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Review

# Dismantling of Printed Circuit Boards Enabling Electronic Components Sorting and Their Subsequent Treatment Open Improved Elemental Sustainability Opportunities

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**Abstract:** This critical review focuses on advanced recycling strategies to enable or increase recovery of chemical elements present in waste printed circuit boards (WPCBs). Conventional recycling involves manual removal of high value electronic components (ECs), followed by raw crushing of WPCBs, to recover main elements (by weight or value). All other elements remain unrecovered and end up highly diluted in post-processing wastes or ashes. To retrieve these elements, it is necessary to enrich the waste streams, which requires a change of paradigm in WPCB treatment: the disassembly of WPCBs combined with the sorting of ECs. This allows ECs to be separated by composition and to drastically increase chemical element concentration, thus making their recovery economically viable. In this report, we critically review state-of-the-art processes that dismantle and sort ECs, including some unpublished foresight from our laboratory work, which could be implemented in a recycling plant. We then identify research, business opportunities and associated advanced retrieval methods for those elements that can therefore be recovered, such as refractory metals (Ta, Nb, W, Mo), gallium, or lanthanides, or those, such as the platinum group elements, that can be recovered in a more environmentally friendly way than pyrometallurgy. The recovery methods can be directly tuned and adapted to the corresponding stream.

**Keywords:** recycling; strategic metals; spectroscopy; X-rays imaging; image recognition; artificial intelligence



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## 1. Introduction

Our electronically dependent society creates a large and ever-increasing amount of electronic waste, which is reported to be the fastest increasing waste stream, with ~50 million metric tons generated globally in 2018 [1]. Despite the value of the materials it contains (~USD 60 Bn), only 20% of them are currently recycled [2]. Printed circuit boards (PCBs) represent on average 8% of this electronic waste, yet only about 10 out of the 60-plus chemical elements that they contain are routinely recovered and recycled [3–5]

The recycling of waste printed circuit boards (WPCBs) depends on both an informal sector focused on precious metals recovery using highly polluting approaches, and industrial processes based on the pyrometallurgy of centralized waste, treated in depreciated smelters to make use of the calorific value of the WPCB's 30 wt% epoxy resins [6]. Overall, only ~30 w% of the WPCBs' mass is truly recycled [7]. The rest is transformed into fumes or ends up mixed in the bottom or fly ashes. A more sustainable approach would require alternate processes that: (i) allow for improved elemental recovery; (ii) are more "green";

(iii) do not require centralizing waste over a wide geographic area, as waste transportation is becoming an increasing issue, which implies that new processes must be deployable for small waste volumes or geographic zones; (iv) avoid burning epoxy resins that can release toxic gases [8].

Toward that end, recent development and research favours a strategy based on first dismantling WPCBs [9–11] followed by efficiently sorting electronic components (ECs). This allows obtaining various fractions: (i) bare boards; (ii) solder; (iii) ECs sorted in elementally enriched subfractions. The goal is for each fraction, or subfraction, to have the simplest elemental composition possible, making them easier to reuse directly or recycle, and making it now possible to recover valuable metals [12] such as Ti, Ga [13] Ba, Ta, Nb, W, Lanthanides [14,15]. It should be noted that classically recycled metals such as Au, Ag or Cu are not covered in this review as their recovery is already extensively discussed in the literature. The objective of this review is indeed to shed light on other relevant chemical elements for which recycling opportunities could arise from the emerging technologies described here.

The PCB dismantling research effort is currently little considered in the USA but is very vibrant in the European Union and parts of Asia. It pushes for: (i) increased recovery yields; (ii) reduced CO<sub>2</sub> footprints; (iii) processes that are adapted to small volumes and reduced capacity to export waste and landfilling. Indeed, for many countries, options are currently limited as many cannot afford to build pyrometallurgy plants, exporting waste PCBs is becoming increasingly difficult, and landfilling is very costly due to the hazardous nature of such wastes. Some countries, such as Japan, show a real effort in improving the treatment and the importation of hazardous waste from developing countries [16]. Under the implementation of the Basel convention, the Japanese government wants to promote and develop the official recycling route to prevent illegal exports and treatments leading to environmental pollution.

It is anticipated that the dismantling approach will create new business models where, for example, the factory is brought to the waste instead of the waste to the factory, which is enabled by the containerization of processes.

## 2. Materials and Methods

To separate ECs from WPCBs, several methods can be used. Mechanical dismantling of WPCBs is the most common and straightforward way to obtain ECs, and can be performed manually or automatically for bigger scales. Other ways involve melting or chemically leaching the solder to release the ECs. These methods will now be reviewed. A qualitative comparison of their relative costs in terms of operating expenses (OPEX) and capital expenses (CAPEX) is given in Table 1.

### 2.1. Mechanical Dismantling

#### 2.1.1. Manual Dismantling

It is still common, mostly in the informal sector, to dismantle WPCBs manually, using chisels, hammers, and cutting torches, taking advantage of cheap labour or of non-professional and occasional gatherers. They usually focus on high-value components, such as aluminium heat sinks, batteries, inductances (copper coils), capacitors, Integrated Circuit dual in-line packages, Random-Access Memory (RAM) devices, Chip carriers, and Central Processing Units (CPUs) [17]. It is usually performed early in the WPCB recycling process. Components are then resold or sent to the corresponding treatment processes.

When this is performed by the informal sector, it is detrimental to the downstream licensed collecting and recycling industry since the remaining parts have little value, which significantly decreases the economic viability of recycling processes.

**Table 1.** Qualitative comparison of the different dismantling processes (+ cheap, ++ mid-priced, +++ expensive).

Process	CAPEX	OPEX	Advantages	Drawbacks
Manual dismantling	+	++++	Easy to implement Selective disassembling	Hard manual work Requires manpower Slow process Polluting
Surface cutting knife	++	+	Non-polluting High disassembly rate	WPCBs are treated one by one
Crude heating	++	+	Large capacity	Toxic emissions Heat damage ECs
Infrared radiators	++	+	High disassembly rate	Small volumes Heat damage ECs
Hot air heating	++	++	High disassembly rate Little maintenance Non-polluting	Low accuracy control Low energy efficiency
Solder bath heating	++	+++	High disassembly rate	Difficult to automate Toxic fumes emissions Dangerous working conditions
Hot fluid heating	++	++	High disassembly rate High thermal efficiency High solder recovery rate	Generating toxic waste fluids and fumes
Heated centrifugation	++	++	High solder recovery rate Solder elements separation	High temperature Heat damage ECs
Solder dissolution	+	++	Selective process	Requires further treatments Hazardous chemicals used
Hydrothermal and supercritical fluids treatment	+++	++	Target metals or resin No toxic product released Reusable reagents	Cannot target solder Requires further dismantling treatments
Epoxy resin treatment	++	++	Recover functional circuits Chemicals can be recycled	Need further dismantling process Hazardous chemicals used
Robotic Dismantling	+++	+	Combined dismantling and sorting Low manpower	Low throughput
Fragmentation by high voltage electric pulse crusher	+++	++++	High capacity Non-polluting	Low energy efficiency Expensive initial investment

### 2.1.2. Surface Cutting Knife Dismantling

One can find examples of processes in which ECs are automatically removed mechanically by a surface cutting knife or blade. In the Chinese patent CN105818189A [18] a circular saw is mounted parallel to a conveyor belt and cuts large ECs off the WPCBs (Figure S1, Supporting Information). This approach allows for the treatment of large quantities but leaves both some solders and electrical pins onto the bare board. It also requires careful monitoring and air purification to avoid exposing the workers to toxic powders or nanoparticles generated by the mechanical process.

## 2.2. Dismantling via a Heat Treatment

In this section, we only review reports on technologies that dismantle WPCBs by removing solder (mechanically, thermally, or chemically), hence recovering three fractions: solders, ECs, and bare boards.

### 2.2.1. Crude Heating

Crude homemade or commercial tools can be found that combine heating with mechanical treatment of WPCBs. Heat can be generated either electrically, by a gas torch, or wood fire. A temperature control or an exhaust gas purification unit is not always used, which may harm the workers and the local environment. There is little scientific literature on this as it is usually performed by small private operations, informal and/or

unlicensed companies. Some insight into the tools available can be achieved to some extent using patent records, but since a patent review on PCB recycling has been published recently [19], we will not develop it further here. A commercial example, however, is a tool that integrates some level of engineering control and is commercialized by Suny Group in China [20]. It combines a gas-heated rotating tumbler with a waste gas treatment unit to absorb, split and decompose toxic gas emissions (comprising of three steps: water and oil spray towers, activated carbon adsorption system, and a photocatalytic catalyst system, respectively). Treatment is performed by batch and capacity is up to 500 kg/h (in two to three batches per hour). Two fractions are finally recovered: (i) bare boards; (ii) mixture of ECs and solder particles. An additional physical separation step is therefore needed to separate ECs from the solder powder. Work in our laboratory also shows that at least 10% of the solder remains on the bare board and the ECs, which is a contamination that one has to deal with later in their recycling. This is an issue common to most methods reported in this subsection, except for centrifuged heating (see Section 2.2.6).

### 2.2.2. Infrared Radiators (IR)

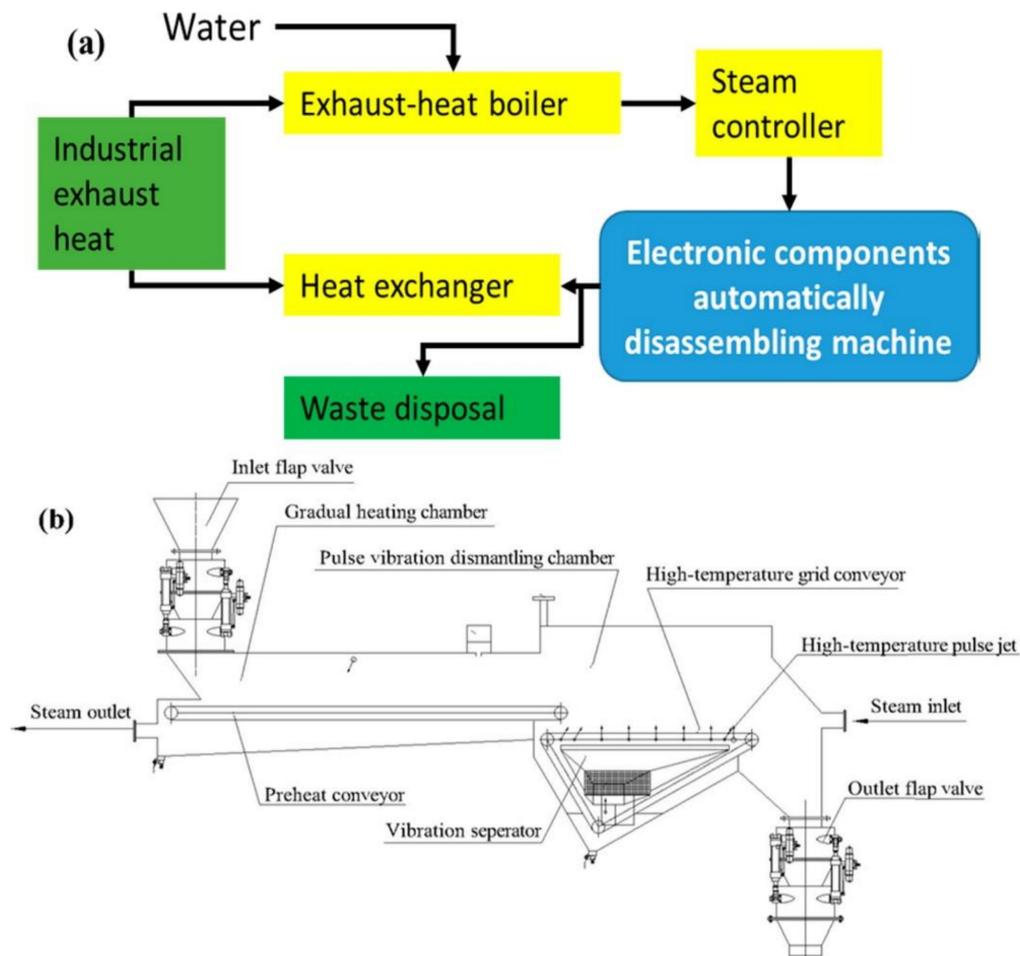
Controlled infrared radiation from quartz halogen lamps in a conveyorized mode is one of the most advanced methods to bring solder to its melting point. It has been first demonstrated in an automated disassembly process by NEC Corporation [21,22]. Infrared radiators are normally positioned on both sides of the WPCBs conveyor belt. Once the solder melts, ECs can be detached from the bare boards by robotic grippers or by gravity using their own weight; the rest is removed by a shearing machine or a brush [23]. Recently, using this method, Park and coworkers set up an automatic rotating rod and sweeping steel brush-type apparatus, which then achieved a 94% disassembling rate (disassembling rate,  $DR (\%) = 100 \times \text{Number of ECs released from PCBs} / \text{Total number of ECs on PCBs}$ ) at an operating temperature of 250 °C and feeding rate of 0.33 cm/s [24].

The characteristic that makes this method superior to others is that the damage ratio is low [25]. Additionally, the de-soldering process is normally fast because of direct heat transfer while little maintenance and cleaning are required due to the non-contact heating. However, as the IR radiator could not focus the beam on just the solder, combined with the differences in heat absorption rate, ECs are heated at varying temperatures, which precludes the possible recovery of functional ECs. Therefore, to enable functional EC recovery, focused IR radiation is needed, such as laser (see Section 4.1.1 for an example of tantalum capacitors recovery). In addition, despite many efforts to increase the throughput rate by using impacting rollers, the method is still unable to deal with large volumes.

### 2.2.3. Hot Air Heating

In a similar fashion to IR, forced heated air with temperatures of up to 260 °C was widely employed to melt solder, in combination with vibration and a pulsed jet to disassemble ECs from WPCBs [26–30]. For better mobility, portable heat guns are usually used in this process. Compared to IR heating, hot air convection is slower, thus lowering the odds of ECs overheating. This method offers a low capital cost, high disassembly rate, little maintenance, and is environmentally friendly. However, it has rather poor control accuracy. When heating a large area, the stream of hot air is unable to secure the expected spatial variation and distribution of temperature. Low heating efficiency is another concern in this technology. Fortunately, to solve this issue, Chen et al. proposed an industrial eco-friendly system using hot steam produced by industrial exhaust heat coupled with an automated heated gas disassembling equipment (Figure 1). In this system, through the flap valve, the WPCBs are dropped onto the pre-heated conveyor belt and further heated by the steam from the pulse vibration disassembling chamber. After that, the WPCBs are brought to the high-temperature grid conveyor where the electronic components are extracted with the assistance of hot steam and a vibrator or pulsed jet. With the much higher heat capacity of water ( $4.18 \times 10^3 \text{ J kg}^{-1} \text{ K}^{-1}$ ) compared to that of air, this process is better temperature-controlled; hence the potential liberation of toxic chemicals can be

avoided [23,31]. A possible concern in terms of CAPEX and safety resides in the use of a ~5 bar functioning pressure inside the semi-automated disassembling equipment [28].



**Figure 1.** Schematic (a) Process flow and (b) illustration of automated heated-steam PCBs disassembling equipment employing industrial exhaust heat. Reprinted with permission from [28] Copyright 2013, American Chemical Society.

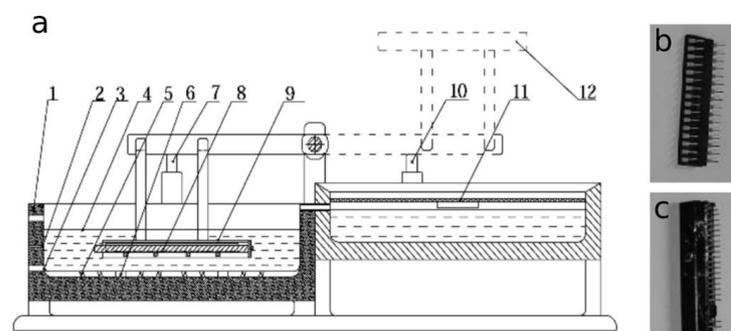
#### 2.2.4. Solder Bath Heating

De-soldering through direct contact of molten solder can be traced back to nearly three decades ago but has only been employed by a few ewaste recycling companies in China [23]. The first recycler to adopt this process is based in Suzhou, and has an operating temperature of 230 °C and a capacity of half a ton per day.

In this method, waves of melted solder are artificially generated in the bath, which contact the WPCBs suspended over the pool. ECs should not be overexposed to the liquid solder, as it could cause overheating. Therefore, the process is rather difficult to automate. Instead, most factories in China adopted a mechanical approach in which the WPCBs are pulled out of the solder bath then manually beaten by a hammer to separate the ECs [32]. Though fume hoods were used, negative long-term health effects were experienced by the workers. Worse still, the process is highly hazardous as the manual workers are exposed to high-temperature liquid baths of toxic metals. Some factories tried to minimize this danger to the workers by having an aluminium foil layer over the bath to stop splashing. However, due to its contact with solder, the wrinkled aluminium skin is then also be considered as hazardous waste.

### 2.2.5. Hot Fluid Heating

Another common route to melting SnPb based solder is to employ hot fluids [33,34]. The temperature of the liquid bath must be higher than the fusion temperature of the solder. In addition, the hot liquid should be risk-free and innocuous to humans and the environment. Additionally, most importantly, the hot liquid must be chemically inert with the materials in PCBs [35]. Because of these requirements, hydrophilic ionic liquids, diesel, paraffinic oil (methyl phenyl oil) or silicone oil are employed, which normally operate at around 215 °C and sometimes as high as 250 °C [23,33]. Figure 2a shows the diagram of the equipment system [35]. The clamped WPCBs are immersed horizontally in the hot fluid (ECs face upwards). Then, to efficiently remove ECs from the solder joints, a vertical dynamoelectric mechanical vibrator is used alongside two ultrasonicators installed at the bottom of the bath. The mechanical vibrator helps the solder to slip through the holes and drop into the fluid bath. Another vibrator is placed in a neighbouring position to move the ECs onto the griddle. Meanwhile, the addition of ultrasonication with cavitation and agitation effects makes the solder stay in liquid form in the fluid, enhancing de-soldering efficiency [35].



**Figure 2.** (a) Schematic diagram of the hot fluid PCBs disassembling system. 1. Insulator; 2. Temperature sensor; 3. Liquid exit; 4. Heating liquid; 5. Electrothermal wheel; 6. Ultrasonicator; 7. Dynamoelectric vibrator; 8. PCBs; 9. Clamp; 10. Dynamoelectric vibrator; 11. Griddle; 12. The separation position of the bare boards and the components. Reprinted with permission [35]. Copyright 2007, Springer London. (b,c) Comparison of (b) hot fluid disassembling approach versus (c) hot air approach. Reprinted with permission [35]. Copyright 2007, Springer London.

This method of de-soldering offers lots of advantages such as highly efficient disassembly and thermal energy consumption due to a better heat transmission coefficient. It is noteworthy that both soldered and unsoldered ECs can be retrieved with higher quality (Figure 2b,c) compared to those recovered using the hot air method [35]. However, the process faces the challenge of disposing of the liquid waste as well as of the need to rinse the bare boards and ECs for subsequent steps, hence generating wastewater that requires further treatment.

Regarding the heating media, a focus should be given here to ionic liquids (ILs) as their use is now percolating into industrial sites. ILs are salts that are used in their liquid state, often at, or close to, room temperature, but not always. They have received a renewed interest in green chemistry as, when compared to many organic solvents, they are good solvents for many organic molecules, they have excellent electrochemical and thermal stability (<400 °C) [36], low vapor pressure, and are non-flammable, making them safer to handle [37]. Scientific research reports on recycling PCBs using such solvents are, however, still fairly scarce with about ten articles per year for the past five years. This can probably be explained due to their cost, the issue of their recyclability (e.g., they cannot be distilled) as well as the variability of their properties as a function of their water content. All these issues render difficult process stability and reproducibility as well as the economic viability of any industrial process using them.

Most of the research reports that can be found on the use of ILs in the recycling of PCBs deal with the recovery of precious or strategic metals, and only a few discuss the removal of solder. The first report on this issue was published by P. Zhu et al. in 2012 [38,39], in which they report the recovery of solder using 1-ethyl-3-methylimidazolium tetrafluoroborate ([EMIm]<sup>+</sup>][BF<sub>4</sub><sup>-</sup>]) at 240 °C while stirring at 150 rpm for 10 min. This IL is reported as one of the most thermally stable ILs [36]. The process being above the solder's melting point (ranging from 183 to 232 °C), the latter is expelled using gravity and centrifugal force and collected at the bottom of the stainless steel vessel. In this respect, it is more a heating process approach than a leaching one. The advantage of the IL here is its stability at these fairly high temperatures, which are at the limit of even silicon oils. However, some delamination of the WPCBs was also observed as the IL somewhat dissolves, or at least swells, the epoxy resins of WPCBs, but this was not clearly demonstrated as such delamination is also observed when WPCBs are heated at such temperatures. This delamination could even prove useful for the recovery of silica fibres as well as copper layers. It should be noted that stirring is reported to play an important role in the process of maximizing solder collection. The report also did not study the processing of WPCBs with components still attached. Hence, it does not really address dismantling nor gives solder recovery or component dismantling rates.

This later issue was investigated in-depth one year later by Zeng et al., in a similar approach, but using instead the water-soluble ionic liquid 1-Butyl-3-methylimidazolium tetrafluoroborate ([BMIm]BF<sub>4</sub>) due to its large working temperature range [36], adapted viscosity and lower cost (few dollars per kilogram in bulk quantities). It was determined that the optimum process parameters were 250 °C, 12 min, and 45 rpm, for the reactor's temperature, the retention time and the WPCB basket holders' rotation, respectively. With this process window, 90% of the electronic components could be retrieved from the WPCBs. However, at this operating temperature, more than 1% in mass is lost every 10 h [36], which must be taken into account, both from an OPEX, and a health and safety point of view. The economic assessment of the process, however, suggested that it could be economically viable on an industrial scale. Additionally, indeed, a roughly one cubic meter pilot size reactor that enables such dismantling, as well as the separation and recovery of bare boards, ECs and solder has been developed by the same group, and is described in a patent application filed in 2018 in China [40]. It uses baskets to hold the PCBs that can be rotated around a central axis, thus creating turbulences that detach ECs. A secondary reservoir recovers the IL, while components are retrieved in a sieving basket located at an intermediate level (Figure S2, Supporting Information). Overall, it reports a disassembly rate of components exceeding 95%, and a solder recovery rate greater than 90%, although the issue of the amount of solder remaining on ECs is not discussed.

A significant advantage of using hot liquid media is that operations must be oxygen/air free, which decreases the amount of fume emissions so significantly that they can now easily be captured using a single activated carbon filter. Hence, despite the additional OPEX associated with the cost of the IL, overall CAPEX and OPEX seem to be significantly lowered since no complex and expensive fume treatment facility seems to be required.

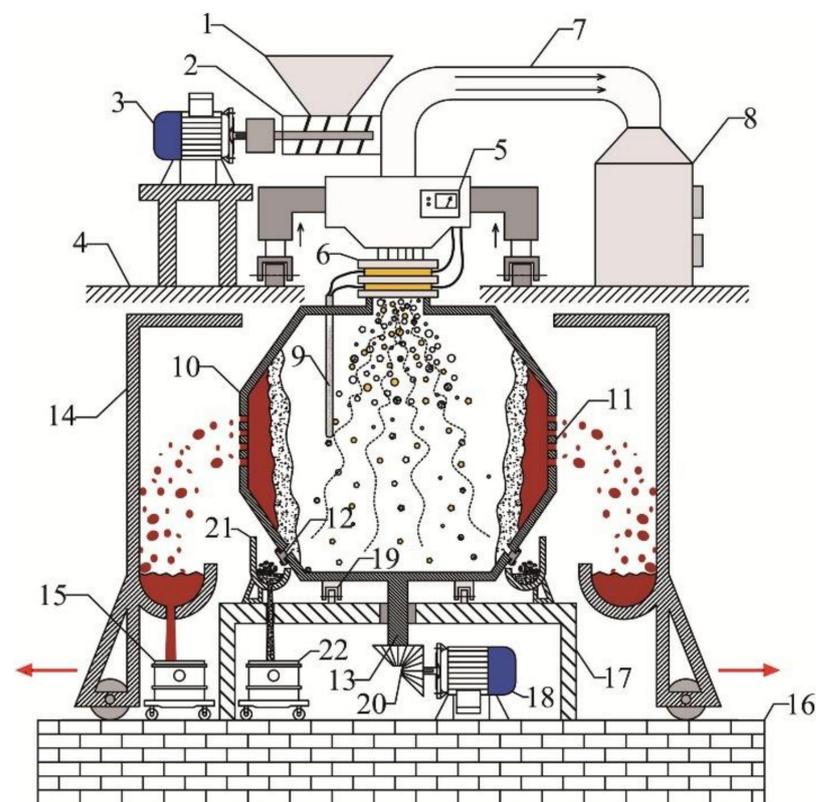
#### 2.2.6. Heated Centrifugation

The various methods described previously use heat to melt and recover solder, loose components and bare WPCBs. If such approaches enable WPCB dismantling, in some cases with high efficiency, reported solder extraction yields are rarely above 92 wt%. This comes from the fact that various effects play a role in molten solder extraction, mainly gravity, capillarity, the solder's viscosity, and surface/components' wetting properties. To increase yields, one can increase temperature to decrease viscosity, but this leads to more chemical decomposition of WPCBs and ECs and the generation of toxic emissions.

To circumvent this problem, it was proposed in 2010 by Y. Zhou and K. Qiu to dismantle WPCBs using heated centrifugation. They showed that heating WPCBs of various natures up to 240 °C and centrifuging them intermittently at 1400 rpm (g-force was

not given in the article) in oil, allowed for solder recovery and WPCB dismantling [41]. It should be noted that a similar apparatus was recently patented by others [42].

This approach was further developed towards high temperatures in 2018 by Z. Guo and coworkers, on grinded WPCBs, but it could, in theory, be applied to whole WPCBs or broken down WPCBs. In a first report, using the optimized temperature of 420 °C and a gravity coefficient of 1000, Pb and Sn were recovered at only 57.16% and 59.49%, respectively. However, if instead temperatures of up to 1300 °C were used, then the total recovery fraction for Cu, Zn, Pb, and Sn would be 93.23%, 80.86%, 94.54%, and 97.67%, respectively. The large-scale centrifugal separation apparatus diagram proposed in reference [43] is reproduced in Figure 3.



**Figure 3.** Schematic diagram of large-scale centrifugal separation apparatus (1. Hopper; 2. Spiral feeding rod; 3. Motor; 4. High platform; 5. Microwave generator; 6. Conductive sliding ring; 7. Flue pipe; 8. Exhaust gas treatment tower; 9. Thermocouples; 10. Rotating centrifugal gravity separation reactor; 11. Porous filter plate; 12. Residue discharge port; 13. Axis; 14. Liquid metal receiver; 15. Metal liquid storage tanks; 16. Low foundation platform; 17. Rotating centrifugal gravity separation reactor platform; 18. Rotating centrifugal gravity separation reactor motor; 19. Ring bearings; 20. Gear; 21. Slag receiver; 22. Tailings storage tank). Reprinted with permission [43] Copyright 2018, Elsevier Ltd.

### 2.3. Chemical Methods

If the most widespread approaches to dismantling WPCBs, by melting and removing solder, are via the thermal treatments listed above, some drawbacks can be found in their use, such as: (i) possible toxic emissions; (ii) high operating cost (energy); (iii) heat damage to components that otherwise, in some cases, could be reused; (iv) residual solder remaining on the WPCBs that can pollute downstream hydrometallurgical processes [43,44]. To circumvent these issues, some chemical approaches have been developed that dissolve more or less specifically some, or all, of the exposed metals. Hereafter we therefore review methods that allow WPCBs to be dismantled by chemical dissolution of the solder or even of all exposed metals.

### 2.3.1. Solder Dissolution

Regarding the recycling of WPCBs beyond pyrometallurgy, since copper represents a significant mass fraction of PCBs, a strong focus has been given to the leaching of copper using either acids or ammonia-based solutions [45,46]. Since such approaches have recently been reviewed [47], we will focus only on the latest developments. Although ammonia-based solutions are often more specific, acidic approaches are more economically viable especially when using  $\text{H}_2\text{SO}_4$  [48–50]. The choice of the right oxidizer (for example, oxygen or halogen gases, peroxides, or oxidizing metal ions) is key as it can render the solution: (i) extremely corrosive and/or; (ii) dangerous for the workers and/or; (iii) complex, with new chemical elements that must then be separated from other metals to be recovered, all of which adds to the process CAPEX or OPEX [51–56].

To avoid these drawbacks, ionic oxidizers have recently been favoured, and to avoid contamination,  $\text{Cu}^{2+}$  appears to be a better choice than  $\text{Fe}^{3+}$  [57–60]. Various formulations have been investigated in systems such as  $\text{HCl-CuCl}_2\text{-NaCl}$  and  $\text{H}_2\text{SO}_4\text{-CuSO}_4\text{-NaCl}$ . All use chloride as a stabilizer for cuprous ions, which favours further copper oxidation, despite the corrosivity that NaCl salt adds to the system.

Hence, an optimized formulation comprising of 0.5 M  $\text{H}_2\text{SO}_4$ , 0.4 M  $\text{CuSO}_4$ , and 4 M NaCl was recently proposed and reported, disassembling WPCBs by leaching all exposed metals. Air was bubbled within the reacting solution to help the regeneration of the oxidizing cupric ions. Copper metal could then be retrieved electrochemically [61].

An original approach, which has the advantage of reduced OPEX, coprocesses WPCBs with used tin stripping solutions (TSS). TSS are waste solutions generated during new PCB production. Despite being a waste, TSS can still remove solder and hence enable the subsequent dismantling of WPCBs [10]. TSS leaching power for WPCBs' solder is due to its content in various oxidizing metal ions as well as residual nitric acid. Therefore, when TSS waste reacts with WPCBs at room temperature for 2h, it is reported that 87% of the SnPb solder, 30% of the Cu, 29% of the Fe, and 78% of the Zn could be dissolved. Furthermore, it enabled ~87% of ECs to be dismantled. Finally, close to 99% of the dissolved Sn, Pb, Fe, Cu, and Zn could be retrieved by selective precipitations (Figure S3, Supporting Information).

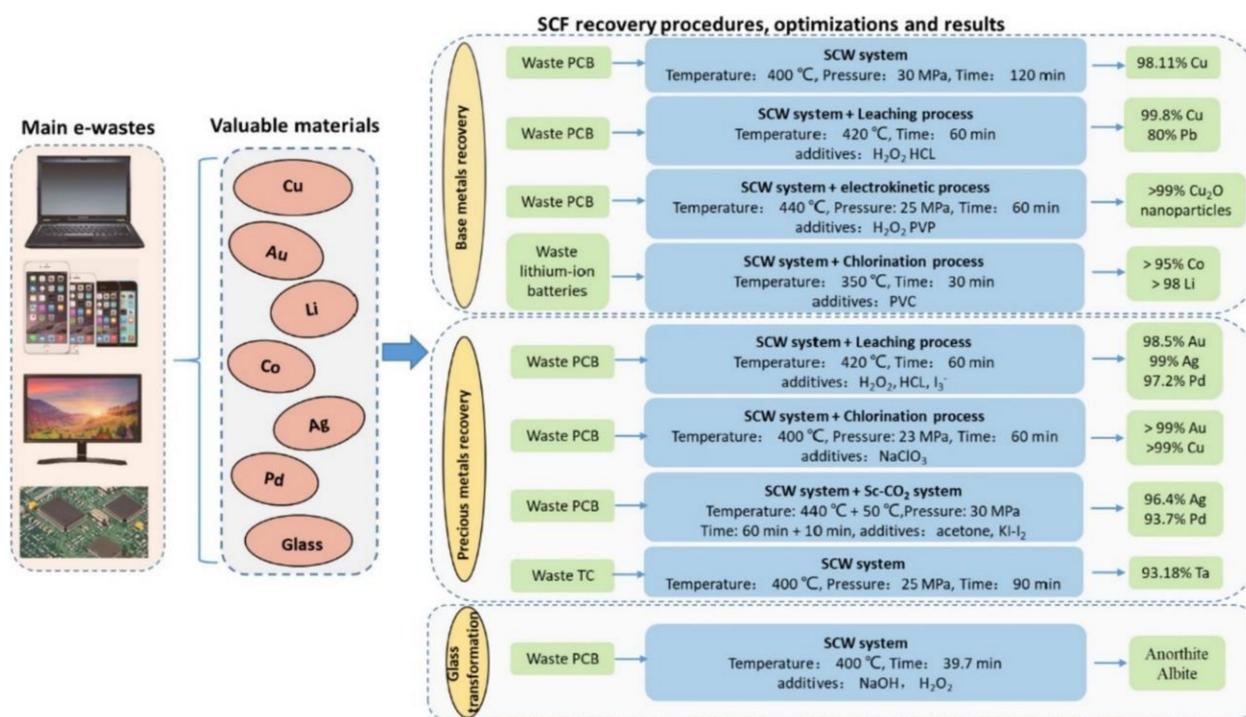
Overall, the chemical approaches listed above lack specificity and not only leach solder but also force a costly oxidation/reduction copper cycle. If this could be avoided, it would render the dismantling much more cost-effective. Such approaches, targeting solely the recovery of Sn element have been reviewed by Yang et al., in 2017 and will therefore not be described further here [17]. The spotlight could, however, be turned on even more recent publications reporting specific solder leaching accelerated by ultrasound [62]. In this report, Zhu et al., use both fluoroboric acid and (costly) hydrogen peroxide to dissolve lead-tin solders at room temperature. The need for ultrasound comes from the SnO passivation film that forms early on at the surface of the solder. The passivation layer can be attacked by ultrasonic cavitation, which enables the leaching mixture to dissolve pristine SnPb solder. Finally, the kinetic comparison of the leaching indicates a two to three-fold efficiency increase when using ultrasound.

It should be noted that the use of ultrasound activation in a proprietary process (no description available) was also reported and licensed by Quinnovations Ltd. and affiliates [63,64].

### 2.3.2. Hydrothermal and Supercritical Fluids (SCF) Treatment

Various reports using sub- or super-critical water indicate that some specific reactivity can be achieved, whether one targets the epoxy resin, base metals, or precious metals. Supercritical water is defined as the state of water above its critical point ( $T = 374\text{ }^\circ\text{C}$ ;  $P = 221.1\text{ Bar}$ ). In such conditions, water becomes a strong oxidant that can easily oxidize organic matter or metals, so much that only a few alloys can somewhat resist its corrosive power (for example Inconel<sup>®</sup> 600 or Incoloy<sup>®</sup> 825) [65]. Specificity can be achieved by changing process parameters and/or additives. However, one cannot find a process that solely targets SnPb or other solder alloys to enable subsequent EC removal (Figure 4) [66,67].

If other conditions are used, it is now the epoxy that can be targeted, leading to the equivalent of pyrolysis but without the emission of toxic gases [65].



**Figure 4.** Resource recovery processes of ewaste by SCF technology. Reprinted with permission from reference [66] Copyright 2019, Elsevier Ltd.

Some epoxy resins contain large amount of bromine containing flame retardant that are known to be polluting and hard to recycle. Xing et al. [68] present a new process based on the use of subcritical acetic acid (220–260 °C, 2.6–3.6 MPa) to decompose brominated epoxy resins. After a one-hour reaction time using acetic acid of 49.90% mass concentration under a pressure of 2.6 MPa, most bromine was transformed into HBr and extracted in the aqueous phase. The remaining glass fibre and copper foil were liberated and easily recovered after epoxy degradation. This process presents an environmentally friendly way to treat brominated epoxy resins as the acetic acid can be recovered and reused at the end of the process.

### 2.3.3. Dismantling via Epoxy Resin Treatment

Compared to previous reports, a shift in paradigm for WPCB dismantling was reported for epoxy resin treatment. It consists of first dissolving or decomposing their epoxy resins constituents. Early reports proposed to use concentrated nitric acid (8 N) at 80 °C for 3h, but yields were small [69,70]. Zhu et al., proposed to use Dimethyl Sulfoxide (DMSO) instead [39,71]. DMSO is a highly polar and water-miscible organic liquid, and is often claimed to have an exceptionally low level of toxicity. However, recent in vitro studies report otherwise [72]. Tests on ~1 cm<sup>2</sup> samples at 60 °C showed that: (i) after 45 min, delamination started to occur and; (ii) after 210 min, total exfoliation of WPCBs could be observed allowing the separation and recovery of ECs, copper foils, and glass fibres. Since this process requires DMSO to diffuse within the laminated structure, larger samples need more time and/or higher temperatures to be treated efficiently. DMSO was recycled using a rotary evaporator, which, in addition to its toxicity, could represent an OPEX limitation as DMSO is a high boiling point solvent, which requires a significant amount of energy to be recycled. In a similar approach, N,N-dimethylformamide (DMF) was also shown to efficiently dissolve brominated epoxy resin, resulting in the delamination of WPCBs and

its components' separation. The process parameter window was 135 °C for 3 h using 1 cm × 1 cm with WPCB: DMF ratio of 300 g/L. DMF regeneration was also performed using a rotavapor [73].

A promising new approach using a soft chemistry route was published recently by Chen et al. [3], which dissolves a large range of anhydride epoxy resins at temperatures below 200 °C (Figure S4, Supporting Information). It used ethylene glycol (EG) as a solvent, as well as a reactant, in which hydroxyl groups can react in a transesterification reaction with an ester bond present in the resin's polymerized network. The exact reacting mixture was comprised of EG, N-Methyl-2-Pyrrolidinone (NMP), and the reaction catalyst, 1,5,7-Triazabicyclo [4.4.0]dec-5-ene (TBD), in a molar ratio of 1:1:0.02, respectively. The role of NMP is to accelerate the diffusion of both EG and TBD by swelling the resin.

Amazingly, this process allows the electrical circuit to be recovered unaffected and still functional. In a way, it is not a complete dismantling of WPCBs but it definitively could be the first purification step in such a process. Finally, the report also mentioned that the chemicals used could be recycled.

Recent works have shown that epoxy treatment assisted by microwave can enhance the efficiency both in terms of yields and time. Microwave-assisted organic swelling present new and effective strategy to delaminate multilayer structure of WPCBs. In a promising approach, Monteiro et al. [74] achieved delamination of WPCBs in only two cycles of 10 min for a solid liquid ratio of 300 g/L of 225 mm<sup>2</sup> WPCBs parts using NMP at 200 °C. Huang et al. [75] present an optimized microwave-assisted process to recover non-metallic parts of WPCBs based on nitric acid. Under 95 °C for a concentration of 7 mol/L of HNO<sub>3</sub> with a liquid/solid ratio of 10 mL/g and 300 W microwave power, 88.42% separation was achieved between epoxy and glass fibre after 12 h. The products of the reaction can be recovered and reused for the next separations, making it a sustainable process. Moreover, this process is economically viable, with a net profit of \$ = USD 3.165 for 1kg of the non-metallic materials of the PCB only.

#### 2.4. WPCBs Fragmentation by High Voltage Electric Pulse Crusher

Dismantling WPCBs by High Voltage Electric Pulse Crusher is considered the most promising technique even though most research on electrodynamic fragmentation focuses on mineral ores. The idea of employing electric shockwaves for disintegration was first coined in 1961 when Yutkin discovered that suspended particles are crushed by electric current during water electrolysis [76]. Figure S5 in the Supporting Information displays the structure of the SELFRAG Lab High Voltage Pulsed Power Fragmentation System. The system is composed of a reaction unit filled with deionized water and a high voltage pulse generator which is continuously charged by a high voltage power supply [77].

Depending on the rise time of the electrical discharge, two mechanisms can be observed. When the pulsed rise time is higher than 1 μs, electrohydraulic crushing takes place. If the pulsed rise time is smaller than that threshold, the deionized water shows a high dielectric constant which is in high contrast with that of PCBs. Consequently, electrodynamic fragmentation occurs within the solid PCBs only [78,79]. In this latter case, (i) electrical pulses form plasma channels that move through the material and generate a locally high temperature and pressure (the pressure can be as high as 103 MPa); (ii) the plasma channel's diameter increases significantly (from μm to hundreds of μm) resulting in the fragmentation of the WPCBs due to excessive tensile stresses [80]. Since stresses mainly happen along phase boundaries, this technology can selectively break PCBs along those boundaries; hence minimizing EC damage [77–79]. In addition, as the breakage happens in water, it does not produce secondary pollution as seen in the solder bath method or hot fluid method. Additionally, the process is closed, making the wastewater easier to manage; hence the risk to the environment and workers is lowered.

Over the past 20 years, an increasing effort has been devoted to realizing the potential of High Voltage Electric Pulse Crusher for processing WPCBs [77,81]. Fragmentation's performance is highly affected by four main parameters, namely; level of filling, voltage,

pulse rate, and number of pulses. However, high energy usage is the critical challenge holding this technology back from wide deployment [82]. Yield is indeed hindered since the majority of input energy is converted into heat, leading to fairly low energy efficiency, ranging between 12 and 35%, which contradicts theoretical predictions [83]. Additionally, high initial investment and low throughput hinder the scalability of this technology. Nevertheless, some commercial products have been put on the market that can deal with ~100 kg/h (see for example ImpulsTec GmbH in Germany, or ITHPP-Alcen in France).

### 3. Electronic Component Sorting

Once dismantled, ECs are diverse. If a chemical analysis is performed on the unsorted components, the number of elements that are present is high. The sorting step's scope is to reduce chemical element complexity by separating ECs by composition. The advantages are twofold: on the one hand, elemental recovery rates are increased and, on the other, recycling techniques can be chosen and tuned for each EC category. A qualitative comparison in terms of OPEX and CAPEX is given in Table 2.

**Table 2.** Qualitative comparison of the e different sorting processes (+ cheap, ++ mid-priced, +++ expensive).

Sorting Process	CAPEX	OPEX	Advantages	Drawbacks
Physical separation	+++	++	Large capacity Simple and existing processes	Requires multiple processes and machines
Machine vision	+	+	Easy to implement Focus on specific ECs	Does not focus on the elemental composition Expensive device
Spectroscopy sorting	++	++	High accuracy elemental sorting	No industrial process developed yet

#### 3.1. Sorting by Physical Separation

Except for sieving, physical separation methods are often binary. Items are separated into two bins depending on their physical properties. These methods are aimed at coarsely sorting components at the beginning of a sorting process. Individual EC types are not targeted specifically.

The literature describing such methods for powder from whole PCBs crushed/milled is extensive [84–86]. However, it lacks studies that apply physical separation methods directly on disassembled PCB components. Currently, the notable study dealing with physical separation of disassembled components is from Lee et al. [87]. The authors performed a thorough three-step physical separation study using disassembled components. They used: (i) binary sieving (9.5 mm), (ii) magnetic separation and (iii) dense medium separation.

By simply using sets of sieves, components can be sorted by size. The purpose of sieving is also to separate components from dust, scrap and other compounds of the disassembly step. For example, if the disassembly involves heat, sieving may remove dust and ashes from the waste stream. Afterwards, magnetic separation, which takes advantage of the ferromagnetic properties of some elements such as Fe, Co and Nickel and some rare-earth alloys (Nd, Dy, Sm, and Gd), can separate magnetic components from the waste stream, for example, inductance coils, which usually have an iron core. Finally, dense medium separation can be carried out by assessing the floatability of the components in a liquid (typically water). If the component is denser than the liquid, it sinks; if it is of similar density, it stays in suspension; if it is less dense, it floats.

The strength of Lee et al.'s analysis is shown in Figure S6. They performed chemical analysis at every step of the separation process. Their objective was to assess how much of each element can be concentrated in each sorting bin. This approach is interesting because it disregards the nature of the component and focuses on the chemical composition. Their results notably showed that 87% of Al, 96% of Sr, 74% of Nb, and 72% of Mn that were initially present in the waste could be concentrated in separated branches. For example,

87% of the total aluminium mass went into the division where components were: smaller than 9.5 mm, magnetic and floating. However, for some elements, such as Cu, Ag, and Sb, the method was unsuccessful in concentrating the elements in a single fraction.

It is worth noting that Hayashi et al. successfully separated tantalum capacitors and quartz resonators using a column type pneumatic separator [88,89] in which air flows upward within a column that has orifices located at different heights. The devices were fed from the bottom. Depending on their mass and shape, the air drag varies and leads each EC type to different orifices.

Combined with this approach, other physical separation methods could be of interest, such as Corona discharge or gravity shaking, which are not yet reported to have been used with disassembled components, except for plastic parts [90,91].

### 3.2. Machine Vision

Machine vision refers, in simple terms, to the analysis of images to provide decisions. Machine vision has been used in industry for various purposes for decades. This approach can be highly accurate while only requiring a camera and a computer.

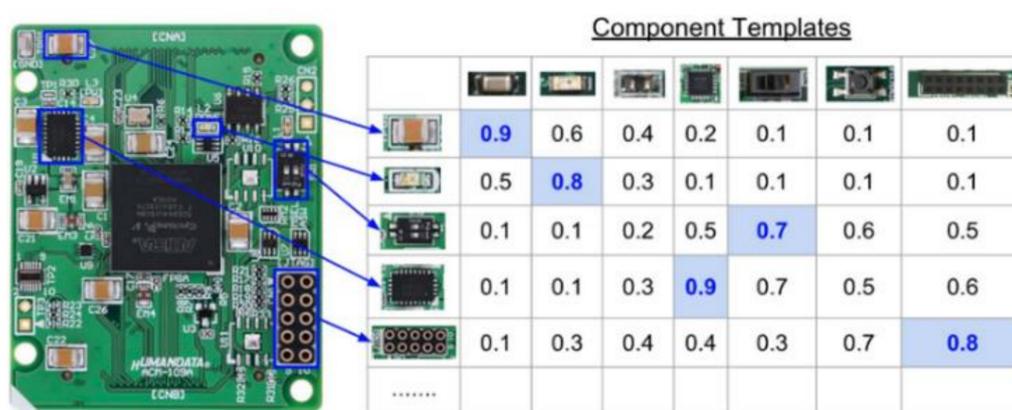
For recycling PCB components, machine vision is promising because ECs have distinctive features that are recognizable optically. The machine identifies these traits and features and classifies the components into different classes. These traits can be colours, topography, edges, sizes, etc. To classify components, researchers must focus on: (i) data quality, (ii) pre-processing techniques, (iii) classification algorithms.

Particular attention must be paid to data quality, which needs to be representative of a real waste influx with a high heterogeneity; thus, collaboration with the recycling industry is key. Pramerdorfer et al. provided a public dataset specifically purposed for computer vision analysis [92]. Their data arises from a recycling facility. The authors highlighted the lack of public, high-quality data since most articles do not disclose their dataset. Later, Mahalingam et al., provided another dataset designed for machine learning [93]. They provide only four categories of components: integrated circuits (ICs), resistors, capacitors, and inductors. All these datasets contain images of PCBs that are not dismantled. Therefore, the components are first localized on the PCBs and classified. However, within the current focus of this review on dismantled PCBs, there is currently no public dataset on dismantled EC components, probably due to the proprietary value that such a database represents. For recycling, these datasets could also be generated or improved by providing better classification based on the chemical composition. For instance, the class “capacitors” contains components with many different compositions.

The data type is also important. Typically, a conventional image contains three channels: Red, Green and Blue (RGB). Several studies use hyperspectral images that contain several channels to form reflectance spectra [94–96]. These images provide significantly more information than conventional RGB images and could enable higher classification accuracies. Interestingly, the infrared channels of a hyperspectral image can highlight metal traces [96].

Once the data are of high quality, images can be processed to identify and extract key features (edges, shapes, colors, etc.). Researchers have focused on extracting useful features [97–102]. Some simple features include colours as a sorting criteria [103,104]. This approach is simple but lacks accuracy for many components. For example, both CPUs and capacitors are often black. Another criterion is the object size. Copindean et al., use their apparatus to detect the object size and combine this information with the colour [104].

To classify efficiently, researchers use machine learning algorithms such as principal component analysis [105], support vector machine [96,98], nearest neighbour [98,101] and neural networks [106–108]. More specifically, convolutional neural networks are the most promising as they perform well for image classification despite requiring large training datasets. In Figure 5, the researchers used a convolutional neural network to predict the EC types from a PCB image [108]. They computed a similarity score compared to a reference EC template.



**Figure 5.** Detection and recognition of components on a PCB using convolutional neural networks. The right table shows the similarity score computed by the neural network for each class template. Reprinted with permission [108]. Copyright 2019, IEEE.

As an alternative approach, some researchers performed text recognition on PCB components [103,106,107]. Their analysis is thorough, but it is unclear how well text information can help component sorting. They further suggested using text information in combination with other methods, but this approach might not be compatible with many of the dismantling approaches that can destroy the marking by heat or chemical attack.

Machine vision techniques have limitations as many components may have different appearances but have similar chemical compositions. It is worth noting that the reverse is also true. In these cases, the classification system needs prior knowledge of the chemical composition of these ECs to merge them in a single class. The lack of chemical information is the major flaw of currently published works. Additional characterization techniques that assess composition are needed to tackle this challenge.

### 3.3. Sorting Using Spectroscopy

Unlike previously described methods, various spectroscopies can target the chemical elements within a given EC to enable their identification and quantification. The technical specifications of such a method are to be compatible with an automated sorting system (e.g., a conveyor). Therefore, acquisition speed is a key factor for industrial deployment. Additionally, the technique should not require any chemical treatment beyond those described in Section 1 or EC destructive sample preparation. As an example, high-accuracy methods based on inductively coupled plasmas (ICP), which require the digestion of the component in acids, are inadequate for sorting ECs. In this section, a few relevant techniques are highlighted.

Laser induced breakdown spectroscopy (LIBS) consists of ablating a surface with a single laser pulse. This creates a plasma that emits light whose spectrum can be characterized using a spectrometer. LIBS can target almost all elements of the periodic table; thus, it has been used extensively to characterize electronic waste [109–113], including Ta, Zr, and Ba in capacitors [114,115], Au and Ag in ICs [110] and rare-earth elements in specific components (speakers, Hard Disk Drive magnets) [116]. We note that the literature is not limited to ECs on WPCBs. LIBS is extremely useful in characterizing other fractions of PCBs such as their plastics/epoxy fraction, where it performs classifications with accuracies of up to 98% [117]. This is of particular importance to measure the level of toxic elements that pollute the polymer (forbidden brominated flame retardant, Sn, Pb, As, Hg, etc.) [118]. The drawback of LIBS is that it is a surface characterization technique. Therefore, it is not applicable to ECs that are heterogeneous. For instance, if a component has a thick polymer housing, LIBS does not capture the composition of the materials within the EC.

Another technique that is complementary to LIBS is X-ray fluorescence (XRF). It is expected that XRF could be powerful, although the existing studies use XRF to characterize PCBs but not specifically for sorting ECs [119–126]. A single measurement can yield

information about the presence of more than 40 elements in the periodic table. Moreover, XRF becomes more sensitive as the atomic weight increases, so high value heavy metals are easily detected. Another advantage of XRF is that, as opposed to LIBS, the measurement can be performed on the whole component by using a broad X-ray beam. Additionally, by using high-energy X-rays, the entirety of the component volume can be measured; thus, going through any housing that the component may have. With XRF, the elemental information of a component obtained is more accurate than those acquired by LIBS. The main downside of XRF is its acquisition speed. To obtain a high signal over noise ratio spectrum, the system must acquire the signal for at least a few seconds. In this regard, XRF characterization might be useful for building an elemental composition database or for slower, high accuracy sorting but will be unable to deal with a high volume waste flow.

Furthermore, XRF only yields elemental analysis without spatial information. For sorting, any conveyor elemental mapping is obviously impossible. In this regard, X-ray imaging might be of interest. In the latter case, the X-ray beam is projected onto the components and an X-ray transmitted image of the component is obtained from an X-ray camera placed on the other end. X-ray images yield two types of information. On the one hand, the X-ray image can be analysed to identify patterns present in the internal structure of the components. On the other, the absorption strength yields information about the atomic weight of the elements present in the component, or of its thickness. Despite this, there is currently no scientific literature on X-ray imaging being applied to electronic waste, probably because this method cannot differentiate a thick light element from a thin heavy one, which both give a similar absorption.

### 3.4. Combinatory Approaches

From the methods described previously, it appears that none of them can be used alone in an industrial sorting system. In other words, there is no perfect EC sorting technique since they all have their strengths and weaknesses. Thus, the right strategy should come from a combination, as suggested by Kleber et al., of both LIBS and computer vision [127]. Several sorting methods assembled along the sorting line should yield higher sorting efficiencies. For instance, combining the predictions of vision and spectroscopy machine learning models could yield high sorting accuracies. However, cost must also be considered. For example, computer vision could be implemented in almost all cases whilst spectroscopy techniques might be employed for a subset of specific components.

A pending patent from Gabriel et al., highlighted the possibility of combining both X-ray and optical images (Figure S7a,b) [128]. They used two convolutional neural networks trained separately for each data type. Thereafter, the predictions probabilities of both models were merged. Figure S7c in the Supporting Information is the normalized confusion matrix showing the prediction accuracy of the combined predictions. They obtained promising results with accuracies of at least 93% for all classes and an average accuracy of 95%.

## 4. Recycling at the Electronic Components Level: Processes and Opportunities

Once PCBs have been automatically dismantled and ECs have been sorted, one now has access to streams of single types of ECs, and hence to new and enriched sources of chemical elements that, up to now, were seldom accessible for recycling. This is for example the case of Ba, Ti, Sb, Zr, Mn, REEs, Ga, Si, and refractory metals such as Nb, Mo [129], Ta, W, Re. It should be noted that refractory metals are often overlooked and not quantified in WPCBs' composition analysis as there is currently no standard for such analysis of PCBs [130,131]. Having access to those elements could represent new business opportunities and would also allow some countries to be less reliant on natural extraction or importations, and reduce the environmental impact of the ECs' life cycle.

We therefore review hereafter a selection of classes of ECs that could represent interesting new business opportunities due to their recent, or soon to be, availability.

#### 4.1. Refractory Metals

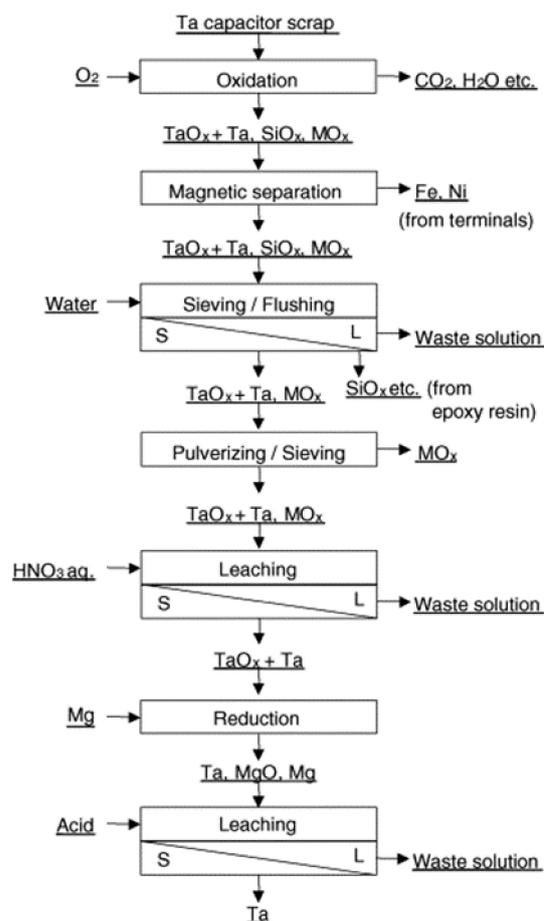
##### 4.1.1. Tantalum and Niobium-Based Capacitors

In ECs, niobium and tantalum are mostly found in electrolytic capacitors. If tantalum was the material of choice for such components until 2000, a spike in its price in the early 2000s pushed for its replacement by niobium. There is currently little industrial effort to recycle them. It was reported in 2017 that the end-of-life recycling rate for tantalum is only 1%, despite the fact that tantalum is regularly listed as a critical raw material since: (i) it comes from refining conflict minerals [132], and (ii) its supply chain's resilience strongly depends on its recycling [133]. Fortunately, even if only a few recycling reports were published during the fifteen years following the pioneering work of Hoppe and Korinek on recycling tantalum in 1995 [134], there seems to be a ramping up of research efforts on this issue. Indeed, 90% of all reports on recycling tantalum-based ECs have been published in the past five years (based on a Web-of-Science search on the key words tantalum AND recycl\*). This, combined with previously mentioned progress in dismantling and sorting, leads us to believe that this low recycling rate could soon go up. Not many reports focus on Niobium, probably because of its lower value as well as current smaller volume of end-of-life Nb-rich waste streams.

The simplest approaches reported tend to thermally or hydrothermally process these capacitors (burning off all organic matter) followed by various grinding and physical separations including size, density, magnetic and corona electrostatic separations. Afterwards, specific leaching of other materials and non-refractory metals ( $\text{SiO}_x$ ,  $\text{MnO}_2$ , silver, remaining solder, etc.) may also be employed. The advantage of this general approach is that it refrains from oxidizing tantalum metal further and therefore saves the cost of its reduction. Meanwhile, its drawbacks are that: (i) high-temperature combustion or pyrolysis generates harmful halogen compounds from the decomposition of brominated flame retardants and other halogenated materials; (ii) it does not allow the recovery of high purity grade Ta [135].

To avoid the first issue, Katano et al., proposed using steam gasification with sodium hydroxide to enable the decomposition of organic matter at lower temperatures as well as the neutralization of the halogenated gases. The report, however, does not mention any further purification of the tantalum sintered compact, which is likely to contain various contaminants [136]. Another solution was to use environmentally friendly supercritical water, as described in Section 2.2.3. For example, references [137] and [67,138] used it to either depolymerize or oxidize (by adding +10%  $\text{H}_2\text{O}_2$  in the mix) all organic matter, at 425/400 °C, 25 MPa, for 120/90 min, respectively, enabling the recovery of Ta electrodes (purity = 93.18%).

To reach higher grades, one needs to include either: (i) a fairly classical halogenation purification step [139,140], with some possible innovation to avoid the toxicity of halide gases [141], for example by using  $\text{CCl}_4$  [139], or  $\text{FeCl}_2$  as a chlorination agent [142,143]; or (ii) additional hydrometallurgy and reduction steps. For the latter case, most published works proposed alternative approaches that avoid the use of hydrofluoric acid for the Ta lixiviation step [144]. Hence, in 2005, Mineta and Okabe published a complete process (Figure 6), including hydrometallurgy and a magnesiothermic reduction step of the intrinsic capacitor  $\text{TaO}_x$  dielectric, which enabled the recovery of 99 w% pure Ta with an overall 90–92% recovery efficiency [145]. Piotrowicz et al. used 8 M  $\text{HNO}_3$  as a leaching agent for Ag, Sn and Mn; the thermal reduction step used Ag, Mg and Si as reducing agents. Overall, it enabled a 96% anode recovery. Full Ta reduction was only observed for the melted piece with Mg. Overall, the recycling efficiency into pure tantalum metal form was above 50% [146].



**Figure 6.** Flowchart for tantalum recycling from waste capacitor. Reprinted with permission from reference [145], Copyrights 2005, Elsevier Ltd.

Alternative solutions to solubilize significant amounts of tantalum and niobium, such as oxalate-based aqueous media [147] have also been proposed and investigated. The authors further indicate that recycling of the relatively expensive oxalic acid is needed to make the process economically viable.

An alternative to the thermal approach, which also keeps tantalum in its metal form, is to leach out all other materials but the tantalum itself. Such a process has been proposed by von Brisinski et al. who used Lewis acidic mixtures of dialkylimidazolium halides and aluminium trichloride to react with through-hole tantalum capacitors [148]. Although the exact mechanism of the reaction is not explained, not only was the epoxy dissolved but so was the silver, tin (contacts) and most of the manganese oxide electrolyte, leaving only the pure tantalum anode cylinder and traces of  $\text{MnO}_2$ . The report does not give any details regarding its exact purity or the recyclability of the IL, so one cannot perform an economic assessment of the possible OPEX and viability of such a process.

Further purification, by selective extraction of tantalum from an acid leaching solution, has been proposed using ILs instead of an organic solvent in a liquid–liquid extraction process. To emulate the role of the organic solvents used for Ta extraction, such as methyl iso-butyl ketone (MIBK) or tri-butyl phosphate (TBP), ILs were rendered task-specific by functionalizing them with ketone or phosphonate groups, successfully making them selective to tantalum for its extraction from a sulfuric acid leachate [149–151].

Additionally, three oral presentations at the E-MRS Fall meeting 2019 in Warsaw described innovative approaches for recycling Ta based capacitors. In the first presentation, Rousseau et al. from Chimie ParisTech proposed performing a sequence of pyrolysis, crushing, magnetic separation, and sieving followed by a final purifying step using a high efficiency Ar/ $\text{H}_2$  plasma torch (20 kW, 5 min Ar followed by 5 min  $\text{H}_2$ , with  $T_{\text{max}}$

= 1200 °C; energy consumption calculated to be less than 10 kWh/kg<sub>Ta</sub>). The latter step reduces MnO<sub>2</sub> into Mn and extracts the manganese metal by condensing it in another part of the reactor, leaving behind high purity Ta metal [152]. This is to be compared with the industrial approach to produce Ta using sodiothermic reduction followed by ebeam melting/purification, which requires 37 kWh/Kg<sub>Ta</sub> [153–155].

The second presentation, by Degabriel et al., also from Chimie ParisTech, outlined a proprietary process based on an assisted lixiviation of tantalum oxide using layered double oxide (LDH). Indeed, when the pH and solution's potential are chosen at a value at which a Ta polyoxometallate is the stable oxide, then the LDH can remove it from the solution via its intercalation into the LDH. Hence, even though Ta oxide's solubility is low in phosphoric acid, since it is constantly removed by intercalation into the LDH, its lixiviation is driven to the right of the dissolution equation as per Le Chatelier's principle. Since the intercalation is specific, Ta can be recovered in a second step. A 100% recovery rate was reported [156]. This approach, however, requires a reduction step to recover Ta metal.

The third presentation, by Choi et al., proposed a process flow in which one particularly noteworthy innovation was the use of an optical detection of Ta based capacitors, followed by its dismantling using laser heating pulses, melting the solder, and picking up the device via a robotic arm [157]. As part of this project, Sauer et al. presented a method to extract and purify tantalum from capacitors up to 98% purity [158]. The process relies on three main steps: cryogenic grinding and leaching using 6-molar hydrochloric acid followed by an electrolysis. More details on this automated disassembly line of end-of-life electronics was published by Noll et al., as we wrote this article [159]. It described a "dismantling and sorting line for a piece-by-piece processing of cell phones and printed circuit boards with seven interlinked machines", which was installed at a recycling facility. The subsequent steps were more classical, with mechanical treatment (including a cryogenic crushing) allowing for the efficient removal of silver (of which 90 wt% stays in the epoxy fraction). Globally, it appears an elegant approach for disassembly although the reported recovery speed of 0.3 s per capacitor could prove to be a hindrance for industrial adoption when tons/h is the treatment rate of WPCBs that needs to be dealt with. However, it might prove economically viable for the recovery of high value components, such as CPUs or rare-earth-based magnets [160].

Recently, Ramon et al. showed that recycling of tantalum capacitors using automated selective dismantling is economically viable [161]. Using a machine vision system and a trained convolutional neural network, the Ta capacitors are detected on the PCBs. Once spotted, the capacitors are removed mechanically using a scraper mounted on a robotic arm. The economic study of the process shows that in the current situation an initial investment of EUR 60,000 would allow a minimum return on investment of 15% over 4 years. This process requires an average of 2.5 s per capacitor. As the selective dismantling of the tantalum capacitors is a time consuming (hence, costly) process, a process based on the systematic dismantling of all the components on the PCBs followed by a visual sorting process would reduce the average removing time and make this process even more cost-effective, especially if the tantalum capacitors are not the only electronic component recycled.

Niobium capacitors are gaining market shares due to their lower price and good reliability compared to tantalum capacitors. Hence, the niobium capacitors' market has seen its value double in the past five years and is expecting to keep growing as new opportunities for their use emerge [162,163].

For this reason, chemical processes to recover niobium are being developed. Montero et al. present a column process that is based on the leaching of crushed PCBs by a sodium cyanide solution of 4 g/L concentration [163]. This process, besides recovering niobium, also enables the recovery of gold, silver and copper and is said to be economically viable, with a 27% financial yield over 10 years, representing a net present value of USD 105,926. However, the high toxicity of cyanide raises important environmental concerns and there are still doubts if this process will constitute a real opportunity in the future. More environmentally friendly processes still need to be developed.

#### 4.1.2. Tungsten and Molybdenum

If tungsten was massively used in the early ages of electronics in vacuum (electron) tubes, there is nowadays very little of it in WPCBs, as it is mostly used as a copper migration thin film barrier of interconnects in integrated circuits, and despite this critical role, it only represents a minute mass fraction.

However, tungsten is present on the EU critical raw materials list and its recovery can have both economic and ecological benefits. The current production and recycling processes of PCBs produce wastewater, which contains small amounts of tungsten. Hence, Avinash et al. showed that they could selectively recover 80% of the tungsten present in wastewater by using an emulsion liquid membrane [164].

In electronics, molybdenum has also been used since the early days, for example in multilayer ceramic circuits, but can still be found in various modern ECs. It is used for thermal-management purposes as a required material in high-power diodes and rectifiers. It can also be a key component of field-emitter flat-panel displays, as well as physical-vapor deposition masks, which are not within the scope of this article.

Despite its price being in the tens of thousands of US dollars per metric ton (depending on trading date and metal grade), we could not find any research article or patent on the recycling of molybdenum from such ECs. Therefore, there is a possible opportunity arising here, whether from a research or business point of view.

#### 4.2. Platinum Group Metals (PGM)

Until 2010, of all the platinum group metals, only Pt and small amounts of Pd were recycled from WPCBs, yet only 5% of Pt consumed was recovered [165]. Due to low Pd reserves, limited to mainly Russia (41%) and South Africa (36%), along with a consistent increase in demand, Pd is considered strategic by Europe and the US [166]. Therefore, exploiting recycling streams such as WPCBs is an area of instant economic interest. On PCBs, the majority of Pd is used in multilayer ceramic chip capacitors (MLCC), while smaller portions can be found in conductive tracks in hybrid integrated circuits (HIC) and for plating printed circuits. It is calculated that Pd makes up to nearly half of the economic value derived from computers or laptops after the ECs have been disassembled and sorted [167]. However, little research has been conducted on Pd recovery in WPCBs [168–171]. The difficult part in Pd recycling is its enrichment from a base metal (mainly Cu) since the Cu content in MLCC is much higher than Pd and they have similar chemical properties [172]. The current methods for the selective leaching of Cu are inefficient, while Pd is partially dissolved during Cu leaching.

Some progress have been made, particularly on liquid–liquid extraction, which can now offer reliable, highly selective, highly purified metal ways of recovering Pd [173–175]. In 2017, has been in-depth investigation of Pd leaching in acids (including nitric, hydrochloric, or sulphuric) in a mixture with hydrogen peroxide [166]. However, until recently, all systems used either corrosive acids or strong oxidant to extract Pd. Zhang et al., proposed a multistep, environmentally benign, non-acid process to recover Pd from WPCBs [176]. First, Pd was enriched during Cu recovery and dissolved in  $\text{CuSO}_4$  and NaCl solution. Then, by using diisoamyl sulfide ( $(\text{C}_5\text{H}_{11})_2\text{S}$ ,  $\text{R}_2\text{S}$  for short) to extract the dissolved Pd to the organic phase ( $2\text{R}_2\text{S} + \text{PdCl}_4^{2-} \rightarrow \text{PdCl}_2 \cdot 2\text{R}_2\text{S} + 2\text{Cl}^-$ ), 99.4% of  $\text{Pd}^{2+}$  was separated at optimized conditions (10% diisoamyl sulfide, aqueous to organic ratio of 5, and 2 min of extraction time). It is noteworthy that there was an insignificant effect on the base metal (Cu) owing to the far stronger nucleophilic substitution of diisoamyl sulfide with  $\text{PdCl}_4^{2-}$ . Afterwards, 99.5% of the  $\text{Pd}^{2+}$  extracted was stripped out from diisoamyl sulfide/dodecane at an aqueous to organic ratio of 1, by forming a much more stable complex with ammonia ( $\text{PdCl}_2 \cdot 2\text{R}_2\text{S} + 4\text{NH}_3 \rightarrow 2\text{R}_2\text{S} + \text{Pd}(\text{NH}_3)_4\text{Cl}_2$ ), resulting in an overall dissolution–extraction–stripping Pd recovery rate of 96.9%. What made this process even more cost-effective and environmentally friendly was that the diisoamyl sulfide could be regenerated and used for up to five cycles. This, combined with the advances made

in dismantling and sorting PCBs, is giving hopes for a future in which the Pd consumed comes mainly from recycling.

Besides that, other Platinum Group Metals, including Os, Ru, Ir, and Rh, have not been recycled yet and therefore represent a recycling opportunity at the component level as they make up part of the active materials in transistors, thick-film hybrid circuits, integrated circuits (ICs), semiconductor memories, thick-film resistors, and conductors. Among them, Ru alloyed with Pt, Pd or Ir is the most important PGM in ECs (hard drives, hybridized ICs, etc.) [165].

#### 4.3. Gallium

Even though the stock price for gallium is near to its lowest value in ten years, with the consumption of electronic devices increasing steadily (almost doubling from 188 tons in 2006 to 331 tons ten years later), Ga production has experienced a high annual growth rate of ~20% in the period of 2004–2014 [177,178]. It is also expected that the global Ga market will expand by a third in the next four years and even faster when 5G is widely available [179]. It is important to note that Ga is also part of the EU's list of critical raw materials due to the availability and distribution on Earth criteria.

On PCBs, Ga exists in the form of gallium nitride (GaN), gallium arsenide (GaAs), and gallium phosphide (GaP) found in power transistors, optoelectronic components, and semiconductors in ICs. Approximately two thirds of the Ga used in 2010 was for ICs [180,181]. For instance, a 2011-generation smartphone contains 3–15 mg of Ga, which is steadily increasing with the fast development of the semiconductor industry [182,183]. Until now, due to its special characteristics, Ga was considered irreplaceable and labelled as a critical raw material for electronics. However, with high-quality requirements of electronic goods, merely 20–30% of Ga is eventually incorporated in the final products; the rest is lost during intermediate processes such as etching, polishing, etc. [184]. Although at the moment, Ga is being recycled from preconsumer scrap generated during semiconductor manufacturing, mainly from waste waters and liquid phase deposition, its recovery percentage only makes up 35% [165,184]. If Ga-based waste, especially GaAs, could be recycled properly, not only we would have a “new” plentiful source of Gallium but it would also be a big step toward avoiding the damage to organisms caused by arsenic. Specifically for Ga-based WPCBs containing GaAs and GaP, there are a number of means to retrieve Ga, namely, nitridation in ammonia, chlorination by chlorine gas, oxidation by oxygen, and thermal dissociation [184]. The main challenge in recycling Ga is due to the difficulty of separation from the other elements such as In, Sn, Ge, Pb, Ag, Cu, and Au. This is where the advances made in PCB dismantling and sorting described above will be useful and simplify that problem by lowering the number of elements in the mixture. For instance, with proper ECs sorting, Zhan et al. were able to recycle Ga from GaAs-based ICs by an efficient and environmentally friendly hydrothermal-buffering method [181]. In optimum conditions (275 °C; 15 min of residence time; 300 rpm; 5% (v/v) H<sub>2</sub>O<sub>2</sub> as the oxidant; buffer initial pH of 7.8, and buffer concentration 0.001 M), 99.9% of Ga and 95.5% of As were recovered.

#### 4.4. Rare-Earth Elements

Many rare-earth elements (REEs) are considered as critical elements in many countries [185,186]. Indeed, lack of access to them would cost the concerned economies dearly, as they play many functions in the electronics and energy sectors [187–189]. To minimize dependence, as well as reduce the environmental impact due to the heavy mining and processing (particularly purification by hydrometallurgy [190–194]) required for their production, they must be recycled [195–197]. This is even further emphasized when one takes into account the balance problem between the ore concentration in each of the REE that does not match their respective market's request, and leads to an overproduction of light REEs and an underproduction of heavy REEs [198]. In the field of WPCBs, one can find REEs in various places, such as permanent magnets (Nd, Pr, Gd, Dy and Tb), that are also needed in large amounts for other applications, such as wind turbines or electrical

vehicles. Hence, it is projected that within the next 25 years, the demand for neodymium and dysprosium's could increase by 700% and 2600%, respectively [199].

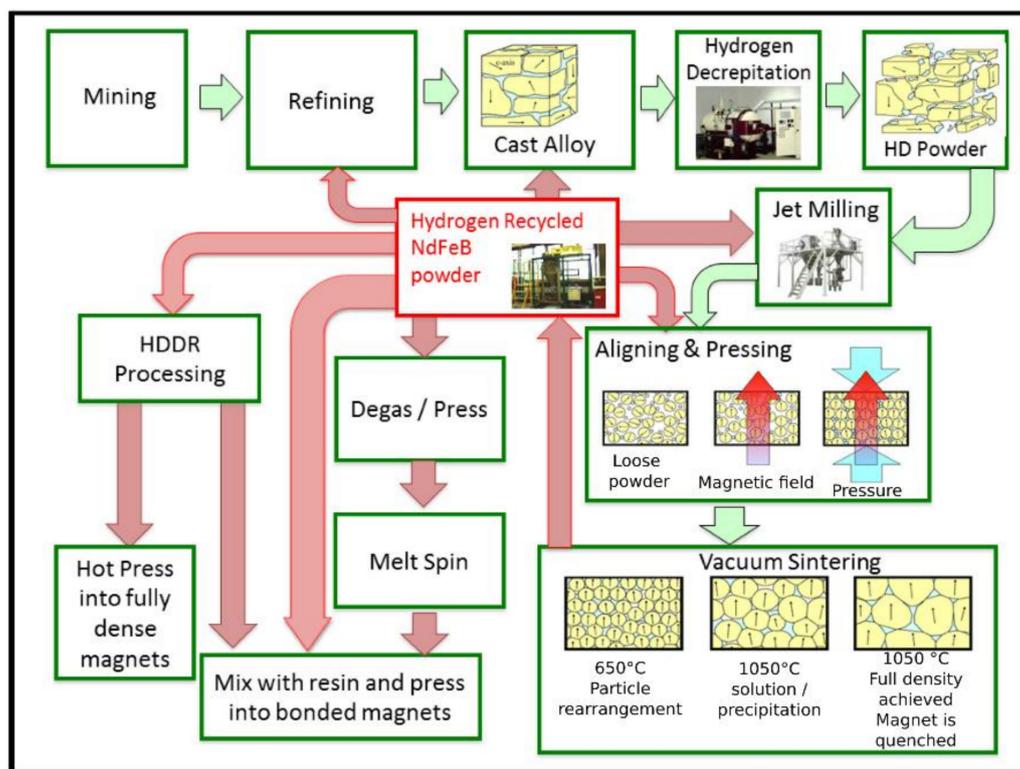
The recovery of REEs from whole WPCBs has been proven feasible but is challenging since their overall concentration is fairly low (in the tens of ppm), and recovering them requires high CAPEX equipment, such as smelters and large electron beam apparatus, as well as OPEX [200]. Such approaches can only be made profitable in large centralized operations. It therefore does not appear to be adapted for smaller communities nor for the development of sustainable processes. The dismantling/sorting approach seems here again perfectly adapted to increase REE concentration in the ore of urban mines. This is especially true because physical methods that use early grinding tend to break the magnet's protective packaging, leading to the deterioration of REE based magnets into magnetic powder that is attracted by magnetic machinery parts, where it quickly oxidizes and cannot to be retrieved [201–203].

Despite those difficulties, Joseph et al., studied the economic potential of a neodymium recovery process from ewaste [204]. Comparing three extraction processes based on a HDEHP-HCl system, an EHEHPA-HCl system and a P350 extractant system, the study concludes that the latter presents the best economic opportunities and that it would be suitable for recovering neodymium in Malaysia with optimum revenue return. This process would represent an annual revenue of USD 388,910 in the conditions described in the article.

Similarly to what has been said for tantalum, an ideal process would not oxidize the metallic alloy of the permanent magnet, and would avoid the reduction process costs. This is, however, more difficult here, as these alloys oxidize easily in air, hence their covering with a protective coating. Various groups have therefore reported a general process that first performs a hydrogen exposure, which creates a volume expansion of the magnet as metal hydrides are formed [205–210]. This allows the casing to break, and the alloy to turn easily into fine powder. This process also has the advantage that, if carefully performed, it protects the alloy from oxidation [211]. Additional milling, sometimes at cryogenic temperatures, has also been reported [205]. Once hydrogen is removed, by heating in neutral atmosphere (~200 °C), one can then reprocess the powder via compaction (with additives, binder, reducing agents, etc.) then follow with a sintering step to produce new permanent magnets with the best performances possible (Figure 7). Many reports and patents can be found on this last portion of the process [205–207,209,212–219], which most likely only represents the tip of the iceberg when compared to the unpublished trade secrets from the permanent magnet industry.

If, however, more purification or elemental separation must be performed, additional process steps are required such as: (i) chlorination using safer agents such as  $\text{NH}_4\text{Cl}$  [220,221]; (ii) advanced leaching [222–236]; (iii) or even bioleaching using bacteria which avoids the use of strong acids [237]; (iv) hydrometallurgy (solid–liquid or liquid–liquid extraction) [15,210,238–244]; as well as (v) electrodeposition at the reduction step, including in ionic liquids [245–253] and molten salts [254–259], with their wide electrochemical stability [260].

It should be noted that the weaknesses of the hydrometallurgy processes are their complexity and therefore the lack of in-depth understanding of their kinetics and thermodynamics. This creates a real issue when applying such processes to recycling. Indeed, if the composition of an ore coming from a mine is usually fairly constant, a wide range of compositions is observed from one recycling batch to another. This is a road blocking issue as one cannot spend years of R&D to develop a new process for each batch. To solve this bottleneck and enable a fast processing development, computer controlled and automated microfluidics platforms integrated with characterization methods (infrared [261,262], X-ray fluorescence [14], ICP [263]) have been developed. A review of the state of the art on that front was recently published by [14].



**Figure 7.** Possible magnet re-processing flowchart. Green frames indicate primary route for the making of sintered NdFeB magnet. Red frames with arrows indicate where recycled powder can be injected into production cycle. Adapted with permission from reference [209] Copyrights 2015, Published by Elsevier Ltd.

## 5. Conclusions

This review highlights the importance of dismantling and sorting for recycling WPCBs. The dismantling step is critical to ensure that all components are physically separated from the board in a safe, environmentally friendly, and economically viable manner. It must minimize the production of additional waste streams. From this point of view, the solder bath dismantling technique seems very harmful to both humans and the environment, whereas the use of Ionic liquids solves most of the issues of the former approach.

The sorting of ECs for the sole purpose of recycling is still an emerging field, hence a large portion of the literature on sorting, focuses on defect detection on ECs manufacturing lines. Nevertheless, with the acceleration of research in this field and an increase of industrial tool propositions, we expect this research field to grow significantly. To favour the use of machine vision, image recognition experts will need to build, or have access to, large high-quality datasets. Indeed, these vision systems are becoming increasingly performant by using ever improving machine learning algorithms. Convolutional neural networks could play a major role in classifying ECs based on images. Because of their low cost, vision systems should be at the core of the sorting strategies. However, vision systems do not target the chemical composition. For this reason, physical separation and spectroscopy techniques should be combined with machine vision. On the one hand, physical separation techniques are low-cost and effectively split the waste stream without the need to handle the ECs with an additional tool (e.g., diverters, pushers or robotic arms). The drawback is that they seem to be efficiently enriching the stream for only a handful of components. On the other hand, spectroscopy techniques can be expensive to implement, but have the potential to sort with high accuracy by targeting the elemental composition.

Overall, the EC sorting approach results in the production of many different waste streams that are each enriched in specific elements. Therefore, there are more streams than with conventional raw-crushing methods, with higher concentrations of the high value

elements. This might allow the integration of such waste streams in classical ore treatment plants or might yet require industries to develop new treatment lines and possibly increase their costs. Fortunately, these increased expenses could be compensated by the recycling of high-value metals using element-specific pyro- or hydrometallurgy processes, which until now could not be recycled. For example, tantalum recovery can be specifically recovered from some capacitors with recovery rates close to 100%. Other metals for which significant business opportunities are proposed in this review are Ga, Nb, W, Mo, Pd, Os, Ru, Ir, Rh, Nd, Pr, Gd, Dy and Tb.

Finally, although dismantling and recovery are the research focus of increasing numbers of publications, we note that yet more research is needed to address the sorting of ECs. In this regard, it is possible that innovative sorting approaches are already being used by recycling plants but are still part of industrial secrets. Nonetheless, it is positive to observe that the overall literature dealing with the recycling of electronic waste is growing exponentially in order to cope with the equally exponentially growing waste volumes.

**Supplementary Materials:** The following are available online at <https://www.mdpi.com/article/10.3390/su131810357/s1>, Figure S1: Diagram of the dismantling section, or ECs cutter unit, of the WPCBs recycling tool adapted from reference (Fang, Li et al., 2017) and described in the source article as composed of “a frame (1), a driving motor (2), two cutting knives (3), material carrier chain and belt (4, 6), an ECs collecting bin (5) with its bin support (8), a knife mounting fixture (7), a dust filter (9) and an exhaust fan (11).”, Figure S2: Water soluble ionic liquid-based reactor pilot (~1000 L) for the disassembly of WPCBs and the recovery of wide range of tin based solder alloys. Reproduced from CNIPA 2018 patent application CN201811488109.9A (Li, Dong et al.). Figure S3: Process flow of the co-processing of WPCBs with spent TSS and metal recovery using selective precipitation. Reprinted with permission from (Yang, Li et al., 2017) Copyright 2017, American Chemical Society. Figure S4: Recycling of WPCB by epoxy dissolution. (a) WPCB and the re-acting vessel. (b) The recovered PCB with its ECs. (c) Recovered fiberglass fiber with their SEM micrograph. Reprinted with permission from reference (Chen, Yang et al. 2019). Copyright 2019, Springer Nature Limited. Figure S5: Schematic illustration of SELFRAG Lab High Voltage Pulsed Power Fragmentation System. Reprinted with permission from reference (Duan, Han et al., 2018). Copyright 2018, Elsevier. Figure S6: Variation of an element concentration for each step of the sorting process. Reprinted with permission from reference (Lee, Kim et al. 2012), Copyright 2012, Elsevier B.V. Figure S7: Optical Image (a) and X-ray transmission image (b) of dismantled Tantalum and Aluminium capacitors. The components are manually labelled for the training of the neural networks. (c) Normalised confusion matrix showing the predictions accuracies of the combined model for each class.

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## Abbreviations

BMI<sub>m</sub>, 1-Butyl-3-methylimidazolium tetrafluoroborate; CAPEX, Capital Expenditure; CPU, Central Processing Unit; DMF, N,N-dimethylformamide; DMSO, Dimethyl Sulfoxide; DR, disassembling rate; EC, electronic component; EG, ethylene glycol; EM<sub>m</sub>, 1-ethyl-3-methylimidazolium tetrafluoroborate; HIC hybrid integrated circuits; IC, integrated chips; ICP, inductively coupled plasmas; IC, integrated circuit; IL, ionic liquid; IR, Infrared radiators; LDH, layered double oxide; LIBS, Laser induced breakdown spectroscopy; MIBK, methyl iso-butyl ketone; MLCC, multilayer ceramic chip capacitors; NMP, N-Methyl-2-Pyrrolidinone; OPEX, Operating Expense; PCB, Printed circuit board;

PGM, Platinum group metals; RAM, Random-Access Memory; REE rare-earth element; RGB, Red, Green and Blue; SCF, supercritical fluids; TBD, 1,5,7-Triazabicyclo[4.4.0]dec-5-ene; TBP, tri-butyl phosphate; TSS, tin stripping solution; WPCB, waste printed circuit board; XRF, X-ray fluorescence.

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