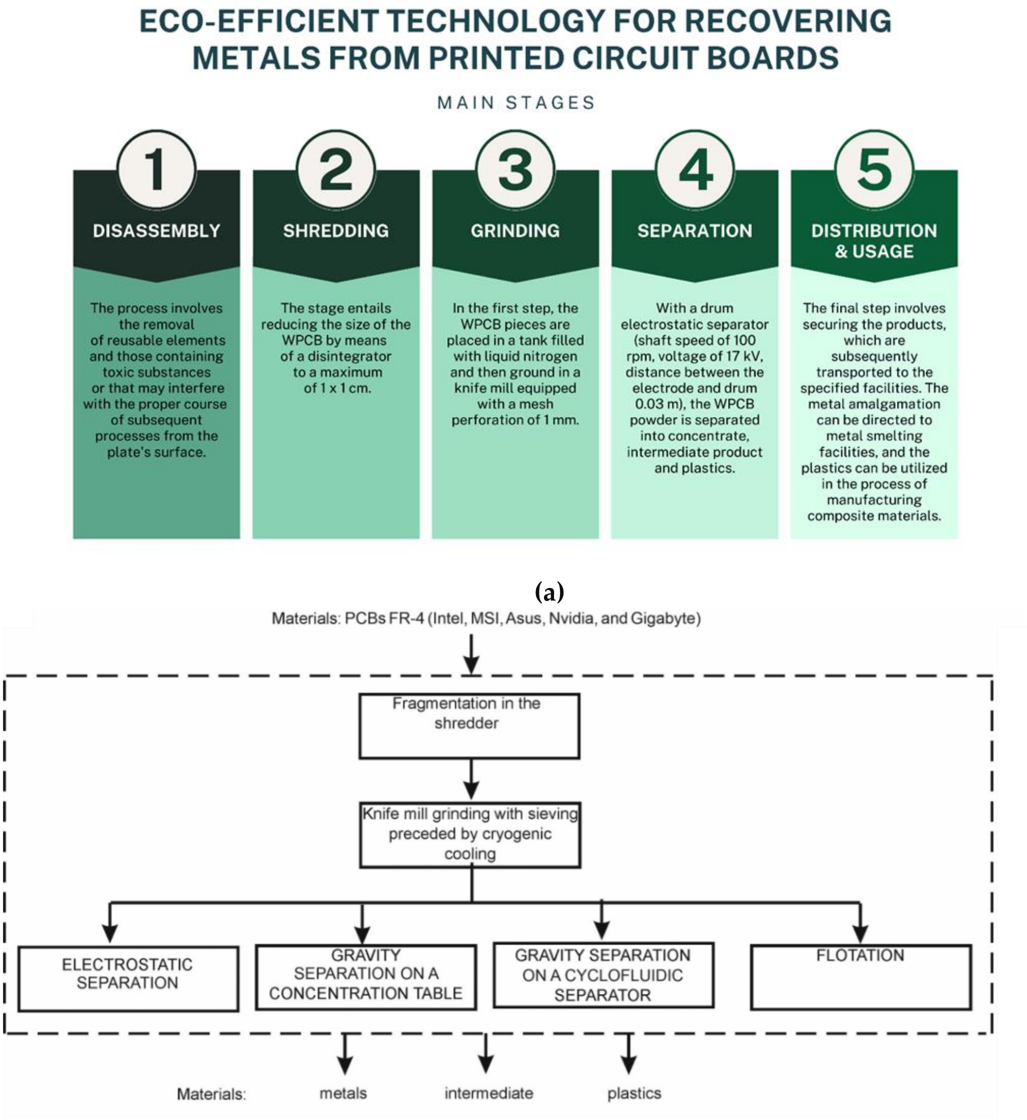
Separation of metals from the WPCB composite was carried out using four laboratory devices i.e. an electrostatic drum separator, a shaking table, a cyclofluid separator, and a flotation machine. These devices were fed with the same material. The separation was optimized for all of the devices in order to obtain the best possible separation result. A shaking table, cyclofluid separator, and flotation used water for separation, so the resulting products had to be dried.

The first separation process studied was electrostatic separation, which is based on differences in the surface charge storage capacity and conductivity of the dry particles. In electrostatic drum separators, nonconductive particles adhere to a rapidly rotating drum, which is electrified by friction against a brush that also mechanically scrapes these particles (plastics). Conductive particles (metals) rapidly release their surface charge, allowing them to easily be 'stripped' from the drum surface. To increase the efficiency of this process, the electrostatic separator is equipped with a high-voltage electrode that bombards the particles with electrons. A Boxmag Rapid Laboratory Electrostatic Drum Separator (Aston, Birmingham, UK) was used, which was designed to receive three products, i.e., metal, middling’s, and plastic particles. Separation was carried out under the following conditions: a shaft speed of 100 rpm, a voltage of 17 kV, and a distance between the electrode and the drum of 0.03 m [22].

The second method of separation investigated was gravity separation, which was used to separate particles with different densities. Due to the wide variety of devices and different designs amongst gravity separators, it was decided to use two types of gravity separators that interacted with the particles in different ways. In both cases, the separation was performed in aqueous medium. The first piece of equipment used in gravity separation was a shaking table. Separation was carried out using a laboratory shaking table equipped with a grooved plate. Separation was carried as follows: table load of 9 dm^3/min (water suspension with material), water flow rate of the first nozzle of 5.7 dm^3/min, water flow rate of the second nozzle of 5.4 dm^3/min; table stroke of 1.5 mm, table movement frequency of 260 strokes/min, longitudinal inclination angle of 1^o and transverse inclination angle of 6^o [23]. The table made reciprocating movements so that the suspension with the particles, which were fed onto the surface of the table, were separated along the table according to their density. The lightest particles (plastics) were stripped from the table surface as the fastest flowing, while the heaviest particles (metals) overcame the hydrostatic pressure of the liquid and were carried along the table surface as a result of its movement. Spray nozzles were placed on top of the table to increase the efficiency of the separation process.

The second device was a laboratory cyclofluid separator, in which separation was achieved by vertical fluidization of the particles, where the vertical movement of the liquid caused the particles to separate according to their density. The heaviest particles (metals) fell to the bottom of the separator, while the lightest particles were lifted to the surface. The principle of operation of the laboratory cyclofluid separator involved the use of a semi-industrial U-shaped cyclofluid separator with continuous movement [24]. This separator was equipped with a water-filled tank in which a cylinder was placed. The cylinder containing the WPCB powder suspension was closed at the bottom with a 0.5 mm mesh size sieve and performed a vertical reciprocating motion. This created a cyclical fluid thrust in the cylinder and allowed the particles to be separated by density. In the cyclofluid separator, separation was achieved by fluidizing the particles in a fluid in a vertical direction. A laboratory cyclofluid separator was used for the separation. The following parameters were used: water volume of 13 dm^3, cylinder stroke of 4 cm and suspension movement frequency in the cylinder of 53 movements/minute [23].

The final separation process studied in the research was flotation, which involved the separation of particles on the basis of differences in their ability to repel or attract water. The flotation process took place in aerated water so that hydrophobic particles could 'stick' to air particles and float to the surface of the vessel. Flotation was carried out using a laboratory flotation machine Mechanobr, from IMN Gliwice (Instrumentation Building Plant of the Institute of Non-Ferrous Metals, Gliwice, Poland), with a 1 litre flotation tank. During the flotation process, a flotation reagent (Dimethoxy dipropyleneglycol) was used at a concentration of 157 mg/dm^3. The rotation speed of the magnetic stirrer, the rotation speed of the rotator in the flotation tank and the flotation time were 100 rpm, approximately 400 rpm, and 5 minutes, respectively [25].



(a)

(b)

Figure 1. a) The main stages of an eco-efficient WPCB recycling technology (in stage 4, electrostatic separation was presented, i.e., separation characterised by the best selectivity); **b)** Flowchart of WPCB recycling, illustrating four methods for separating metals from plastics together with the comminution steps.

After selecting the separation method, the final and fifth step was distribution and use. Plastics (waste) obtained as a result of WPCB recycling were used in the research as an additive in the production of composite materials, while the metal mixture (concentrate) can be sold.

2.1.2. Life Cycle Assessment (LCA)

LCA study focused on assessing the environmental impact of different options for separating metals from plastics from ground WPCBs. LCA was carried out according to ISO 14040/44 standards, which includes four main phases; goal and scope definition, inventory analysis, impact assessment, and interpretation [26–29]. The main objective of this LCA was to assess and compare the environmental impacts of four different separation options for WPCBs. These options included electrostatic separation, flotation, and gravity separation on a shaking table and in a cyclofluid separator. The study aimed to identify the most environmentally friendly separation method that could be scaled up for industrial applications. The functional unit for this LCA was defined as the treatment of 1 kg of WPCB, which ensures consistent comparisons between different separation processes. In addition to the separation options, the environmental impact of the comminution processes was also evaluated to understand the impact of the overall process.

System boundary

The LCA system boundary covered the steps of the WPCB recycling process from fragmentation in the shredder (stage 2 in Figure 1a) to producing a saleable metal mixture (stage 4 in Figure 1a). Each step is evaluated separately. As shown in Figure 1b, the WPCB recycling system included different scenarios of the following steps: fragmentation in the shredder, knife mill grinding with sieving preceded by cryogenic cooling and four separation options, including electrostatic separation, flotation, and gravity separation, on the shaking table and in the cyclofluidic separator.

The inventory analysis was carried out using the Ecoinvent 3.0 database, a comprehensive and widely recognised source of life cycle inventory data. The SimaPro 8 software was used to model processes and compile inventory data [30]. The inputs for each method were meticulously recorded and analysed. Data for shredding and cryogenic grinding, such as energy requirements and the usage of liquid nitrogen, were included along with the separation methods. The inventory table (Appendix 1) is listed in the supporting information.

The environmental impacts of the recycling processes were evaluated using the Tool for the Reduction and Assessment of Chemical and Other Environmental Impacts (TRACI), developed by the US Environmental Protection Agency (EPA) [32]. TRACI is a midpoint-orientated technique that evaluates environmental burdens across various impact categories, making LCA results more interpretable and actionable. Impact categories considered in this study were: Ozone depletion (kg CFC-11 eq), Global warming (kg CO_2 eq), Smog formation (kg O_3 eq), acidification (kg SO_2 eq), Eutrophication (kg N eq), Carcinogenic effects (CTUh), Non-carcinogenic effects (CTUh), Respiratory effects (kg PM2.5 eq), Ecotoxicity (CTUe), and Fossil fuel depletion (MJ surplus).

The results of the TRACI impact assessment were analysed to identify the most environmentally sustainable option to separate metals from plastics and to demonstrate the overall environmental impact of the presented recycling. The findings were interpreted to provide information on the potential environmental benefits and drawbacks of each process.

2.1.3. Using WPCB Recycled Plastic to Produce Composite Materials - Strength Parameters

In the case of plastics for the production of composite materials, the impact of the share of recycled powder in the matrix on the strength parameters of the final product was evaluated. The composites were prepared by gravity casting with a filler (plastics obtained from recycling) content of 2.5, 5.0, 7.5 and 10.0 wt% and were marked for silicone, polyester resin, and epoxy resin as: 2.5_F, 5_F, 7.5_F and 10_F, respectively. Before fillers were introduced into the matrix, hardening agents and matrix cross-linking initiators were added. The silicones and resins with the above agents were then placed in a rotary mixer and the filler was gradually added while mixing at a speed of 150 rpm until the assumed filler contents was achieved. After the fillers were placed in the matrix, the resulting mixtures were placed in appropriate forms. For polyester resin, the moulds were heated at approximately 80°C for 24 hours. Then 72 hours after pouring, the samples were removed from the moulds and subjected to mechanical and physical tests. Static tensile tests, Shore A type hardness tests, and Schopper-Schlobach abrasion resistance tests were performed for samples made of silicone materials, while static tensile tests, Shore D type hardness tests, and pin-on-disc tests were performed for resin materials. All tests were carried out at room temperature (22°C) and 50% humidity. Tensile tests were carried out according to [32] for five samples (type 5-B) cut from the composites and silicone samples. The tests were carried out on an Instron 4465 tensile tester (Instron, Norwood, MA, USA) equipped with a mechanical contact extensometer. The test speed was 500 mm/min. The tensile strength and elongation at break were determined. Shore A and D hardness tests were carried out according to [33]. Measurements were made with a Shore A-type hardness durometer Zorn (Zorn Instruments GmbH & Co. Hansestadt, Germany). Five measurements were made in each sample. The abrasion test (pin-on-disc) for epoxy and polyester resign was performed on the CSM Tribometer Instruments (Needham, MA, USA) according to ASTM G99. The test samples were cylindrical in shape, 10mm high and 12mm in diameter. Before the test, the samples were cleaned with technical ethanol. The ball moving after the sample, with a dimension of 6 mm, was made of zirconium dioxide. The ball was pressed against the sample with a force of 20 N. The ball's linear speed was 10 cm/s. The abrasion of the materials tested was defined as a change in the coefficient of friction (μ) on the 20 m road. For each group of materials tested, 5 measurements were made, for which the arithmetic mean was given. The abrasion wear of silicones was measured according to EN ISO 4649:2007 on an APG Schopper-Schlobach apparatus (APG Germany GmbH, Friedberg, Germany). For the abrasion test, the arithmetic mean of three measurements was given.

2.1.4. Using WPCB Recycled Plastic to Produce Composite Materials - Phytotoxicity Test

To determine the impact of recycled WPCB plastic on the surrounding environment, phytotoxicity tests were carried out on the leachate resulting from washing the powder with distilled water according to the PN-EN 12457-2:2006 standard. The plastics were leached on a ROTAX 6.8 shaker for 24 hours. The liquid/plastic ratio (L/S) in the tests was 10:1. The leaching test was carried out at 22°C. The eulate was filtered prior to use.

Phytotoxicity tests were performed for two dicotyledonous plants: mustard (Sinapis alba L.) and cucumber (Cuccumis sativus L.) and one monocotyledonous plant: wheat (Triticum spp). For comparison purposes, phytotoxicity was also determined for distilled water (control). For each species, the test was performed in Petri dishes in 5 repetitions. For this purpose, 10 ml of sample (distilled water and leachate) was placed on each plate with filter paper and 10 seeds were seeded. After 5 days after sowing, the number of germinated seeds was counted and 14 days after sowing, the lengths of the stem and root were measured. The tests were carried out according to PN ISO 11269-2 and PN-ISO 11269-1.

2.2. Materials

2.2.1. Material for Recycling

The materials used for recycling were popular motherboards such as Intel, MSI, Asus, Nvidia, and Gigabyte. The boards were dismantled from desktop computers from 2007-09. The PCBs belonged to the FR-4 group, in which the laminate was composed of glass fibre and epoxy resin [34].

2.2.2. Characteristics of Polymers and Fillers for the Production of Composite Materials

Three materials were used as a matrix to prepare the composites: polymer material in the form of silicone Mould Star™ 30 manufactured by Smooth-On, Inc. (smooth-on.com), AROPOL M 105 TB polyester resin (INEOS Composites) with BUTANOX M-50 initiator (INEOS Composites) (https://c-l.pl/), LG 120 epoxy resin (Havel) with HG350 hardener (GRM Systems) (www.grm-systems.cz/en/epoxy).

An addition to the matrix used was a filler in the form of recycled WPCB plastics. The powder grains were not larger than 1.2 mm and more than 40% of the grain masses were smaller than 0.125 mm. Plastic consisted mainly of fibrous and needle-shaped grains [21].

3. Results

3.1. WPCB Recycling

The results of research on the separation of metals from plastics using an electrostatic drum separator, a shaking table, a cyclofluid separator and a flotation machine have already been presented in articles [21,23,25]. The aim of this study is to select the most selective and environmentally friendly method (see LCA) among those analyzed and to retest the best one using a different and larger amount of feed (15 kg).

To demonstrate which of the analyzed methods is the most selective, it was proposed to use the specific density of the products as a parameter characterizing the quality of the obtained separation products (density is a parameter distinguishing metals from plastics) and to plot the dependence of the product yield on the cumulative (weighted) density of the separation product, i.e., the upgrading curve (Figure 2). The results indicate that electrostatic separation was the most selective method, therefore only this method is shown in Figure 1a and the separation was repeated for a larger amount of feed.

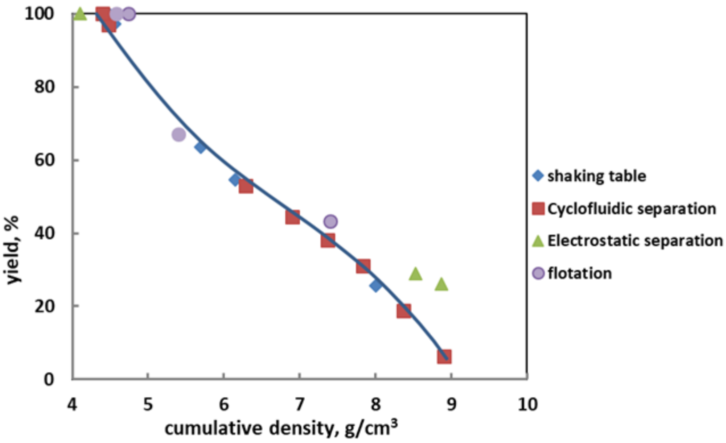
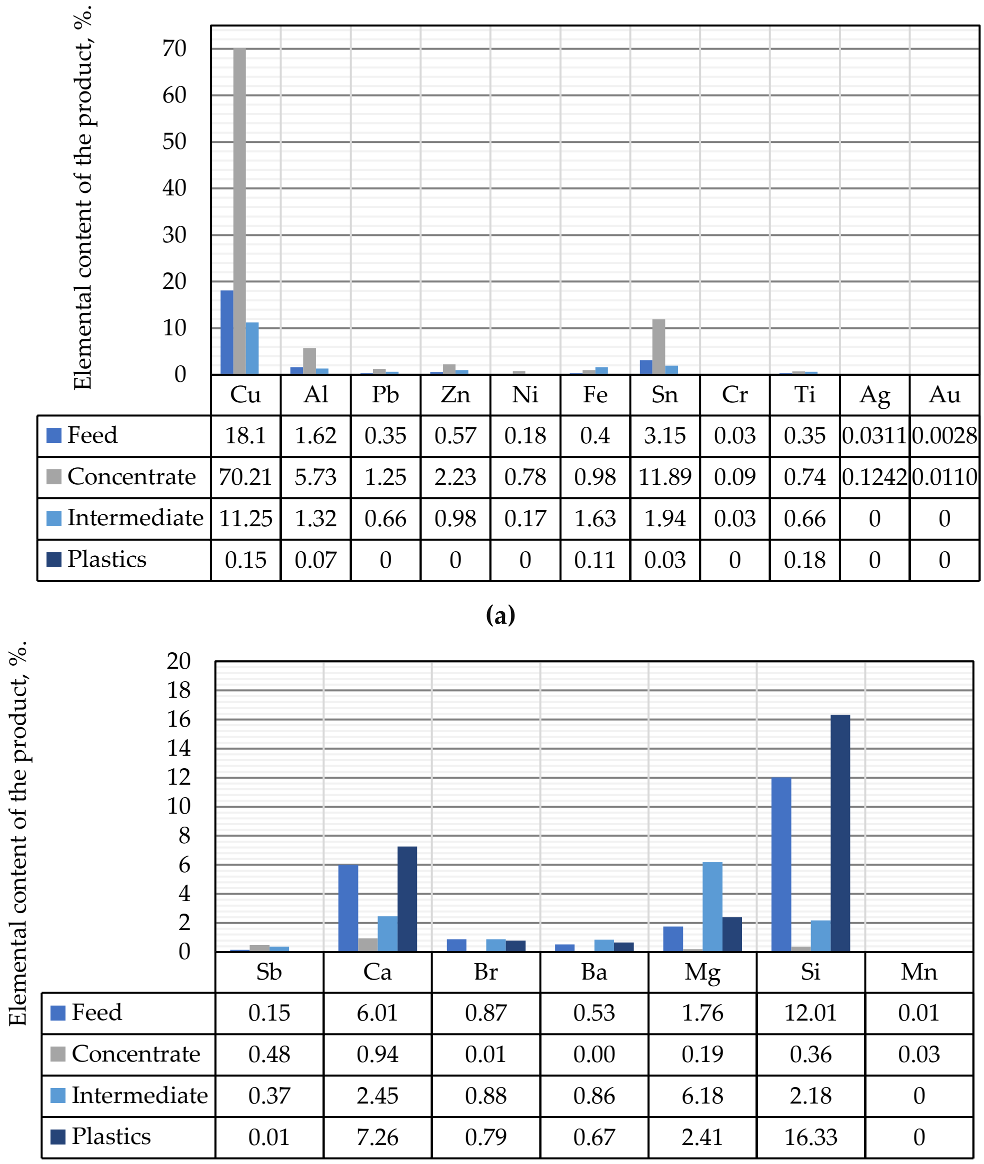


Figure 2. Correlation between product yield and cumulative density of separation product.

Three products were obtained in this way: concentrate, intermediate, and waste. Concentrate and waste yield were 25.1% and 72.5% of feed, respectively, with densities of 8.94 g/cm^3 and 2.15 g/cm^3. These confirm the high purity of the resulting metal concentrate and plastic as waste product. The intermediate or middling's product, whose yield was only 2.4%, consisted mainly of mixed in nature particles that cannot be physically separated. The density of this product is 5.41 g/cm^3.In order to release the metal from this fraction, these particles would need to be recrushed under the appropriate conditions to liberate metal fraction. Alternatively, to process middling's, another environmentally friendly method such as bioleaching can be used, in which microorganisms are catalysts for the metal leaching process, however, this cannot be branded as physical separation method. For metal recovery, the most widely used bacterial strain is Acidithiobacillus ferrooxidans, which is able to dissolve the metals contained in WPCB [35]. This method is usually used as an auxiliary method due to the mechanics of the process and its long kinetics [35–37]. However, for such a small amount of intermediate product, a small bioleaching installation would be an effective complement to complete the WPCB recycling process.

The chemical analysis of electrostatic separation products indicates that more than 94% of valuable metals (that is, Cu, Al, Pb, Zn, Ni, Fe, Sn, Cr, Ti, Ag, Au) are found in the concentrate (Figure 3). It could be assumed that because of the multilayer structure of the PCB, it is advantageous to obtain the smallest possible grains. Wu et al. 2008, however, report [38], that very fine grains, i.e. less than 0.125 mm, may contribute to inefficient electrostatic separation due to grain aggregation effects on the drum and electrode surface. In the work of Wu et al. 2009 [39] showed that this effect can have a significant impact on the stability of the separation process. The settling of plastic-structured dust on the electrode surface was observed in the study presented in this paper. However, the aggregation effect was observed to be strongly reduced when the material was separated using cryogenic temperatures during milling.



	Cu	Al	Pb	Zn	Ni	Fe	Sn	Cr	Ti	Ag	Au
■ Feed	18.1	1.62	0.35	0.57	0.18	0.4	3.15	0.03	0.35	0.0311	0.0028
■ Concentrate	70.21	5.73	1.25	2.23	0.78	0.98	11.89	0.09	0.74	0.1242	0.0110
■ Intermediate	11.25	1.32	0.66	0.98	0.17	1.63	1.94	0.03	0.66	0	0
■ Plastics	0.15	0.07	0	0	0	0.11	0.03	0	0.18	0	0

(a)

	Sb	Ca	Br	Ba	Mg	Si	Mn
■ Feed	0.15	6.01	0.87	0.53	1.76	12.01	0.01
■ Concentrate	0.48	0.94	0.01	0.00	0.19	0.36	0.03
■ Intermediate	0.37	2.45	0.88	0.86	6.18	2.18	0
■ Plastics	0.01	7.26	0.79	0.67	2.41	16.33	0

(b)

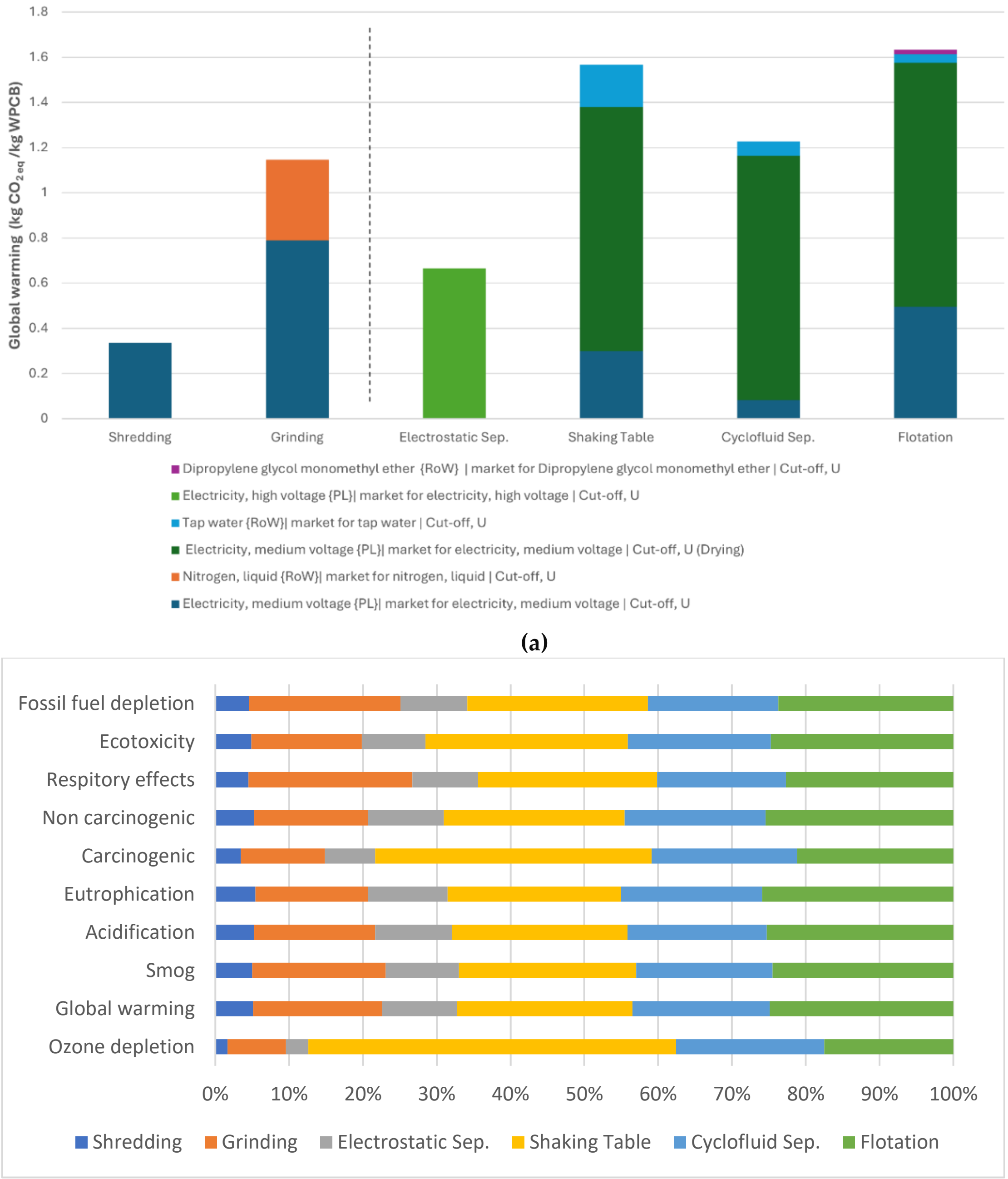
Figure 3. Chemical composition of feed and electrostatic separation products: (a) – valuable elements, (b) – non-valuable elements (value „0" i.e. below detection threshold).

3.2. Life Cycle Assessment (LCA)

The global warming potential (GWP) of the four WPCB separation options shows that they are ranked from most to least influential as follows: flotation, shaking table, cyclofluid separator, and electrostatic separation (Figure 4a). The latter has an impact of 0.665 kg CO_{2eq}/kg of WPCB. The

doi:10.20944/preprints202601.0216.v1

several times higher GWP for flotation, shaking table, cyclofluid separator, is mainly due to the need to dry the products obtained from these processes. For flotation, the impact of tap water consumption and flotation reagent consumption is very small, while the major GWP comes from energy used for mechanical flotation cell operation.



(a)

(b)

Figure 4. a) Global warming potential of four WPCB separation options along with the comminution processes. **b)** TRACI midpoint categories showing the relative contribution of all WPCB recycling processes.

Hence, it can be concluded that, due to the impact on the environment of the selected physical separation processes, electrostatic separation was shown to be the best option for WPCB recycling. While the GWP for the entire physical recycling of WPCB, including the comminution processes, is 2.147 kg CO_{2eq}/kg of WPCB and this is mainly due to energy consumption for the electricity and, to a lesser extent, the use of liquid nitrogen before grinding. Although, its use does still not allow for full metals recovery, i.e. the extraction of metals in the form of pure and fully liberated particles. While the result is a mixture of metals from which individual metals can only be obtained by chemical processing. Physical recycling separates plastics that can be utilized further, but they make up 70% of the total mass of WPCB. As a result, approximately 70% less material would go to chemical extraction by leaching.

Currently, WPCB recycling methods are often based only on chemical leaching methods such as hydrometallurgy or by thermal processing by pyrometallurgy. These methods do not involve pretreatment to remove plastics, resulting in significant greenhouse gas emissions and waste generation, as well as the consumption of energy and reagents [40]. For comparison, GPW for chemical recycling of WPCB by hydrometallurgical method using glycine solution as an environmentally friendly main leaching agent is 12.4 kg CO_{2eq}/kg WPCB [41]. Assuming that in the case of physical recycling 70% less mass would go to hydrometallurgy, the GPW for it would be lower by 8.66 kg CO_{2eq}/kg WPCB. In this way, the GPW for physical recycling is compensated for and the plastics generated in it can be used to produce other consumer goods. As a result, hydrometallurgy recycling can also produce less waste and use fewer leaching reagents.

The remaining midpoint categories shown in Figure 4b follow trends similar to GPW, with flotation, shaking table, and cyclofluid separator accounting for approximately 70% of total impacts. The exception is ozone depletion, for which the shaking table is close to 50% burden.

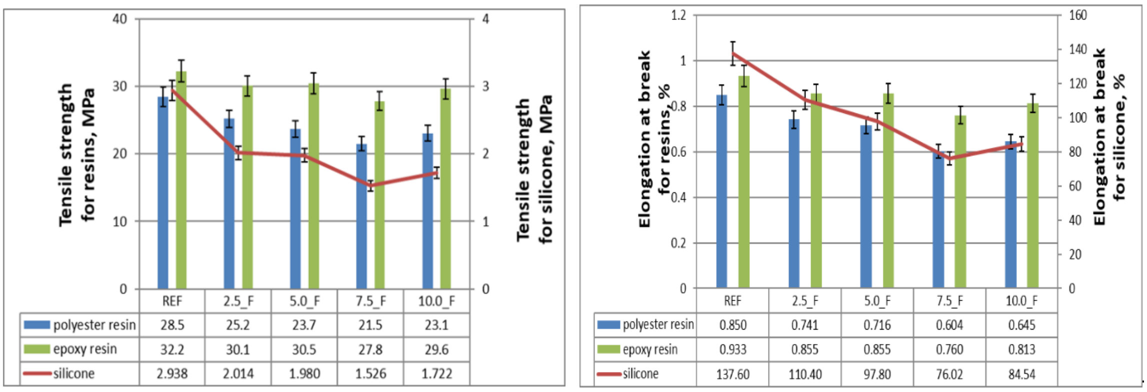
3.3. Strength Parameters of Composite Materials

3.3.1. Tensile Tests of Composite Materials

The results of mechanical tests of composite materials obtained by adding plastic powder obtained from recycled WPCB to the matrix and for pure epoxy, polyester resins and silicone (REF) are presented for tensile strength in Figure 5a (for silicone and resins) and elongation at break in Figure 5b (for silicone and resins).

For silicone used as a matrix, the highest tensile strength (2.94 MPa) was found in a sample made of material without filler. In the case of silicone-based composite materials with the addition of PCB powder, the tensile strengths were lower than those of the reference samples (REF). Tensile strength values slowly decreased with increasing filler content in the composite. The lowest value was measured for samples with a PCB powder content of 7.5%. For the sample with such a powder content, the tensile strength was 1.53 MPa and was lower than the reference value by 1.41 MPa. This value was 48% lower than for the sample with the highest tensile strength measured for the reference sample (2.94 MPa).

Tensile strength also decreased when powder obtained from recycled printed circuit boards was added to polyester and epoxy composites. The lowest value was also obtained for the samples with the addition of powder in the amount of 7.5%. This value was 21.5 and 27.8 MPa for the polyether and epoxy resins, respectively, while for the reference samples it was 28.5 and 32.2 MPa. Therefore, the tensile strength values were lower than the reference value by only about 15% for polyester resin and 13% for epoxy resin.

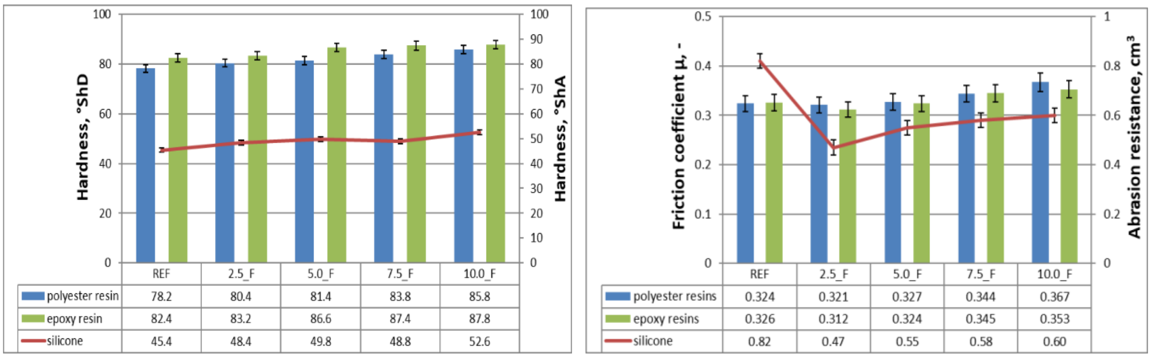


	REF	2.5_F	5.0_F	7.5_F	10.0_F
🟦 polyester resin	28.5	25.2	23.7	21.5	23.1
🟩 epoxy resin	32.2	30.1	30.5	27.8	29.6
🟥 silicone	2.938	2.014	1.980	1.526	1.722

(a)

	REF	2.5_F	5.0_F	7.5_F	10.0_F
🟦 polyester resin	0.850	0.741	0.716	0.604	0.645
🟩 epoxy resin	0.933	0.855	0.855	0.760	0.813
🟥 silicone	137.60	110.40	97.80	76.02	84.54

(b)



(c) (d)

Figure 5. Test results of strength parameters of composite materials; **a)** tensile strength for epoxy and polyester resins as a matrix in MPa and for silicone as a matrix in MPa, **b)** elongation at break for epoxy and polyester resins as matrix in % and for silicone as matrix in %, **c)** Hardness for epoxy and polyester resins as matrix in °ShD and for silicone as matrix in °ShA, **d)** friction coefficient for epoxy and polyester resins as matrix and abrasion resistance for silicone as matrix in cm$^{3.}$

For the elongation in the break tests, the strength trends were similar. Reference samples (without filler) with silicone as a matrix showed an elongation at break of 137.6% (Figure 5b), while filled samples showed a lower elongation at break. The lowest value was recorded for samples containing 7.5% of PCB powder (76.02%). This value was more than 45% lower than for the reference sample.

Similar trends in elongation were observed in break tests for samples with polyester and epoxy resins as the matrix. A slow reduction in the elongation at break is observed for the samples with increasing amounts of added powder. Again, the smallest value was obtained for samples with the addition of powder at the level of 7.5% and was 0.604 and 0.760% for polyester and epoxy resins, respectively. For comparison, the elongation at break values for the samples without the addition of filler (REF) were 0.850 and 0.933% for the polyester and epoxy resins, respectively (Figure 5b). The decrease in this parameter ranged from about 30% to about 18% for polyester and epoxy resins and was lower than for samples in which silicone was used as a matrix.

In the case of samples with fillers, for all matrixes used, both tensile strength and elongation at break were the highest for samples with the lowest filler content (2.5%). After that, the parameter values slowly decreased. For samples filled with powder in the amount of 2.5 and 5%, the values of tensile strength and elongation at break were similar. Generally, the 7.5% and 10% powder samples had lower tensile strength and elongation at break than the 2.5% and 5% powder filler samples.

For the epoxy resin used as the matrix, much smaller changes in the analyzed parameters were observed than for the polyester resin, when the content of the powder obtained from the recycling of PCBs increased.

3.3.2. Hardness Tests of Composite Materials

The hardness of the tested materials, both for the matrix in the form of silicone and the matrix in the form of resins, increases with increasing amount of powder added from PCB recycling (Fig. 5c). This is especially noticeable in the case of silicone. With 10% powder addition, the hardness increases from 45.4^{o}ShA for the reference sample (REF) to 52.6^{o}ShA. For polyester and epoxy resins, for the addition of powder in the amount of 10% by weight, the hardness increases from 78.2 for the reference sample (REF) to 85.8^{o}ShD and from 82.4 (REF) to 87.8^{o}ShD, respectively.

The hardness of the resin-prepared composites was more than two times higher than that prepared on a silicone matrix, which could be expected.

3.3.3. Abrasion Tests of Composite Materials

The results of the tests to evaluate the friction coefficient and abrasion resistance for resins and silicone as a matrix are presented in Figure 5d. The friction coefficient value for pure polyester resin was 0.324 and the epoxy resin 0.326. These two values should be treated as equal. Similar trends were observed for both polyester resin and epoxy resin. For both resins, the lowest friction coefficient value was recorded for composite materials containing 2.5% filler. Then, as the filler content increased from 5 to 10%, an increase in the friction coefficient was observed. The highest friction coefficient was observed for composites containing 10% filler for epoxy resin and polyester resin.

The highest value of abrasive wear was observed for the REF sample (0.82 cm^3). Material 2.5_F (0.47 cm^3) has the lowest abrasion resistance value among all tested materials. This value is 43% lower than the REF value. For composite materials with higher filling values, it was observed that with increasing filler content, the abrasion resistance value increases. The abrasion resistance values for materials 7.5_F and 10_F can be treated as equal.

3.4. Phytotoxicity Tests

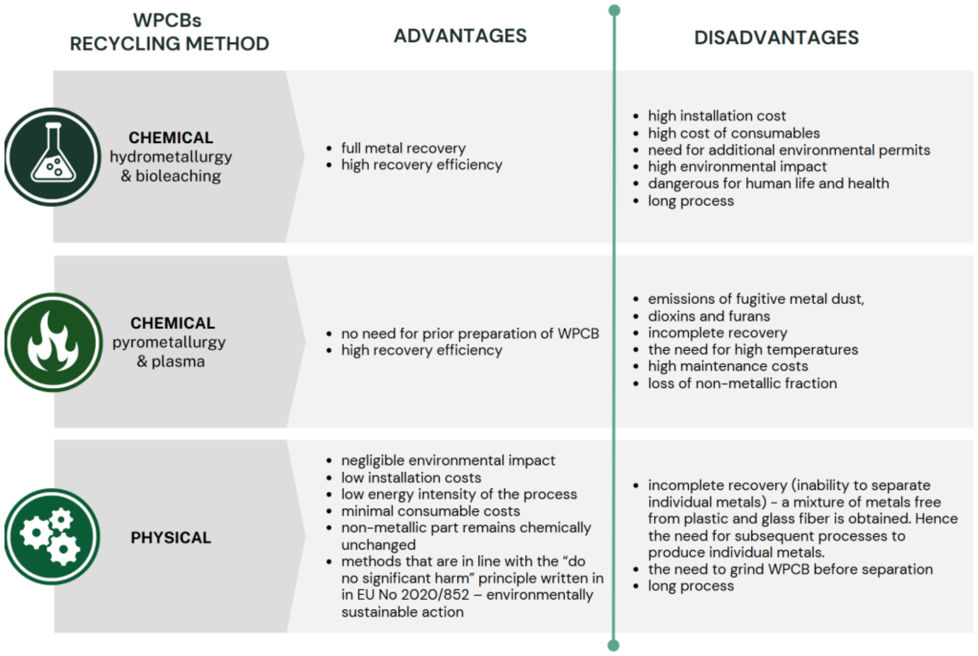
On the basis of phytotoxicity tests, it was found that all plants germinated 5 days after sowing. Importantly, after 14 days, no inhibition of root or stem growth was observed. In contrast, in most cases, an increase in root and stem length was observed in plants watered with eluate compared to plants watered with distilled water. This is natural plant behavior. Distilled water does not contain nutrients, while some nutrients may be available in the eluates. This happens provided that they do not contain toxic substances. In general, the increase in root length was greater than the increase in stem length of the studied plants and amounted to 163%, 218% and 192% for cucumber, mustard and wheat, respectively. The increase in stem length for these plants was 131%, 139% and 94%, respectively. Only in the case of the length of the wheat stem was a growth inhibition of 6% observed. Therefore, it can be concluded that the tested plants were not sensitive to substances leached from plastic. In contrast, the substances presented in the eluates promoted the growth of the tested plants.

4. Discussion

As a result of the use of physical recycling, three products are created: concentrate, a mixture of metals, waste, plastics with a small amount of glass fibres, and an intermediate-middling's product, i.e., particles containing a mixture of plastics and metals. The concentrate obtained, due to its high purity and valuable element content, can be sold to metal producers or processors and represents a significant financial benefit to the WPCB recycler. Given the current metal prices and the average metal content of WPCB, it is estimated that in one tonne of WPCB, the price of the metals contained is more than $91 800. This consists of the price of Au - $84,556, Ag - $1,928, Cu - $1,770, Pd - $1,532, Sn - $1,139, Pt - $454, Ni - $305, Al - $51, Pb - $39, Zn - $30. Metal prices in WPCB were estimated based on: [11,13]. This is, of course, the value specified for unprocessed WPCBs. It should be noted that, despite the small quantities, gold is the key metal determining the economic viability of WPCB recycling. However, since the metal content of WPCB varies depending on the producer, the year of production, and the technology used, recycling of other metals should not be ruled out as a predictable source of financial benefit.

In Poland, the annual weight of the collected WPCB per 1 million inhabitants is estimated to be about 27 Mg (mass without sockets, resistors, transistors, processors, RAM, and other elements on the surface of the PCB), with an estimated value of metals of about 2.5 million USD/year (the mass balance of WPCB and the estimated value of metals and plastics in WPCB were determined on the basis of data from three companies located in the Silesian Voivodeship dealing with WEEE collection). For example, in the Silesian Voivodeship, where 4.4 million people live, potentially about 120 Mg of WPCB are collected annually, and the value of the metals can be estimated at USD 11 million. Therefore, this represents a major opportunity for local WEEE recyclers. For the whole of Poland, the value of metals is even $96 million. Because of the high purity of the products, the valuable metal mixture can be sold to local metal smelters, while the product consisting of the nonmetallic parts can be used as a filler for the production of composite materials based on polymer materials with the addition of fillers from powder derived from WPCB recycling. Due to the fact that the addition of recycled plastics improved abrasion resistance and hardness, especially for epoxy resins as a matrix, these materials could be used in the production of resin floors and composite boards. Importantly, consumer products manufactured in this way are not toxic to plants. Composite materials with the addition of recycled plastics have slightly lower tensile strength. Therefore, they should not be used in products that are exposed to such impacts. The annual mass of recycled plastics for the Silesian Voivodeship in Poland reaches almost 20 tonnes per million inhabitants per year. This is a significant amount and could mean that fewer raw materials will need to be purchased to produce new consumer goods such as resin flooring and composite decking.

As a result, recycling using physical separation methods is relatively easy to perform and does not require sophisticated technical equipment or financial outlays. It also has a small impact on the environment (Figure 6). One of the most important processes preceding the separation methods is comminution, whose purpose is to remove metals from the rest of the WPCB. Improper comminution of the WPCB results in a certain randomness in the physical properties of the grains, which is the main reason for incorrect separation and thus the migration of the grains into the incorrect products. Therefore, release of metals from the remaining non-metallic fractions is crucial to achieve high efficiency in separation and high selectivity. WPCB grinding usually involves two steps. The first involves reducing the size of the WPCB using a disintegrator to dimensions not exceeding approximately 5 cm [42,43]. The next step is to grind the precrushed WPCB using hammer mills [44–46] or knife mills [43], preferably at cryogenic temperatures [21]. The resulting WPCB powder is the feed for the separation processes. Separation in the electrostatic separator occurs as a result of differences in the electrical surface charge of various particles and separation is carried out without the use of process water. Because of its high efficiency in separation and selectivity and the smallest environmental footprint, electrostatic separation is the best option for separating metals from plastics.



WPCBs RECYCLING METHOD	ADVANTAGES	DISADVANTAGES
CHEMICAL hydrometallurgy & bioleaching	• full metal recovery • high recovery efficiency	• high installation cost • high cost of consumables • need for additional environmental permits • high environmental impact • dangerous for human life and health • long process
CHEMICAL pyrometallurgy & plasma	• no need for prior preparation of WPCB • high recovery efficiency	• emissions of fugitive metal dust, • dioxins and furans • incomplete recovery • the need for high temperatures • high maintenance costs • loss of non-metallic fraction
PHYSICAL	• negligible environmental impact • low installation costs • low energy intensity of the process • minimal consumable costs • non-metallic part remains chemically unchanged • methods that are in line with the “do no significant harm” principle written in in EU No 2020/852 – environmentally sustainable action	• incomplete recovery (inability to separate individual metals) – a mixture of metals free from plastic and glass fiber is obtained. Hence the need for subsequent processes to produce individual metals. • the need to grind WPCB before separation • long process

Figure 6. The main advantages and disadvantages of WPCB recycling methods, based on: [15,21,47,48].

Currently, in many countries, WPCB recycling is carried out exclusively by chemical methods, i.e. hydrometallurgy, pyrometallurgy, and/or biohydrometallurgy. Their use generally requires special permits and increased work to prevent environmental degradation. It should also be noted that the chemical transformations during WPCB recycling processes result heavy cross-

doi:10.20944/preprints202601.0216.v1

contamination of non-metallic parts, which in turn creates additional waste (Figure 6). In contrast to the above, methods based on physical separation have less impact on the natural environment [21,25]. These processes do not use hazardous substances that cause significant emissions into the atmosphere as a result the carbon footprint for physical recycling is quite small. By using physical separation methods for recycling allows the recovery of valuable substances (metals) and the use of the non-metal fraction (glass fibre and plastics) without cross-contamination for the production of new products. Thus, physical methods provide opportunities for close to zero-waste recycling of WPCB. In addition, the non-metallic part of the ground WPCB laminate is not chemically altered, creating a wide range of application possibilities, for example, for the production of composite materials. Figure 7 shows the flow diagram of the eco- efficient WPCB recycling technology with mass and energy flows presented using the Sankey diagram. However, it should be noted that the use of physical methods does not provide an opportunity for full recovery, i.e. extraction of metals in the form of pure metals. Since a mixture of metals is created from which individual metals can be further obtained by the follow up chemical treatment. Due to the well-developed metallurgical industry in Poland, it is possible to transfer (sell) such metal mixture from PCB recovery to existing metal production or recovery plants.

During such processing there is about 2.4% of the intermediate product of middling's quality and because of such a small quantity of this product, it is possible to process it by bioleaching using Acidithiobacillus ferrooxidans bacteria. Although, bioleaching is characterized by slow kinetics, and is generally a low-yielding method but it is still more environmentally friendly method than chemical leaching (hydrometallurgy). Alternatively, this material cab be sent directly to chemical leaching, and because of its low mass, it should have a relatively smaller negative impact on the environment because less chemicals will be used for smaller quantity of this fraction.

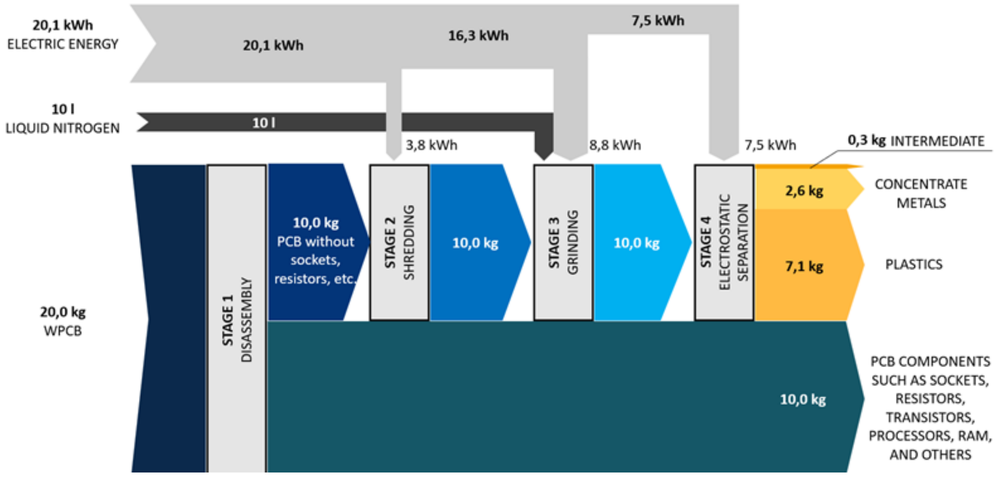


Figure 7. Technology diagram with mass and energy flows presented using a Sankey diagram.

In order for a company to be able to recover metals in accordance with the presented method, it is necessary to additionally consider investment, labour, and administrative costs, including taxes, as well as process costs related to supplying machines and devices with electricity and the use of liquid nitrogen, as well as the management of other products generated during recycling. Table 1 shows the input used in the LCA of each stage, along with the unit prices of the input, and the calculated consumable costs required to process 1 kg of WPCB. The calculations were made for devices operating on a laboratory scale; hence, the consumable costs are high. Without these costs, the most expensive stage is cryogenic grinding, mainly due to the price of liquid nitrogen.

Table 1. Economic analysis of processing 1 kg of WPCB including operation and maintenance costs.

Input	Unit price, USD	Consumption / Price, USD		
		Shredding	Cryogenic grinding	Electr. separat.
Electricity (Sas, 2025), kWh	0.012	0.005	0.011	0.009
Liquid nitrogen, litres	0.59		0.59	
Consumable costs		0.49[1]	1.02[2]	<0.001[3]
Price excl. consumables costs, USD		**0.005**	**0.601**	**0.009**
Total price, USD		0.495	1.621	0.009

[1]cutting shaft elements (36 pcs/2 Mg$_{wpcb}$). [2]mill knives (7pcs/100kg$_{wpcb}$). [3]electrode (1pc/5 Mg$_{WPCB}$).

5. Conclusions

Physical WPCB recycling technology using shredding, cryogenic grinding, and electrostatic separation is in line with the principles of circular economy and sustainable production. It is a pretreatment method that complements conventional chemical recycling methods.

The global warming potential for the entire physical and chemical process is lower by about 70%, which is due to the smaller mass of input material going to the downstream metallurgical processes. Because of this, the entire recycling process generates less waste and uses fewer leaching agents (for hydrometallurgy). In addition, plastics produced through physical recycling can be used to produce consumer goods and thus even bring financial benefits, similar to selling the metal mixture itself.

Author Contributions: Conceptualization and formulation of theses, T.S.; Formal analysis and conclusions, T.S.; methodology, T.S., D.F, M.M. and U.K.; physical separation, D.F, T.S.; LCA analysis, U.K., T.S.; economic analysis, T.S.; testing of composite materials, M.M., D.F.; phytotoxicity tests, T.S. and D.F.; Investigation, T.S. and D.F.; writing - original draft preparation, T.S., D.F. and M.M.; writing - review and editing, T.S. and M.H.; visualization, T.S., D.F., M.M., M.H. and U.K. All authors have read and agreed to the published version of the manuscript.:

Funding: Publication supported by the "Excellence Initiative – Research University" program. Silesian University of Technology, Grant No 06/050/SDU/10-27-01.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: The original contributions presented in this study are included in the article. Further inquiries can be directed to the corresponding author.

Conflicts of Interest: The authors declare no conflict of interest.

Appendix 1. Life Cycle Inventory to Process 1 kg WPCB

Stages			Energy consumption, kWh/kg$_{WPCB}$	Water consumption, dm^3/kg$_{WPCB}$	Liquid nitrogen consumption, dm^3/kg$_{WPCB}$	Flotation reagent consumption, mg/kg$_{WPCB}$
Shredding			0.375			
Grinding		Cooling			1	
		Grinding	0.880			
Separation	Electrostatic Separator		0.750			
	Shaking Table	Separation	0.333	150		
		Drying	1.200			
	Cyclofluid Separator	Separation	0.093	50		
		Drying	1.200			

Flotation Machine	Separation	0.552	30	3140
	Drying	1.200		

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