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PROCESS IMRPOVEMENT ON RECOVERY OF RAW MATERIALS FROM MULTILAYER COMPOSITES

Gytis MILIAUSKAS, Gintaras DENAFAS
Kaunas University of Technology
K. Donelaičio g. 73, Kaunas, Lithuania
e-mail: g.miliauskas@ktu.edu

Abstract
At end of life, waste PCBs (WPCBs) represent both a valuable resource—containing copper, nickel, precious metals, and polymeric materials—and an environmental hazard if landfilled or incinerated. This work develops and optimizes a modular, closed-loop recycling line that (1) delaminates epoxy resin from fiberglass and embedded metal via dimethyl sulfoxide (DMSO) extraction, comparing conventional thermal swelling with microwave-assisted processing to maximize resin yield, preserve fiber integrity, and assess solvent recyclability; (2) mills the residual PCB fragments under a design-of-experiments framework to tune particle size distribution (2–10 mm) and energy use, including evaluation of process-control additives; (3) fractionates milled material through sequential magnetic, eddy-current, electrostatic, and density separation, optimizing separation thresholds and recovering ferrous, non-ferrous, and non-metallic streams; (4) conducts hydrometallurgical extraction on the mixed metal fraction, including alkali pre-treatment, acid leaching (with oxidant enhancement), solvent-extraction, and electrowinning to produce high-purity metal products; and (5) scales the optimized processes from laboratory (2 L) to pilot (200 L) stirred reactors under geometric similitude. Finally, mass and energy balances, closed-loop solvent and water recycling. Key technical uncertainties—including solvent–resin selectivity, milling energy–liberation trade-offs, leaching kinetics in complex residues, scale-up hydrodynamics, and solvent recovery losses—are systematically investigated. The resulting platform offers a viable path toward circular, sustainable PCB recycling with minimized waste and maximized resource recovery.

Key words: *circular economy, organic extraction, PCB recycling, hydrometallurgical extraction, prototype design.*

Multilayer Composites (MC) are widely used in all electronic devices, their composition is rich in materials, exclusive electrical, optical, insulating, and mechanical properties [1]. One of the MC types is Printed Circuit Boards (PCB), which are the mediums used to connect components to one another inside the PCB layers. PCB base materials consist of three structural components: resins, reinforcement layers (glass fiber), and the conductive foil [2]. Brominated epoxy resins are the most commonly used material for adding flame retardancy to the final product.

Many other resin systems are also available, such as epoxy blends with other types of resins [3]. PCBs can have different designs (Fig. 1), however the function of all PCB models is the same - to support a number of mounted electronic components and provide electrical connection as well as insulation between the components if needed. PCBs can be single-sided (one copper layer), double-sided (two copper layers on both sides of one substrate layer), or multi-layer (outer and inner layers of copper, alternating with layers of substrate).

In the last decade, the recycling of waste printed circuit boards (WPCBs) has increased for 2 reasons:

1. WPCBs contain a considerable amount of metallic and non-metallic components such as copper (Cu), iron (Fe), tin (Sn), nickel (Ni), lead (Pb), zinc (Zn), silver (Ag), gold (Au), palladium (Pd), and fiberglass and resins. Thus WPCBs could be used as a source of valuable raw materials.
2. WPCBs are classified as hazardous waste. These substances can be released as a result of landfilling or treating with traditional methods such as incineration. [4].

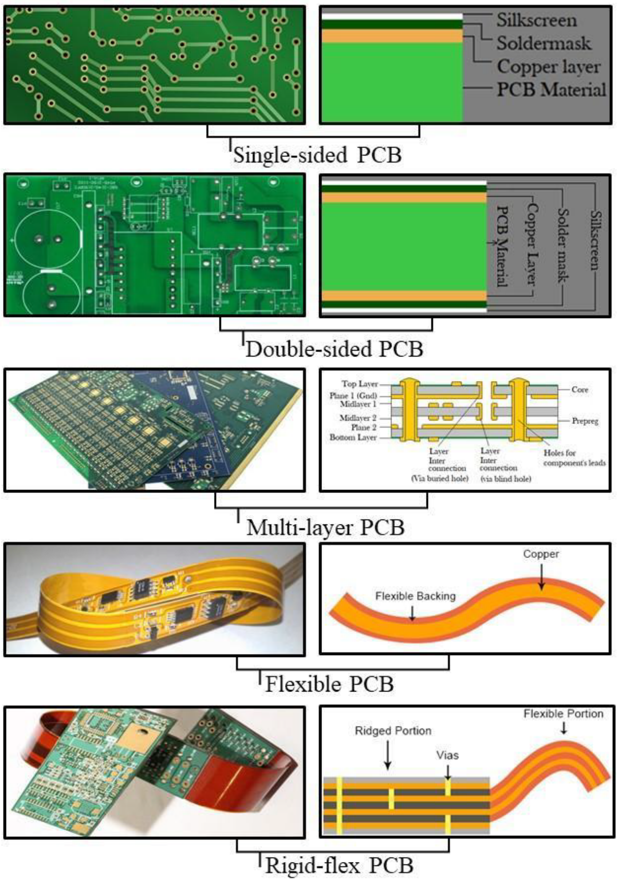

Figure 1. *Examples of PCB types [5],[6],[7]*

Removing harmful materials safely is the key for successful recycling. Removing toxic components fron resins using solvent-based delamination was shown to be the most efficient in terms of environmental impact and cost when compared to other techniques such as thermal treatment and crushing. Other methods have disadvantages in terms of power consumption, release of gases, and recycling rate. In order to be more efficient, a number of investigations have been performed on the delamination of WPCBs by solvents and/or water-soluble oxidants. The most important reason for employing organic solvents is to dissolve resins, separating metal and non-metal components of WPCB's. Using the solvent delamination technique, many components and layers of a full-size WPCB can be seperated and recovered. There are technologies that combine three treatments:

chemical, ultrasonic and mechanical, with the use of dimethylacetamide to dissolve epoxy resin [8]. Delamination with the organic solvents can only be performed at higher temperatures, so microwave medium can greatly enhances the process efficiency, increasing resins dissolution up to 2 times compared to just thermal treatment [9].

We are developing and optimizing PCB recycling process/line (Fig. 2). First step is solvent-based (DMSO) delamination process to separate the resin from the fiberglass and embedded metal. Specifically, we will compare thermal extraction and microwave-assisted extraction by swelling PCBs in DMSO to maximize resin yield and preserve fiber integrity. Out of all the organic solvents, we have chosen DMSO due to it‘s low toxicity (compared to other solvents that were researched) and easy access. We will then investigate the DMSO recyclability by monitoring viscosity, chemical purity, and extraction performance over multiple extractions. There are certain unknowns regarding our process: solvent–resin selectivity across different PCB laminates; kinetics and energy consumption in microwave vs. thermal extraction; limits to solvent reuse before extraction efficiency decreases.

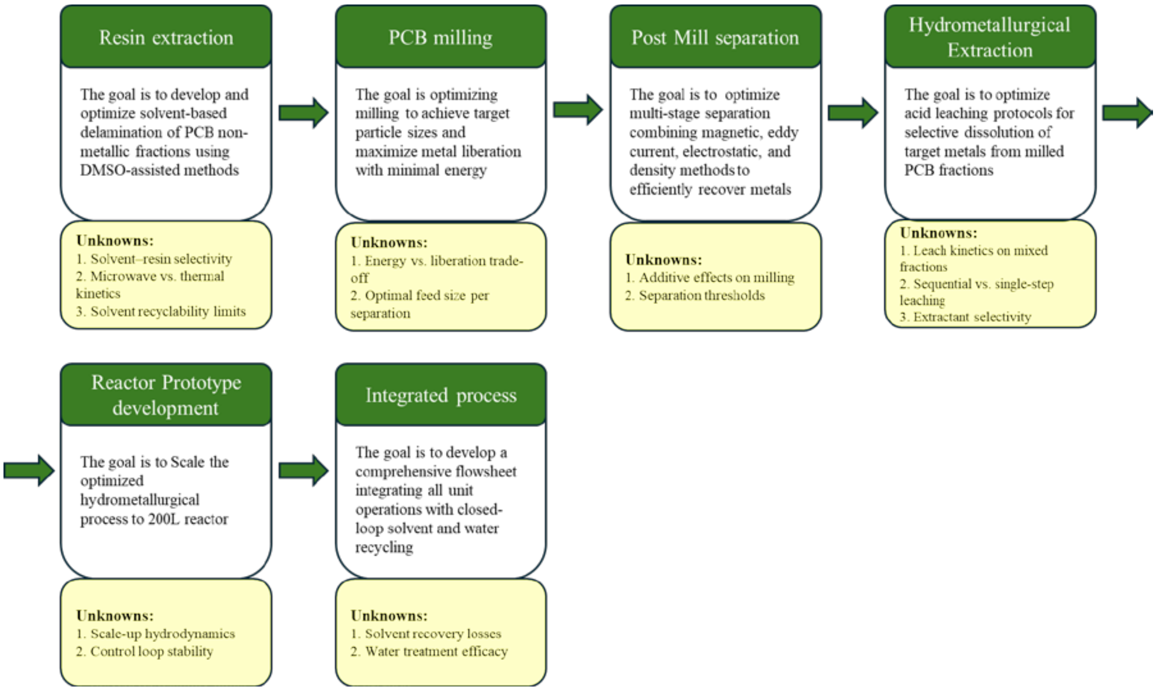

Figure 2. *Project development stages and uncertainty areas*

Following resin extraction and PCB swelling, we will mill the residual PCB fragments in a controlled milling setup. A design-of-experiments (DOE) approach will be used to tune milling: rotational speed, time, and the addition of other material (e.g., silicone oil, ceramic beads, aluminium powder) to achieve target uniformed particle size distributions (2–10 mm) with minimal energy input. During this step, we will investigate the relationship between specific energy input and degree of metal liberation vs. fines generation; the optimum feed size distribution for downstream separation; effects of milling additives on particle morphology and uniformaty.

Post-Mill Separation will be done by employing these steps—magnetic, eddy-current, electrostatic, and density separation—designed to fractionate ferrous, non-ferrous, and non-metallic streams. They will be used all together or in predetermined combination. We will optimize field strengths, rotor and belt speeds, voltages, and media densities to maximize recovery rates of each fraction. The biggest challanges are the cut-off thresholds for conductivity and density that govern separation efficiency and how residual resin or process control agents influence separation performance.

The last stage in developmental stage is the non-metallic and mixed metal fraction pre-treatment to remove surface coatings, followed by acid leaching (sulfuric/nitric acids, with and without oxidants) to dissolve target metals (Cu, Ni, precious metals). We will model leaching kinetics as a function of pulp density, temperature, and oxidant concentration, and then develop solvent-extraction process and electrowinning cells to recover valuable metals. This stage is the most complicated as the PCBs have already passed all the separation stages. The extraction selectivity for multi-metal mixtures under realistic conditions is currently under investigation and proves to be the hardest part (combining economical and safe process).

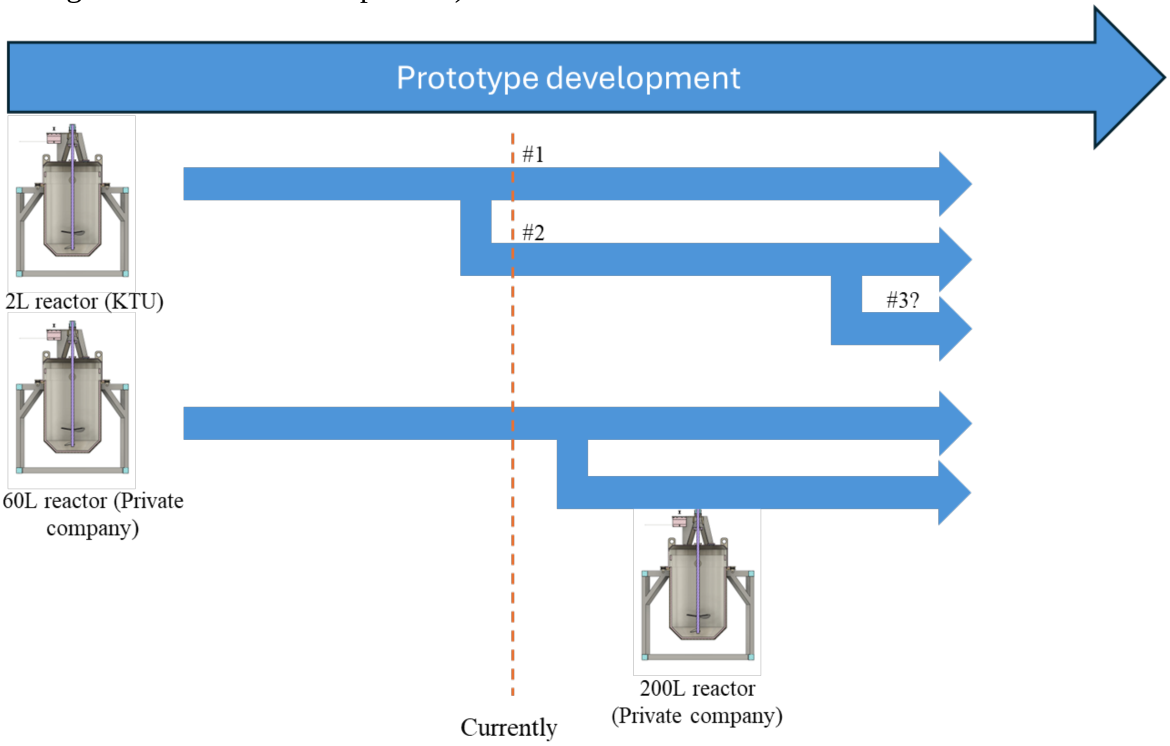

Figure 3. *Current state of prototype development*

Currently, we have developed 2 prototype reactors: 2L reactor at Kaunas University of Technology (KTU) and 60L at private entity. KTU is undergoing a testing of a third prototype model and it's testing. To translate laboratory results to a continuous process, we will design and test reactors at laboratory (2 L) and pilot (60 L) scales under geometric similitude. We will integrate programmable logic controller (PLC) systems for real-time monitoring and control of pH, temperature, and other sensors to evaluate the upscale of the PCB recycling process. The private entity engaged in 200L reactor design and manufacturing. These steps will encounter usuual scale-up hydrodynamics challanges - mixing times, power-to-volume scaling, fluid dynamics, and etc.

As seen in other research, the reduction of PCB is key in „releasing" the metal particles from PCB. Grinding and milling PCB to small particles increases the recovery rate, but also removes the valuable metals from the process (those metals stick to surfaces or lost during the process) [10]. Our studies showed that diffrent delamination process of PCBs have vastly different outcomes (Picture 4), even though A1 and A2 are the same PCB type. Delamination of PCBs might reduce the need for physical milling to small particles, thus eliminating the possible lost of valuable material and having a better economical payback.

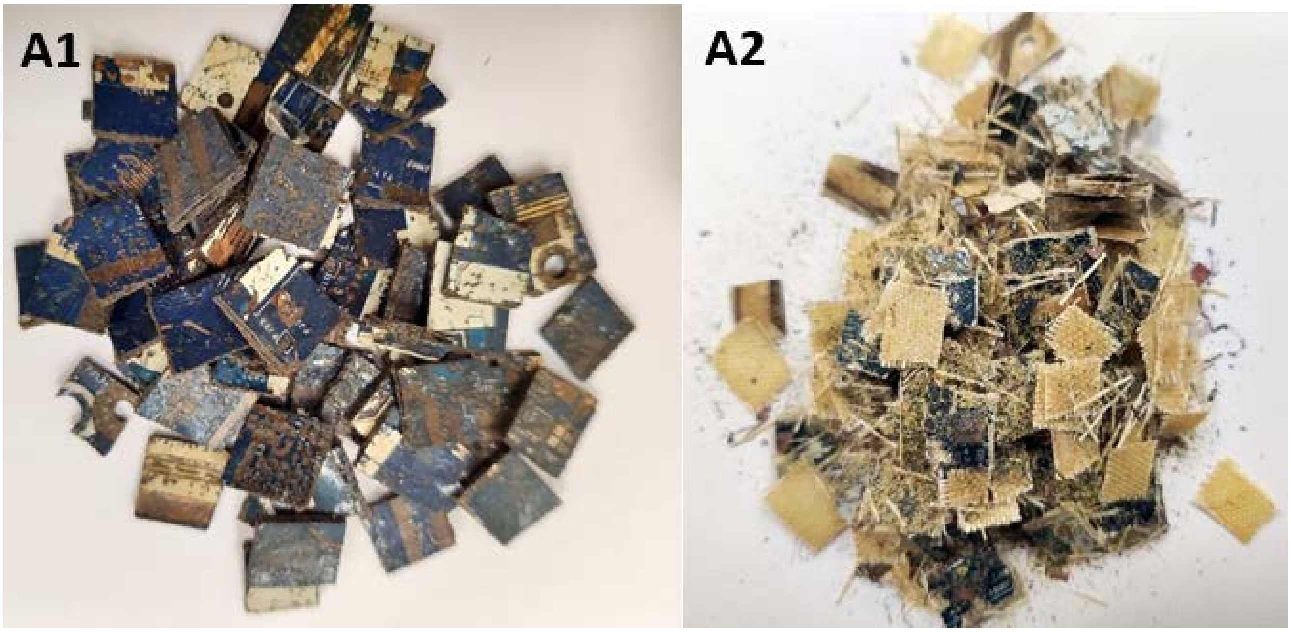

***Figure 4**. A1 - Sample after heating in DMSO in ultrasound bath at 70°C for 56 h. A2 - Sample after heating in DMSO at 140°C for 6 h*

Moveover, recovered resins (Fig. 5) could have a secondary use. Recovered resins have polymeric chains, that can be broken down into monomer, chemically alterted with an active group and reused in different application. For example resin 3D printing, where only the structure of the 3D model is important, not the other properties.

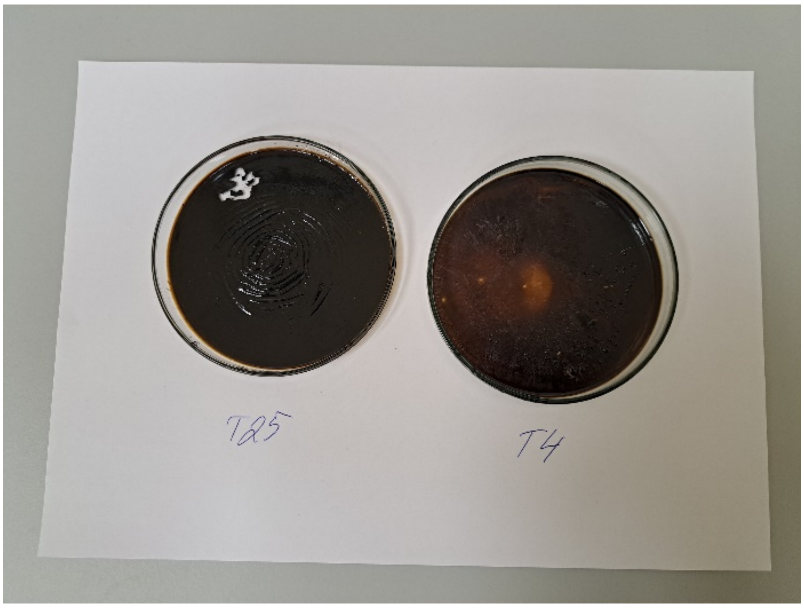

***Figure 5.** Extracted resins from RAM PCBs*

Conclusions

This work establishes first steps for a modular framework for the sustainable recycling of waste printed circuit boards (WPCBs) using solvent-based delamination, controlled mechanical treatment, multi stage physical separation, and hydrometallurgical recovery of metals. It is demonstrated that organic solvent extraction using dimethyl sulfoxide (DMSO) is an effective alternative to traditional thermal and mechanical methods for separating resins from fiberglass and embedded metals. The recyclability of DMSO further supports its use in a closed-loop system, aligning with circular economy principles.

Optimization of the milling process will enable precise control over particle size distribution and energy consumption while minimizing metal losses. The application of magnetic, eddy-current, electrostatic, and density separation effectively fractionates the processed material into ferrous, non-ferrous, and non-metallic streams with high purity and recovery rates. The hydrometallurgical extraction of mixed metal residues—via alkali pretreatment, acid leaching, solvent extraction, and electrowinning—allows selective recovery of high-purity metals under controlled operating conditions.

Scale-up from laboratory (2 L) to pilot (60–200 L) reactors under geometric similitude demonstrates the technical feasibility of translating batch laboratory processes into continuous, automated recycling operations. Integration of programmable logic controllers (PLC) for real-time monitoring and process control ensures reproducibility, safety, and adaptability of the developed system.

This study provides first steps towards an experimentally validated pathway toward a circular, energy-efficient, and environmentally responsible PCB recycling process. The approach minimizes secondary waste, maximizes material recovery, and lays the groundwork for industrial-scale implementation of solvent-based delamination and hydrometallurgical recovery technologies. Future research should focus on long-term solvent stability, kinetic modeling of metal extraction from complex residues, and the valorization of recovered polymeric fractions for high-value applications.

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УДОСКОНАЛЕННЯ ПРОЦЕСУ ВІДНОВЛЕННЯ СИРОВИНИ З БАГАТОШАРОВИХ КОМПОЗИТІВ

Гитіс МІЛЯУСКАС
Каунаський технічний університет
K. Donelaičio g. 73, Kaunas, Литва

Гінтарас ДЕНАФАС
Каунаський технічний університет
K. Donelaičio g. 73, Kaunas, Литва
https://orcid.org/0000-0002-6834-1026

Анотація

У кінці життєвого циклу відпрацьовані друковані плати (WPCBs) становлять як цінне джерело ресурсів — міді, нікелю, дорогоцінних металів і полімерних матеріалів — так і екологічну загрозу у разі захоронення або спалювання. У цій роботі розроблено та оптимізовано модульну замкнену лінію переробки, яка: (1) здійснює деламінацію епоксидної смоли від скловолокна та вбудованих металів шляхом екстракції диметилсульфоксидом (DMSO), порівнюючи традиційне термічне набухання з мікрохвильовою обробкою з метою максимізації виходу смоли, збереження цілісності волокна та оцінки можливості повторного використання розчинника; (2) подрібнює залишкові фрагменти плат у межах експериментального дизайну з налаштуванням розподілу за розміром частинок (2–10 мм) та енергоспоживанням, включаючи оцінку технологічних добавок; (3) фракціонує подрібнений матеріал за допомогою послідовного магнітного, вихрострумового, електростатичного та щільнісного розділення з оптимізацією порогів сепарації та виділенням феромагнітних, кольорових і неметалевих компонентів; (4) виконує гідрометалургійну екстракцію зі змішаної металевої фракції, включаючи лужну підготовку, кислотне вилуговування (з використанням окисника), екстракцію розчинником та електровідновлення для отримання металопродуктів високої чистоти; (5) масштабує оптимізовані процеси з лабораторного рівня (2 л) до напівпромислового (200 л) у змішуваних реакторах із дотриманням геометричної подібності. У підсумку виконано масові та енергетичні баланси, передбачено замкнені цикли використання розчинника та води. Системно досліджено ключові технічні невизначеності, включаючи селективність розчинник–смола, компроміси між енерговитратами на подрібнення та вивільненням компонентів, кінетику вилуговування з комплексних залишків, гідродинаміку масштабування та втрати при регенерації розчинника. Отримана платформа є перспективним рішенням для циркулярної та стійкої переробки друкованих плат із мінімізацією відходів і максимальним відновленням ресурсів.

Ключові слова: циркулярна економіка, органічна екстракція, переробка друкованих плат, гідрометалургійна екстракція, проєктування прототипу.