metals

Review

Sustainable Recovery, Recycle of Critical Metals and Rare Earth Elements from Waste Electric and Electronic Equipment (Circuits, Solar, Wind) and Their Reusability in Additive Manufacturing Applications: A Review

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Abstract: The demand for high-efficiency, low-energy consumption materials, with high durability and stability, has led to the rapid increase of the demand and prices of Rare Earth Elements (REE). The REE monopoly of some countries has held the shift of humanity towards sustainability and renewable energy sources back. The isolation, recovery, and recycle of REE from waste electric and electronic equipment (WEEE) constitute the disengagement strategy and can lead to significant economic benefits, via sustainability. The introduction of critical raw materials (RM), derived from WEEE, as additives to filaments used for the synthesis of composite materials, employed by Additive Manufacturing (AM) applications, has tremendous potential for the performance and the commercialization of the final products by adding unique characteristics, such as antibacterial properties, enhanced mechanical and magnetic properties, and thermal and electrical conductivity. The low cost of the recycled RM, the small numbers of process stages, and the inception of a zero-waste paradigm, present its upscalability, with a realistic view to its industrial employment. Although there are many articles in literature that have reviewed WEEE recycle, a comprehensive review on the conditions, parameters, procedure flow charts, and novel properties of the final composite materials with regards to every RM is missing.

Keywords: rare earth elements; critical raw material isolation; recovery; recycle; circular economy; additive manufacturing

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1. Introduction

The production of enormous quantities of waste electrical and electronic equipment (WEEE) is taking place globally, especially in industrialized countries such as the USA, Europe, Korea, India, Greece, China, etc., due to the rapid economic growth coupled with the swift change of technological advancement in the field of electrical and electronic equipment [1]. WEEE contains large quantities of metals, for example, REE, that can be isolated, recovered, and recycled. On the one hand, their isolation, recovery, and recycling can have substantial environmental benefits, by decongesting landfills of potentially hazardous materials and compounds thereof that can result in elevated risks of cancer and neuro-logical disorders [2]. Further, their reuse in added value materials, following the technological requirements, can lead to economic and societal benefits. More specifically, WEEE contains more than 1000 different substances, which fall under 'hazardous' and 'non-hazardous' categories. Broadly, it consists of ferrous and non-ferrous metals, plastics, glass, wood and plywood, concrete and ceramics, rubber, and other items. Iron and steel constitute about 50 wt% of the waste followed by plastics (21 wt%), non-ferrous metals (13 wt%), and other constituents. Non-ferrous metals consist of metals like copper (Cu), aluminum (Al), and precious metals, e.g., silver (Ag), gold (Au), platinum (Pt), palladium

(Pd), etc. The presence of elements like lead (Pb), mercury (Hg), cadmium (Cd), selenium (Se), hexavalent chromium (Cr), and flame retardants (FR) beyond threshold quantities, classifies them as hazardous waste [2].

REE are present in large quantities in the Earth's crust, but their mixing with other minerals and the resulting low purity leads to their costly extraction. Due to the similarities of their physical and chemical properties, they are usually found together in geologic deposits and their separation process is rather costly [3]. Combined with the relative monopoly of some countries (China has the largest share of REEs production and is followed by the US), their price has risen sharply in recent years [4]. China is a pioneer in the development of REE mining technologies. The need for product development with innovative properties, especially in the last two decades, has led to a rapid increase in demand and in the price of REE. In particular, the shift of humanity to sustainability and renewable energy sources (solar, wind), the demand for extremely high-efficiency and low energy consumption materials combined with high durability, miniaturization ability, and thermal stability, has turned the attention of the scientific community to REE [4].

The properties of REE are mainly due to the distribution of electrons in the outer layer, occupying part of 4f orbital positions. Although the distributions of their electrons are similar, their physical and chemical properties differ significantly, making each element and its application unique, in a wide range of technologies [5].

The current recycling rate of REE is extremely low (only 2% of REEs are recovered by recycling processes against 90% of iron and steel) [6]. Despite their low recycling rates, REE are expected to be used even more in the near future, as their demand grows rapidly. WEEE, batteries and magnets represent a significant opportunity for REE supply chain balance. According to Patil et al. [6], at least 10% of REE used in batteries and magnets and 17% in phosphor lighting could be recovered through recycling processes. Finally, around 10 tons of Tb and 230 tons of Nd (both elements are short in supply) are estimated to be recoverable from WEEE streams every year [6].

In the study of Gutiérrez-Gutiérrez et al. [7], landfill sites were analyzed and it was observed that REE concentrations did not differ significantly. Ce was the most abundant rare metal with a mean concentration of 17 mg/kg of waste. The concentration of Nd ranged between 8.5 and 12 mg/kg and La between 7 and 10 mg/kg. Significant quantities of Cu and Al (mean concentration of 1500 and 15,000 mg/kg, respectively) and fewer quantities of Au and Ag (mean concentration of 0.15 and 3.5 mg/kg, respectively) were also observed. The mobility analysis of the critical metals showed that they are not being vertically transported since concentrations remained similar through the whole range of depth in the landfills [7].

The European Union has increased the priority of regulations on circular economy, WEEE management, sustainability, and environmental agenda. As exponentially increased amounts of WEEE are wasted every year, the excessive need for their recovery, reuse, recycling, or disposal is crucial. Closed-loop supply chains and infrastructures of recycled critical raw materials need to be supported or financed from each respective country's initiatives and the EU as a coordinator, as WEEE European Directives define the requirements for national collection systems but leave to each Country-Member States the responsibility to undertake specific policies and to reach the fixed targets [8]. The analysis of both availability and accessibility of WEEE and the logistics of the closed-loop supply chains will be proven beneficial for policy authorities, as they will constitute diagnostics data which will help them intervene where needed [8]. As recovery and recycle technologies become more feasible, companies will adopt them and WEEE will be treated on site, with high probability of ensuring availability and accessibility in the aforementioned collection points.

New applications, such as magnets, catalysts, batteries, glass screens, and phosphorescent pigments, represent more than 60% of the market, as shown in Table 1, which presents the quantities of REE used in new technologies and intermediates [9]. Each intermediate product can use more than one from a total of 17 REE. In the permanent magnets that find application in wind turbines, praseodymium, neodymium, gadolinium, dysprosium, terbium, samarium (Pr, Nd, Gd, Dy, Tb, Sm, respectively) are used. Magnets are the market with the highest quantitative requirements of REEs, followed by catalysts for oil refining, where lanthanum and cerium (La, Ce) are used [6,7].

Table 1. Global requirements and application of Rare Earth Elements.

Application	Quantity (Thousand Tons)	Market Share (%)
Permanent magnets	51	30
Catalysts	31	18
Metal alloys	30	18
Polishing powder	22	14
Glass	9.5	6
Ceramics	8.5	5
Phosphorescent pigments	5	3
Rest	10.5	6
Total	167.5	100

Current global REE production is approximately 130,000 metric tons of rare earth oxide equivalent (REO) content per year [10].

Printed circuit boards (PCB), monitors, cell phones (CP), photovoltaic modules (PV), and wind turbines (WT) constitute technologies that require technologically advanced electrical and electronic equipment. The aforementioned technologies require specific management of their equipment at the end of their life cycle (EOL), as some of their constituents can be considered hazardous. More specifically, despite an enormous amount of WEEE being generated every year in the U.S. and EU, their treatment relies simply on incineration or landfill [9–14].

2. Critical Raw Materials in WEEE

The electrical and electronic industry is one of the most innovative in terms of products, and this is reflected in the speed with which electronic devices such as CP and personal computers become obsolete, generating large amounts of electronic waste that must be treated [15].

2.1. Printed Circuit Board

PCB are employed in most of the electronic equipment to mechanically support and electrically connect the components using conductive pathways [16]. Practically all electronic devices contain PCB, which are composed of three types of materials: a non-conducting substrate or laminate, printed conducting tracks, and components mounted on the substrate. The substrate is typically composed of glass-fiber-reinforced epoxy resin (that can be used in additive manufacturing), or paper reinforced with phenolic resin (primarily designed to insulate the Cu circuits on outer layers from oxidation from the environment), both with brominated FR. Once the resin layers are completed, identifying information, marks, and sometimes barcodes are printed on them. These marks are called nomenclature [17]. Fiberglass and resin constitute around 40 wt% of the PCB [18]. Printed conducting tracks and components mounted on the substrate are comprised of precious metals and REE. Four main types of materials can be retrieved from PCB: (i) recyclable metals (Cu, Al, Pb, Au, Ag, Pt, Pd) and REE (such as Nd, La, Ce). Boards that have been produced recently may not have Pb in their composition, but may contain other metals such as bismuth (Bi) or Ag; (ii) recyclable polymeric materials; iii) ceramic materials, which can be reused or disposed of more appropriately if they are free of metals; iv) non-recyclable polymers or other contaminants, from which energy can be recovered by combustion and incineration [15].

Compared to traditional wired circuits, PCB offer a number of advantages. Their small and lightweight design is appropriate for use in many modern devices, while their reliability and ease of maintenance suit them for integration in complex systems. Additionally, their low cost of production makes them a highly cost-effective option. They are used in medical, aerospace, military, commercial, and industrial applications [17]. When comparing Printed Wiring Boards (PWB) with PCB, the biggest distinction is that PCB refers to a board with the whole circuitry and components, whereas PWB refers to a board without components. This implies that PCB are finished circuit boards that are ready for installation in the electronic device. The term PWB is used to indicate the rudimentary production of the circuit board or the use of circuit boards in electronics that do not require complex functionality [19].

PCB can also be found in CP. As CP have become an amenity, the desire for upgrade has become a way of thinking. Only in the US, more than 151 million CP were discarded in 2018 [20]. Recycling waste CP would not only be an economic advantage for waste recyclers but could also help avert an environmental catastrophe as they (including their battery) contain Pb, Cd, Hg, nickel (Ni), antimony (Sn), arsenic (As), and bromine (Br), which can pollute the air, soil, and water if improperly managed [21]. Additionally, recycling waste CP could be considered a type of secondary mining of these precious REE, creating a new supply chain for the booming electronics industry and thus reducing primary metal mining. The material composition and the design (partially) of CP makes recycling and dismantling very complex, mainly because of the presence of a wide variety of materials in the devices, such as plastics, glass, and ceramics. Plastics and PCB make up the heaviest parts of a CP; plastic represents almost 40–50 and PCB 20–25 wt%, respectively. In terms of recycling value, PCB are the most important modules of waste CP as they contain most of the precious and valuable metals [22]. The composition of PCB from computers and CP is shown in Table 2 and the price of the metals found in PCB is shown in Table 3.

Table 2. PCB composition derived from personal computer and cell phone (CP) waste.

<table>
<tr><th>Type of WEEE</th><th colspan="12">Metals (%)</th><th>Literature</th></tr>
<tr><td rowspan="2">PCB
PCB (CP)</td><td colspan="2">Pb</td><td colspan="2">Ni</td><td colspan="2">Al</td><td colspan="2">Fe</td><td colspan="2">Cu</td><td colspan="2">Zn</td><td rowspan="2">[18,23–25]</td></tr>
<tr><td colspan="2">2–3
0.3</td><td colspan="2">2
0.1</td><td colspan="2">7
1</td><td colspan="2">12
5</td><td colspan="2">16
14</td><td colspan="2">1.5–2
1–2</td></tr>
<tr><td rowspan="3">PCB
PCB (CP)</td><td colspan="12">Metals (ppm)</td><td rowspan="3">[15,22,24,
26,27]</td></tr>
<tr><td>Au</td><td>Ag</td><td>Pd</td><td>Nd</td><td>Cd</td><td>Pr</td><td>La</td><td>Y</td><td>Ce</td><td>Gd</td><td>Sm</td><td>Dy</td></tr>
<tr><td>110
186</td><td>280
1600</td><td>-
35</td><td>831
269</td><td>1000
1000</td><td>100
30</td><td>361
60</td><td>57
40</td><td>20
8</td><td>75
30–70</td><td>55
41</td><td>75
44</td></tr>
</table>

Table 3. Price of metals found in PCB.

Metals	Price (€/kg)—07/2021	Literature
Lead (Pb)	1.94	[28]
Nickel (Ni)	15.7	[29]
Aluminum (Al)	2.1	[30]
Iron (Fe)	0.18	[31]
Copper (Cu)	8	[32]
Zinc (Zn)	2.49	[33]
Gold (Au)	49195	[34]
Silver (Ag)	707.3	[35,36]
Palladium (Pd)	76700	[37]
Neodymium (Nd)	92.5	[38]
Cadmium (Cd)	2–3	[39]
Praseodymium (Pr)	565	[40]
Lanthanum (La)	144.5	[41]
Yttrium (Y)	27	[42]
Cerium (Ce)	381	[43]
Gadolinium (Gd)	64	[44]
Samarium (Sm)	91.5	[45]
Dysprosium (Dy)	1050	[28]

2.2. Thin-Film Photovoltaic Cells

Thin-film photovoltaic cells (PV) are made by placing one or more films of compounds of photosensitive materials that act as semiconductors on a support substrate such as glass, plastic, or metal. Two main categories of thin-film PV that use REE are examined:

Thin-film Cadmium–Tellurium (CdTe) photovoltaic cells promise lower costs than crystalline silicon photovoltaic systems, as their manufacturing process is cheaper, and due to the extremely small dimensions of the membrane, very small quantities of raw materials are used. Specifically, the cell thickness is in the range of 0.5 μM [46]. The yield of CdTe cells ranges between 11–14% and their lifespan can exceed 15 years [47]. One of the advantages of CdTe cells is that they absorb shorter wavelengths of sunlight compared to crystalline and amorphous silicon cells [47]. The construction of CdTe cells requires the least amount of water, compared to the other solar energy technologies [48]. Like amorphous silicon PV cells, CdTe cells have good heating resistance. PV in their plating, coatings, and wiring contain elements and compounds thereof (such as Cd, Pb, Cr, etc.), which can become dangerous to public health in large concentrations, especially due to the risk of leaching into the surrounding soil [49,50].

Cu, indium (In), gallium (Ga), Se alloy (CIGS) thin-film photovoltaic cells (CIGS): Due to the absence of toxic Cd, but also due to the increased efficiency (14–18%) compared to CdTe, CIGS thin-film PVs have received a large market share, as they are the most effective among thin-film photovoltaic technologies. CIGS cells are manufactured by depositing a thin film on a substrate (glass or plastic) [51,52]. Like crystalline silicon cells, CIGS cells show reduced efficiency due to heating [47]. Disadvantages include the complexity of combining four components, which makes upscaling difficult and costly [53]. Finally, like CdTe, CIGS have a shorter lifespan (approximately 12 years) and require maintenance more frequently than silicon cells to allow long-term outdoor operation [54].

CdTE PV modules account for 6–7% of photovoltaic systems worldwide, while CIGS PV account for 2% [55,56]. In July 2012, the European Union formally revised the WEEE guide, adding photovoltaic components as discarded electronic devices in the WEEE categories [57]. REE in PV systems represent less than 1% of the panel volume (a typical case study recommendation of Aman et al. [58], but their value is significant for the reasons mentioned above [59]. The composition of thin-film PV modules is shown in Table 4 and the price of the metals found in PV is shown in Table 5.

Table 4. Composition of thin-film PV systems.

Composition (%)	CIGS	CdTe
Glass	82	95
Aluminum (Al)	11	<0.01
Silicon (Si)	-	-
Polymers	5.5	3.5
Zinc (Zn)	0.12	0.01
Lead (Pb)	<0.1	<0.01
Copper (Cu)	0.85	1
Indium (In)	0.02	-
Selenium (Se)	0.03	-
Tellurium (Te)		0.07
Cadmium (Cd)		0.07
Silver (Ag)		<0.01
Gallium (Ga)	0.01	-

Table 5. Price of metals found in PV.

Metals	Price (€/kg)—07/2021	Literature
Indium (In)	190–200	[60,61]
Gallium (Ga)	200–220	[62]
Germanium (Ge)	950–1000	[63]
Tellurium (Te)	90	[64]
Selenium (Se)	20	[65]

2.3. Wind Turbines

REE such as Nd, Pr, Dy, Tb are used as raw materials to manufacture neodymium–iron–boron (NdFeB) permanent magnets, which are used as components in generators for WT and in traction motors for electric vehicles [12]. Due to the green and digital transition, REE and permanent magnet value chains are considered crucial for European industrial and financial development. If these value chains move towards outside the EU, the financial ramifications will have an immediate, negative impact towards the green transition.

Although there are different types of permanent magnets, NdFeB are the most commonly used because of their outstanding properties. In terms of their properties, they are equaled only by samarium–cobalt (Sm-Co) magnets; however, these magnets are significantly more expensive [66,67]. Nd and Pr contribute to the magnetic strength, while Dy and Tb improve resistance to demagnetization, particularly at high temperatures. The exact composition of REE within an NdFeB permanent magnet can vary, with different proportions of the different elements leading to different magnetic properties. Nd and Pr can in principle be substituted by other elements, but this is often limited by the operating condition specifications. In the past few years, research has focused on the optimization and potential substitution of the use of Dy and Tb, which are both costly and poorly available elements [67].

In WT, the nacelle containing the magnet and the gearbox weighs as much as 1/3 of the turbine, with the other 2/3 distributed between the tower (mainly cement) and rotor blades (mainly cast iron and steel). For WTs, the composition of an average permanent NdFeB magnet, according to [25,68], is shown in Table 6.

Table 6. Composition of thin-film PV systems.

Composition (%)	Permanent Magnet	Quantity Needed for Energy Production (kg/MW)
Neodymium (Nd)	29.3	198.83
Dysprosium (Dy)	2.7	18.15
Boron (B)	1	-
Terbium (Tb)	0.01	0.54
Iron (Fe)	67	-

An average magnet weighs up to 4 tons [55]. The average mass of a magnet for the production of 1 MW is 680 kg [25,68]. For the main three critical metals, the quantity needed for the production of 1 MW of wind energy is also given [69].

While Dy is used almost exclusively in magnets, Nd, Pr, and Tb also have other applications. Nd and Pr are the most versatile as they are used in batteries, catalysts for cars, specialty glasses, and ceramics; Pr is also used in polishes for particular types of glass. Terbium green phosphors are largely used in fluorescent lamps and in light-emitting diode (LED) screens for televisions [70].

Once ores containing the four aforementioned REE are extracted, they are sent to a refinery for separation and refining of the different elements into individual oxides or metals, which are then used in different manufacturing processes. In the case of permanent magnets, the metals are melted together under a vacuum to obtain an alloy, which is then often turned to powder. There are two methods available for producing magnets from the powder, discovered by General Motors and Sumitomo Special Metals [12,71]. As part of the Hitachi Corporation, Sumitomo Special Metals developed and currently manufactures and licenses other companies to produce full-density sintered $Nd_2Fe_{14}B$ magnets and holds more than 600 patents covering neodymium magnets [72]. In the first method, the alloy powder is mixed with polymers to create a moldable putty (bonded magnets), whereas in the second method, the powder is pressed together under a specific heat profile (sintered magnets). In both cases, magnetization can be induced either during the treatment of the powder or in a separate phase of the manufacturing process. The magnets are then often arranged into arrays for specific applications. Sintered magnets are usually more compact and have better magnetic properties, constituting over 90% of NdFeB magnet production [67]. While the price of some metals is given in Table 6, the price of other metals, used in WTs, is given in Table 7.

Table 7. Price of metals found in PV.

Metals	Price (€/kg)—07/2021	Literature
Boron (B)	70	[73]
Terbium (Tb)	2031	[74]

3. Recovery and Recycling of Critical Raw Materials from WEEE

The main criteria under which the state-of-the-art methods are presented include the following. The flow-chart of the process needs to be composed of a small number of environmentally friendly techniques (such as mechanical removal of parts from WEEE) that are less energy- (and subsequently cost-) intensive, and ensure the use of non-hazardous chemicals during every stage. Furthermore, heat treatment of WEEE must be in compliance with safe-by-design protocols of capturing dangerous and toxic emissions. Finally, the applicability of each method at a laboratory scale with potential upscaling is a major requirement.

3.1. Printed Circuit Boards

As mentioned above, PCBs are fundamental components of almost all electric and electronic equipment. The rapid development of technology has led to the disposal of large quantities of waste PCBs (WPCB). Mechanical and pyrometallurgical methods are the current ways of recycling WPCB. Almost all WEEE recycling enterprises in China use various mechanical methods (such as multi-crushing, grinding, electrostatic separation, gravity separation, fluid-bed separation, density-based separation, and magnetic separation) to separate metals and non-metals from WPCB. After the mechanical separation, approximately 30 wt% of enriched multi-metal components (Cu, Al, Fe, Sn, Sb, Pb, etc.) are separated from the non-metal components. The non-metallic components are often sent to produce modified phenolic molding compounds or non-metallic plates. On the other hand, the enriched multi-metal components are often sold to metallurgy plants (e.g., copper smelting plants) as raw materials to be charged with other ore concentrates to recover valuable metals. However, existing pyrometallurgical and hydrometallurgical recovery processes generate atmospheric pollution because of dioxins, furans, and high volume of effluents release [75,76].

According to Jadhav and Hoecheng [77], the metal content of WPCB can be as high as 40%. Pyrometallurgical recovery processes require heating the WEEE at high temperatures to recover valuable metals. These processes lead to the production of hazardous flue gases that must be removed from the air. Furthermore, they are energy- and cost-intensive and require high purity feeds. Hydrometallurgical processes offer a relatively low capital cost, reduced environmental impact, and high recoveries of metals. They involve the dissolution of metals in alkaline or acid media. Several studies have reported the use of nitric acid (HNO_3), hydrochloric acid (HCl), and sulfuric acid (H_2SO_4). While the same amount of metals can be recovered from large pieces of PCB and pulverized PCB, the polymeric part of the board is easily recyclable in the case of large PCB pieces.

In the study of Jadhav and Hocheng [77], five different acids, i.e., HCl, HNO_3, H_2SO_4, acetic acid ($C_2H_4O_2$), and citric acid ($C_6H_8O_7$) were used as lixiviants for metal recovery from PCB pieces. HCl and HNO_3 removed all metals present on the PCB pieces. HCl required only 22 h to remove all metals from the PCB piece, whereas HNO_3, 96 h. It was also less expensive, making it the appropriate leachant. Since PCB remained in large pieces, the problem of precipitate contamination when recovering the metals from the leach liquors could be avoided. Various options were available for the purification and selective recovery of the metals from the leach liquor, including solvent extraction, adsorption on activated carbon, ion exchange, precipitation, filtration, and distillation [77]. The flow chart is shown in Figure 1.

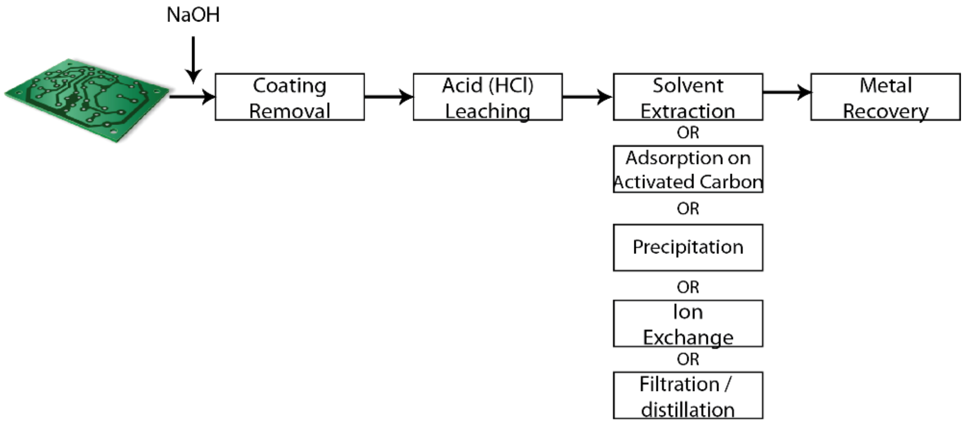

Figure 1. Recovery of PCB metals based on hydrometallurgy with acid leaching.

According to Behnamfard et al. [78], a novel hydrometallurgical process can lead to selective recovery of Cu, Ag, Au, and Pd from WPCB. Their method presents high efficiency (more than 99% recovery for Cu, 90% for Ag and Au, and 100% for Pd), low cost and process time, and is environmentally friendly. In their method, the grounded (less than 300 µM) WPCB content was leached by using two consecutive H_2SO_4 leaching steps in the presence of hydrogen peroxide (H_2O_2) as oxidizing agents. The solid residue of the first leaching was subjected to a second leaching step and the solid residue of the second leaching was treated by acidic thiourea ($SC(NH_2)_2$) in the presence of ferric iron (Fe^{+3}) as an oxidizing agent. The Cu recovery during the second leaching step was more than 99%. The precipitation of Au and Ag from acidic thiourea leachate was optimized by using a specific amount of sodium borohydride ($NaBH_4$) as a reducing agent, reaching 84 and 71%, respectively, in the third step. Finally, the leaching of Pd and remaining Au from the solid residue of the third leaching step was performed in a (sodium hypochlorite-) $NaClO\text{-}HCl\text{-}H_2O_2$ leaching system and the precipitation was optimized by using a specific amount of sodium borohydride ($NaBH_4$) [78]. The flow chart is shown in Figure 2.

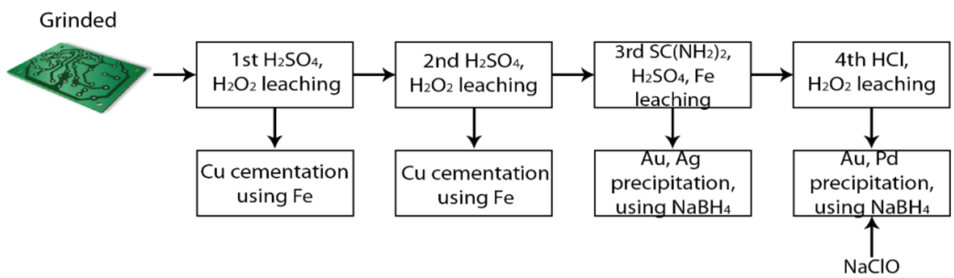

Figure 2. Recovery of PCB metals based on hydrometallurgy with sulfuric acid, hydrogen peroxide, and acidic thiourea leaching.

According to the study of Xiao et al. [79], the melting of 0.1–8 mm WEEE was performed in alsint (Al_2O_3) crucibles in a chamber furnace at 1400 °C for 1 h, under nitrogen (N_2) atmosphere. Two homogeneous molten phases were formed: slag and metal. The total content of the six base metals (Cu, Fe, Sn, Zn, Pb, and Ni) accounted for around 60% and Cu constituted an average 75% of these six metals in the metal molten phase, whereas metal oxides were mostly formed in the slag molten phase. Two major potential leaching agents were evaluated: H_2SO_4, and ammonia–ammonium (NH_3-NH_4^+) salt solutions. H_2O_2 was also added as an oxidizing agent, to speed up the leaching and enable a total leaching efficiency of 46% for the sum of base metals (Zn, Pb, Ni); the Cu leaching efficiency could reach 90%. Zn, Pb, and Ni could be further recovered using selective solvent extraction. Due to the relatively high concentration of Cu in the NH_3 leaching solution (about 20–70 g/L), Cu electrowinning directly from the solution was used for its extraction. Cu of 98.9% purity could be obtained via direct electro-deposition in the NH_3-NH_4^+ salt solution [79]. The flow chart is shown in Figure 3.

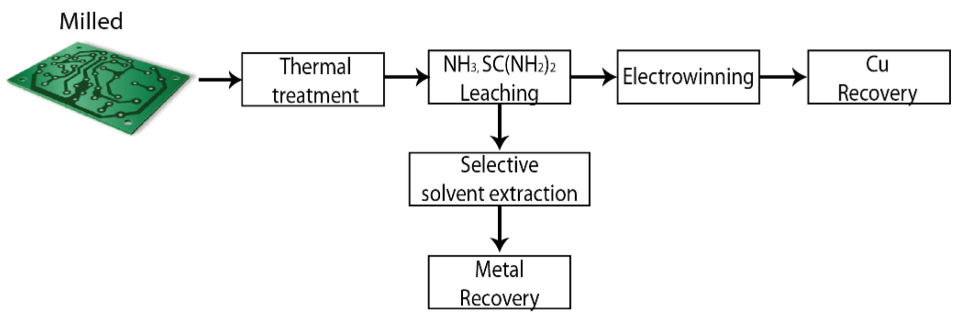

Figure 3. Recovery of PCB metals based on hydrometallurgy with ammonia leaching and thiourea leaching.

As mentioned before, PCB contain a significant amount of REE. Khanna et al. [24], in their study, used WPCB of different origin (computers, CP, motherboards) and proposed a recovery method for REE. The first step included the heat treatment of the WPCB, which was carried out in two different types of furnaces: a horizontal resistance furnace, and a thermal plasma furnace. The plasma furnace generates a high level of turbulence in the heating zone, whereas the only turbulence in the horizontal furnace is caused by the continuous flow of inert gas. At temperatures above 700 °C, a negligible amount of harmful compounds is produced. Lower temperatures than the melting point of Cu were used, as higher temperatures could potentially lead to the formation of mixed alloys [80]. The weights of WPCB were recorded before and after the heat treatment. A Cu-rich red fraction, Al- and Pb-rich whitish metallic fraction, and a dark carbonaceous ceramic phase fraction were the products of the heat treatment. Most of the REE were concentrated in the slag-rich carbonaceous fraction with negligible levels in the Cu-rich as well as in the Al/Pb-rich metallic fractions. This result is observed for all PCB under investigation and for both types of furnaces. The REE recoveries from pyrolysis in the thermal plasma furnace were consistently higher than the corresponding recoveries in the horizontal furnace. These results showed that turbulence in the furnace played an important role in dislodging REE from their host systems and their subsequent recovery in pyrolysis residues. According to the high recovery of La, Pr, Sm, and Y in the plasma furnace compared to an almost null recovery in the horizontal resistance furnace, these REE were tightly bound in PCB and require turbulent conditions for their release, whereas Nd, Gd, Ce, and Dy were relatively easy dissociated and released from the waste (they presented higher recoveries in the plasma furnace, but the recovery from the horizontal resistance furnace was also significant). By using plasma furnace, REE recovery yields increased from 118% to 2500% for various elements. The recovery of a wide variety of REEs from waste PCBs could be proven beneficial for economies with little or no REE resources of their own. Moreover, WEEE recycle will be a sustainable solution towards the problem of the ever-increasing WEEE volume [24].

There have been thorough studies about bioleaching: for example, biological alternatives emerged to reduce the negative environmental impacts caused by the application of hydro- and pyrometallurgical methods, such as generating corrosive residues and consuming large amounts of energy [81]. Most of the studies involve agitated bacteria and fungi cultures, where a stirring device that consumes energy and increases the cost is necessary. The upscaling of these methods would demand high energy input, determining the viability of the process. According to the study of Argumendo-Delira et al. [81], alternatives such as cultivation without agitation in order for the cost to be reduced must be considered. In their study, they used shredded CP, PCB, sterilized with ethanol and digested with HCl/HNO_3. As a carbon source for the fungal consortium (*Aspergillus niger* MPE6 and MX7), they used glucose. They concluded that the use of fungal consortia in a culture without agitation could be an alternative for the recovery of Au from WEEE with lower energy expenditure than in cultures with agitation, while maintaining the environmental hazards to a minimum. The average Au recovery ranged between 30–40%. Furthermore, acidophilic bacteria are widely used in the bioleaching of metals such as Cu, Al, Co, Mo, Zn, Pb, and Ni [82,83]. They can be used as a pretreatment to the PCB to increase the Au/Cu ratio (up to 80% Cu removal), as the excess Cu does not favor Au leaching [78].

3.2. Thin-Film Photovoltaic Cells

The recycling of CdTe thin film in second-generation PV systems, in addition to minimizing the toxicological risk of Cd, brings additional benefits. The increased demand for high-purity Cd and Te can be partially offset by the recovery of the latter from EOL modules, contributing to a relative stabilization of their price. The recycling approach of Sasala et al. [84] was based on low cost, minimization of the environmental impact, and application both to EOL modules, as well as to systems out of specifications that present infantile failures. Any recycling technique for CdTe and glass must first effectively separate them by mechanical or chemical means. The initial separation took place by crushing the frame with a hammer mill and heat treatment, where the thermoplastic polymer film (consisting of ethylene vinyl acetate—EVA) was burned, making the separation possible.

Mechanical removal of semiconductors was achieved by impinging the substrate with water jets of high pressure at 40,000 psi. The method was superior compared to the chemical stripping of semiconductors as the required metals do not come into contact with chemicals. The disadvantage was that the mechanical removal was applied to glass surfaces larger than 0.1 m^2; therefore, it concerned systems that were out of specifications or intact EOL systems, as shown in Figure 4A. Due to the crushing, there were glass fragments with a smaller surface area, so a parallel recycling process was necessary. Chemical stripping with acids was applied to them. Specifically, the glass fragments were immersed in an acid bath, to dissolve the metals and isolate clean glass, which was recycled. The acids used were mainly H_2SO_4 and H_2O_2 under stirring, achieving chemical etching as the semiconductors were stripped of the fine glass. Due to the low solubility of CdTe, portions of the membrane floated in the acid solution and precipitated more slowly than the glass under stirring, facilitating their recovery by collecting the supernatant. Specifically, the first step was to precipitate the majority of Te by adding an alkaline solution to the liquid (mainly sodium hydroxide—NaOH). The second step, after filtering the precipitate, for the recovery of Te, was the recovery of the remaining metals (including Cd) by electrolytic plating with a large-surface Cu mesh electrode, of more than 93%, and finally the electrolytic removal of Cd from the remaining metals of the system with a second electrode. The remaining acid could itself be recycled and used to further strip PV modules [84]. The above is shown in Figure 4B.

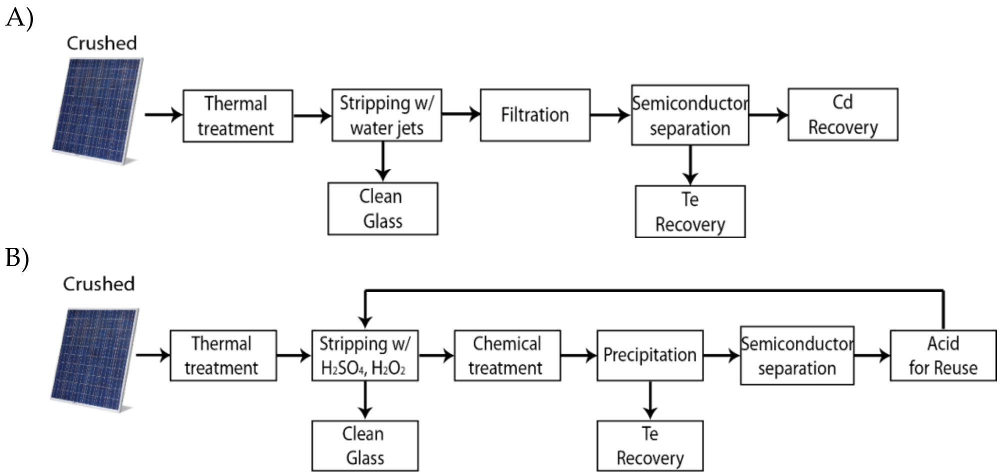

Figure 4. Recycling of CdTe thin-film PV module flow chart using (**A**) water jets, (**B**) chemical stripping.

The cost of mechanical removal of semiconductors reached 0.06 dollars/Watt (W) (capital expenditure, operational expenditure, labor) due to the high required water pressure, while the cost of chemical treatment was 0.04 dollars/W based on a PV system with a capacity of 1 MW/year. Mechanical removal did not use chemicals, which could affect the materials, while their disadvantage was the higher value per W and their application exclusively on large glass surfaces [84]. Even at higher cost, they [84] concluded that the cost was affordable and the recycle process feasible.

The above process was also presented by First Solar, a company manufacturing PV modules, which in the company brochure, states that the EOL PV modules that return to the company are grounded in a shredder and hammer mill to separate the polymer film from the semiconductors. The fragments are then washed with acids, during which the pure glass is separated by about 90%. The mineral-rich liquid can be refined, by external partners, to isolate and recover more than 90% of Cd and Te [85].

It is noted that for a reference solar panel (assumed to be First Solar Series 3^{TM} Black PV module, First Solar, Perrysburg, OH, USA) of an area of 0.72 m^2 and weight of 12 kg, Cu, Cd, and Te constitute 0.011, 0.059, and 0.075 wt%, respectively [86]. Their recycle rate is 90%. From the result of their optimization model, the whole recycling process could make a profit of up to 1.58 dollars per module [86].

In their study, Berger et al. [87] aimed to achieve sustainable strategies for recycling thin-film PV modules, which were based on (liquid) mechanical processes to reduce the amount of chemicals used in conventional recycling, as well as the amount of waste. Two main strategies have been developed, depending on whether the PV module was at the end of its life cycle and the carrier glass was intact or if the module was broken. The main steps in both strategies were the destruction of the polymer film, the separation of the semiconductor from the glass substrate, and the enrichment and the recovery of the semiconductor material.

As for the first strategy, the unit was initially heat-treated and the polymer film between the semiconductor material and the rear glass plate decomposed at 470 °C, leading to the separation of the unit into two glass plates (the substrate on which the semiconductor material was deposited and on the rear plate). A vacuum blasting device was then used. The device created a vacuum on the surface where it was placed and the abrasive (iron powder, glass beads, aluminum oxide were tested) medium hit the surface at high speed. The thin film material and the abrasive got sucked into vacuum, preventing dust emissions and creating a closed circuit for the abrasive, for multiple uses. Clean glass could be completely recycled afterwards.

The second strategy consisted of the crushing/grounding of the PV unit, leading to the partial destruction of the polymer film and the production of glass particles with a diameter smaller than 20 mm with adhesive parts of the polymer film on their surface.

The crushed material was subjected to liquid mechanical treatment, in batch mixers, with different container sizes (25, 50, and 150 L) and stirring speeds. Process parameters such as stirring and mixer speed, water content, and processing time were optimized. During this process, the semiconductor layer of the CdTe and CIGS units was separated from the carrier glass using shear and friction forces on the surface of the fragments, without the addition of chemicals. Finally, the material was washed and sieved so that the diameter of the particles was smaller than 150 µm [87].

To further separate Cd, Te (CdTe PV) Cu, In, Ga, and Se (CIGS PV) from the residues of the aforementioned methods, the two strategies converged and the floatation separation method was followed, which is one of the most widely used methods of useful minerals recovery in the mining industry.

The grounded ore was mixed with water in specialized tanks to form a slurry. Hydrophobic materials were then separated from hydrophilic materials through the use of chemical additives. The tanks were stirred to form bubbles (due to the insertion of air), to which the hydrophobic metals were attached. As a result, the metals rose to the surface where they were collected from the supernatant. The precipitated ore was removed and may be further floated [88]. The flotation tests aimed at enriching the residue with Te and In from the CIGS thin film.

After testing several flotation reagents, Berger et al. [87] concluded that the highest enrichment factor was obtained using potassium amyl xanthate (KAX). The surface of the semiconductors became hydrophobic, allowing them to be collected from the supernatant foaming liquid, as mentioned. The product, derived from the liquid mechanical process, was then treated in an acidic medium to release Cd and Te from the solid matrix. The Cd and Te containing solution was directed to the Cd and Te recovery circuit, where they could be converted to 99.999% metal with additional purification [87]. Both metals could then be compounded to produce CdTe powder or could be used as commercial grade metals. The same procedure was followed for In. The flow chart is shown in Figure 5.

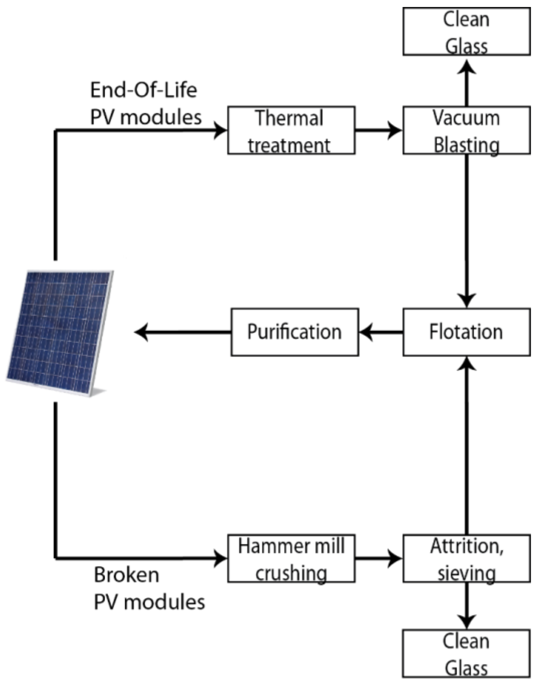

Figure 5. Two CdTe/CIGS thin-film PV module recycling strategies flow chart: intact, at the end of its life cycle and in case the glass plate is broken.

The total recovery of CdTe after the process was 78.7%, while 54% of the CIGS content in the thin film was recovered after shear and friction forces. The elements could be recovered and reach a purity of over 99.9% in order to be reused in CdTe and CIGS thin films, respectively. Due to the lack of heat treatment and therefore the use of non-renewable sources for energy production, the recycling strategy of broken PV systems was more environmentally friendly, with a global warming potential (GWP) of 0.413 kg CO_2-eq/kWh against 0.752 kg CO_2-eq/kWh of the strategy for intact EOL systems [87].

According to Appropedia [89], the same recycling method for CdTe, of Sasala et al. [84] and First Solar [85] as described before, can also be applied to the recycling of CIGS thin-film PV modules. Their calculations of the recycling viability were based on a 1 m^2, 31 kg solar module (Solyndra SL-001-200 200 W). Only the In, Ga, and glass were considered, but metals and plastic used in the frame and backing could also be recycled. Taking the power per unit area, the wasted mass, the disposal cost, and the recycle cost ($20.74 per module) into account, the total profit of recycling of the module could reach $37 [89].

According to the patent of Liu et al. [90], the CIGS thin film was inserted into a ball mill for grinding into particles smaller than 0.18 cm. The powder was then added to a solution of H_2SO_4, heated between 90 and 100 °C, and air was introduced into the heated solution for leaching. Sodium sulfite (Na_2SO_3) was then added to the solution, which was heated above 60 °C and stirred. Na_2SO_3 reacted with H2SO4 to produce sulfur dioxide (SO_2) gas, which then reacted with the selenic acid (H_2SeO_4), and pure Se could be obtained. Finally, NaOH was added to the solution to adjust the pH above 12, while a high-performance Cu extractant (AD-100N) was used to remove Cu in the form of $CuSO_4$, which was then electrolyzed to pure Cu. The supernatant, containing In hydroxide ($In(OH)_3$), was collected, dissolved in HCl, and Zn added to replace In, so it could be collected. In addition to the supernatant, the residue, containing sodium gallate solution ($C_7H_5NaO_5$), was then electrolyzed to pure Ga [90]. The flow chart is shown in Figure 6.

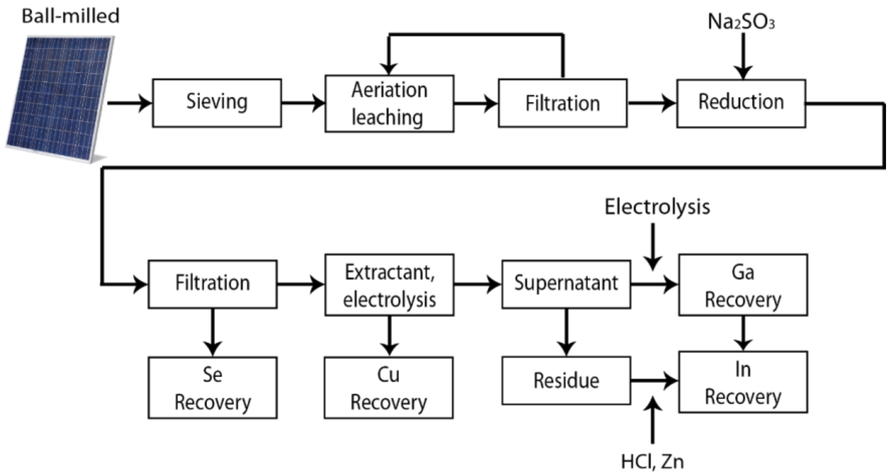

Figure 6. Recycling of CIGS thin-film PV module by acid leaching, extraction, and electrolysis flow chart.

3.3. Wind Turbines

Between 20 and 25% of REEs produced worldwide are used in the production of NdFeB magnets [91]. NdFeB permanent magnets have different life spans, depending on the applications: 2–3 years in consumer electronics, such as hard disc drives in computers and laptops, 20–30 years in wind turbines. The magnets in wind turbines lose their magnetic properties due to continuous heat generation [92,93].

According to Kaya et al. and Önal et al. [91,93], bulk NdFeB magnet pieces were used. No demagnetization took place. They were grinded three times to obtain powder of a size less than 200 µM. In order to prevent a spontaneous explosive reaction between the concentrated HNO_3 and the magnet powder and to obtain a homogenous mixing, the powder was first wetted and then HNO_3 was added. The solution was then calcined at 200 °C for 2 h. Afterwards, the calcined sample was water leached at a ratio of 1:15 (solid:liquid) for 1.5 h under stirring. In the study of Önal et al. [93], the sample was filtered and the HNO_3 could be recovered from the leachant residue via condensation and reused. The pure REE-loaded (>95%) leachate could be directly treated by any downstream process for REE separation (mainly consisting of Dy, Nd, Gd, Pr with minor impurities of B, Nb, Si). Due to the very low calcination temperature, the proposed method was not energy-intensive. The absence of intense particle size reduction processes and drying need, the potential recycle of the HNO_3, along with the applicability on a wide range of magnet feed type, made the process even more attractive. The flow chart is shown in Figure 7A. In their study, Kaya et al. [91] added another step after filtering of the leachant. Specifically, very fine aerosol droplets were obtained from the leach solution using an ultrasonic atomizer with a frequency 1.75 MHz in an ultrasonic field obtained by three ultrasonic transducers. The aerosol was carried with N_2 flow rate 1.0 L/min into a quartz tube between 700 and 1000 °C. XRD (X-Ray Diffraction) analysis of the calcined powders confirmed the formation of a REE-oxide mixture [91]. The recovery of metallic Nd and Pr could be achieved by using molten salt electrolysis [94]. The flow chart is shown in Figure 7B.

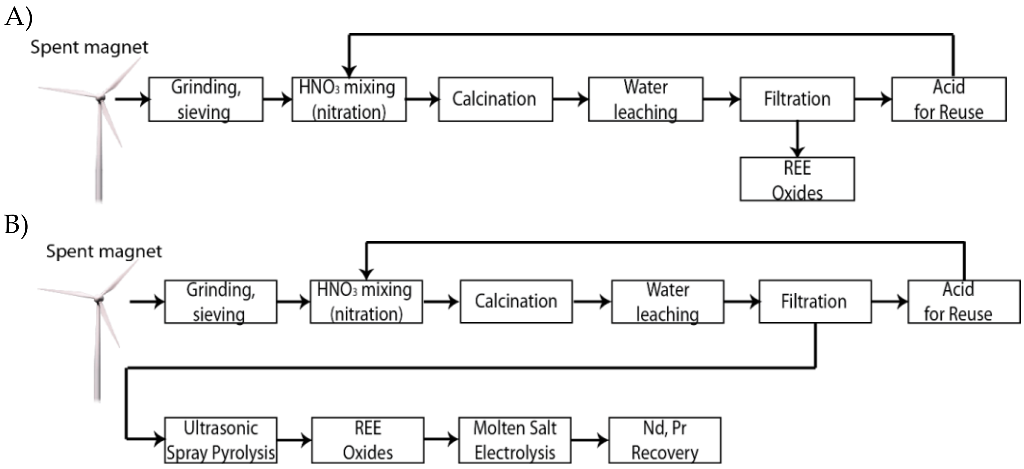

Figure 7. Recycling of NdFeB magnet from wind turbine by acid mixing, calcination, and water leaching (**A**), along with an added step of ultrasonic spray pyrolysis (**B**) flow chart.

In their study, Kumari et al. [95] used spent NdFeB magnets. They demagnetized them by heating the magnet blocks in a muffle furnace at 310 °C for 1 h. The demagnetized sample was then crushed and powdered to <149 µM size. The powder sample was then calcined in a muffle furnace at 850 °C for 6 h and leached with HCl at 95 °C for 5 h. After the completion of leaching, the leachate and leach residue were separated by vacuum filtration. The leach residue, containing impurities such as B, Al, Co, Fe, was dried in an oven at 90 °C for 8 h and filtered to isolate the impurity metals and recover the HCl for recycle and reuse. The REE present in the leachate reacted with the added stoichiometric quantity of oxalic acid ($C_2H_2O_4$) solution at pH 2 and oxalates of Nd, Pr, and Dy precipitated. The mixed REE oxalate precipitate was filtered, dried in an electrically heated oven, and calcined at 800 °C for 2 h to produce mixed REO. Finally, high-purity Nd, Pr, and Dy oxides from the leach solution could be separated with solvent extraction, followed by precipitation [95]. The flow chart is shown in Figure 8.

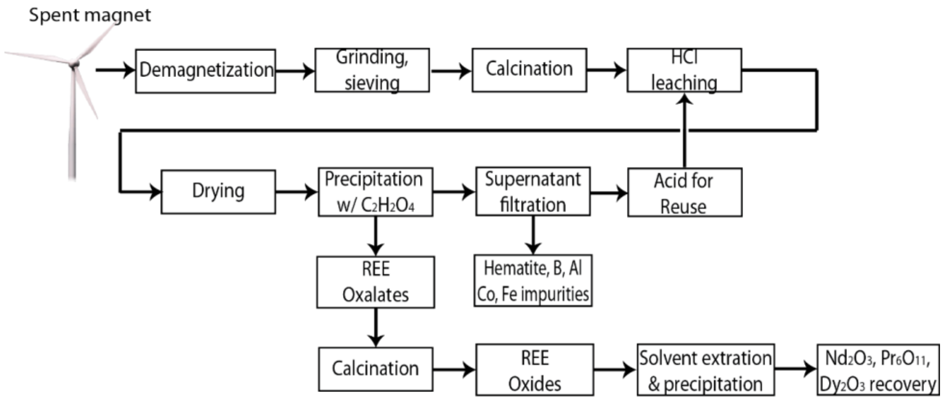

Figure 8. Recycling of NdFeB magnet from wind turbine by acid leaching, calcination and precipitation with oxalic acid flow chart.

4. Additive Manufacturing

Global competitiveness has shifted production chains and markets towards sustainability, for the long-term positive impact on the environment. The adoption of sustainability policies in the supply of raw materials is of paramount importance. The transition to carbon neutrality and further evolvement in the global ecosystem through disruptive products with unique features, while making it autonomous towards its raw materials supply, will have a positive impact on an environmental and economic level.

The recovery and reusability of the aforementioned elements and compounds thereof constitute the inception of a circular-economy, zero-waste, sustainable-by-design value chain paradigm. As such, its benefits can be many-fold. A significant prerequisite in developing commercial exploitation pathways on such promising concepts, is adopting safe-by-design principles, thus ensuring environmental and health safety in every stage of production (namely from the Proof of Concept to the final product).

Three-dimensional (3D) printing, also known as additive manufacturing (AM), is an advanced technology based on a computer-aided design (CAD) model, which features a free interface and layer-by-layer manufacturing procedures [96].

Owing to the convenience of preparing complex and customizable shapes, 3D printing exhibits unique technical versatility, making it integrate gradually into various applications, such as medicine, electronics, sensors, and automation. Three-dimensional printing has received extensive attention due to its unique multidimensional functionality and customizability and has been recognized as one of the most revolutionary manufacturing technologies [97]. It is a rapidly developing technology with short production time, little material waste, and an ability to print complex, customized shapes and dimensions [98]. Additive manufacturing is especially well suited for the fabrication of graded architecture. The technology has demonstrated the ability to fabricate architectures that are impossible to fabricate through conventional processing techniques [99].

Material extrusion is an additive manufacturing process in which material is selectively dispensed through a nozzle. Fused deposition modeling (FDM), fused filament fabrication (FFF), 3D dispensing, and 3D bio-plotting fall into this category [96].

FDM has the advantages of low cost, simplicity, and efficiency. A wide variety of functional integrations can be realized through the diverse combination of multiple extrusion nozzles and raw polymer materials. However, the 3D printable polymer materials for FDM processing are supposed to possess most of the properties of high mechanical strength, low shrinkage rate, suitable melting temperature, non-toxicity, simultaneously, which dramatically restricts the availability of raw materials. At present, acrylonitrile butadiene styrene (ABS) and polylactic acid (PLA) are demonstrated to be the two main commercially available polymers for FDM, which dominate more than 95% of the market. Drawbacks, such as high cost of filament-type consumables, easy clogging of nozzles, difficulty in the printing of elastic materials, and polymer-based micro-/nanocomposite materials, have to be addressed and solutions have to be implemented [97].

Regarding the case of advanced materials, introducing metals REE recovered from WEEE as fillers or additives to materials employed by AM applications can have a tremendous potential for both the performance and the commercialization of the final products. For instance, metals or metal oxides (M/MO) and REE used in filaments for FDM can add unique characteristics to the printed product, such as antibacterial properties, photoluminescence, thermochromism, enhanced mechanical properties, magnetic properties, thermal and electrical conductivity, and others [97].

While customized medical devices and daily life items can be made by 3D printing of thermoplastics, microbial contamination has been a serious obstacle during their usage. A very clever approach to overcome this challenge is to incorporate antimicrobial M/MO nanoparticles within the thermoplastics during or prior to 3D printing. Many M/MO nanoparticles can prevent contamination from a wide range of microorganisms, including antibiotic-resistant bacteria via various antimicrobial mechanisms. Additionally, they can be easily printed with a thermoplastic matrix without losing their integrity and functionality [100]. Most of the essential metals such as Cu, Zn, Mg, and their oxides have strong biocidal effects, while other non-essential M/MO including Titanium (Ti), Ag, Au, and Ce oxide are popular antibacterial agents [101]. Incorporation of M/MO nanoparticles into polymers could improve overall antibacterial efficacy of the nanocomposite, due to the synergetic polymer and nanoparticle effects [102]. Ag, Au, Zn, and Mg oxides have been reported to also present redox properties, prompting reactive oxygen species (ROS) formation, which can alleviate oxidative stress [103].

With regards to Cu, it can be used to give photoluminescence and thermochromism properties, besides antibacterial [104]. It can also increase wettability and improve the flexural strength of polymers (specifically PLA). Finally, it can be used in thermoplastic filaments as conductive filler for the fabrication of resistors in electronic components [105]. In comparison to carbon-black and graphene, Cu offers better mechanical properties [106]. Cu is present in the wiring of all of the aforementioned sources (PCB, PV, WT).

Glass extracted from EOL PV, as well as glass fibers found in PCB, can be used as additives in order to reinforce the polymer and increase the mechanical properties of the final product, namely textural and flexural strength [107].

Electroless plating is a metallization technique that does not require electricity; rather, an immersion in a reducing agent which leads to metal deposition [108]. In their study, Zhan et al. [109] used electroless plating that achieves area-selective metal coating on 3D-printed plastic structures without conventional pretreatments, such as surface etching, which often involves hazardous chemicals (chromic acid) and roughening. They fabricated, using FFF 3D-printing, ABS filaments containing Pd ions (catalyst precursor for Ni electroless plating) using ABS as the polymeric matrix and $PdCl_2$ in acetone solution. The structure was then immersed in a Ni electroless plating bath and Ni was deposited in the selected areas and strongly adhered to the ABS. Many metals (Ni, Co, Cu, Au, Ag, Pt, etc.) and their alloys could be deposited by electroless plating, leading to the wide application of their proposed technology [109].

The FENIX project [110] studies the recovery of precious metals and critical raw materials from WEEE, using similar extraction methods for Au, Ag, Cu, as described by Behnamfard et al. [78], and tested their reusability in novel materials. The commercially exploitable products of such processes according to the project can be: M/MO powder as a product to be used for the formulation of filaments or for the direct use in injection molding and novel composite filament (M/MO and a polymeric matrix) as a product employed by FFF AM applications, giving novel properties to 3D-printed samples thereof or for the production of parts that can be sintered into customizable-shaped metal components [110].

Regarding potential exposure to Cd compounds in plastics, the European Chemicals Agency [111] stated that once encapsulated in the polymer, Cd and its compounds are firmly bound into the matrix and the use of strong acid is required to extract it. Cd is used in pigments as stabilizer, which can find application to colored polymers [111]. Moreover, 3D-printed objects can provide protection against gamma and X-ray radiation if the filament used is infused with high atomic number materials. Banoqitah et al. [112] developed a simulation analysis for thermal neutron applications of ABS 3D printing filament doped with Ga, B, Au, and Cd nanoparticles, which are excellent neutron absorbers. They concluded that ABS infused with Cd can be used for radiation detection and photon shielding purposes in a mixed neutron-gamma field. Finally, Cd can also be used to electroplate steel and protect it from corrosion [113].

Te-containing polymers have become increasingly attractive, due to their unique properties as biomaterials and optoelectronic materials. The relatively large atomic size, low electronegativity, and sensitivity to oxygen of Te add unique properties, such as redox sensitivity. The introduction of Te can endow materials with different responsiveness, which makes it possible for them to be employed as biomaterials for controlled drug delivery. Further research has to take place in order to optimize the optoelectrical properties of polymers enriched with tellurophenes [114]. Te can be used in 3D printing of thermoelectric conversion devices. Thermoelectric (TE) materials exhibit the capability to directly convert thermal energy into electrical energy, based on the temperature difference between the two ends of TE materials, e.g., PV. The majority of applied TE materials are based on metal chalcogenides, such as Te, due to the largest ZT (the dimensionless TE figure of merit ZT, has been utilized to characterize the efficiency of a TE material) [97]. So far, FDM 3D printing technologies have been investigated for the preparation of TE devices. According to Wang et al. [98], PLA can be used as a matrix in composite materials containing Te compounds (specifically $Bi_{0.5}Sb_{1.5}Te_3$), carbon nanotubes (conductive additive), and plasticizers [98]. Finally, ABS polymer matrix, according to Oztan et al. [115], was enhanced with bulk Bi_2Te_3 samples as the TE agent filler via FFF and the thermoelectric properties, such as Seebeck coefficient and electrical conductivity, were enhanced due to the lowered porosity of the printed specimens and the improved connectivity of the thermoelectric powders sintered at higher temperatures (above 500 °C). Cd and Te are present in the CdTe thin-film PV modules.

Indium tin oxide (ITO) is an optoelectric material and can be used in polymer-based electronics. It is highly transparent and has a relatively high electrical conductivity. Sn-doped indium oxide (ITO) can be deposited in organosilicon layers or directly on polyethylene terephthalate (PET), according to Mikoshiba and Sato [116]. The mechanical properties of the ITO layer on PET surface such as abrasion resistance, peel strength, and durability are improved. Indium alloys exhibit low melting points and high ductility [116]. UV (ultraviolet) polymer curing is a process that uses UV light to alter the properties of a polymer, or photopolymer. When a photopolymer is exposed to the UV radiation, it will typically harden and seal, creating a strengthened surface or an unbreakable bond [117]. In their study, Mendes-Felipe et al. [118] produced polyurethane acrylate (PUA) composites with ITO, by UV photopolymerization with filler contents up to 25 wt%, in order to tune morphology, optical, thermal, mechanical, and dielectric properties. The polymerization time and conversion were dependent on the ITO amount. The ITO fillers were well dispersed in the polymer matrix and no large agglomerates were observed, leading to an improvement of the mechanical properties (robust flexibility). They concluded that it is possible to develop high-dielectric constant UV curable polymer composites based on ITO/PUA [118].

Their nonlinear optical properties, electro-luminescence, and electrical conductivities can find application in organic light-emitting diodes (OLEDs), organic solar cells, organic field effect transistors (OFETs), nonlinear optical (NLO) devices, and polymer sensors. General conjugated polymers consist of benzene rings or carbon–carbon multiple bonds in their main chains. By replacing the heavier atom, other properties are often obtained [119]. However, there are only a few reports on the electronic properties of organo-gallium compounds because of their instability to air and moisture. In their study, Matsumoto et al. [119] synthesized the polymers containing Ga atoms by organometal coupling reactions. The synthesized polymers showed good solubility in common organic solvents and enough stability for measuring a series of properties under ambient conditions. The electronic interaction through Ga was supported by UV-Visible spectrometry data and theoretical calculations.

In recent years, Se-containing polymers have been promising biomaterials [120]. Taking advantage of the unique properties of Se (stimulus-responsive, anti-cancer activity antibacterial properties, and ultraviolet blocking action) along with the features of Se-containing polymers (e.g., high sensitivity, programmability, reversibility, and dynamics), they can be used to build smart systems of controlled drug release or synthetic enzyme mimics [120]. According to Deng et al. [121] amorphous Se filaments, fabricated by an approach in which a thin film evolves into an ordered array of filaments in fiber, exhibited a two-orders-of-magnitude change in conductivity between dark and illuminated states, making them suitable for integration in large-area electronics. Cu, In, Ga, and Se are mostly present in the CIGS thin-film PV modules.

As mentioned, AM technology is well suited for the fabrication of architectures that are impossible to fabricate through conventional processing techniques [99]. Additives and fillers such as critical metals, REE, and glass can give novel properties to composites, whilst using sustainably recovered raw materials. Recycling of critical metals such as Cu is growing exponentially. More specifically, a total of around 8.7 million tons of Cu per year come from the recycling of "old" scrap (EOL products) and "new" scrap (generated during production and downstream manufacturing processes) [122]. Only 1% of the REE are recycled from end-products, with the rest deported to waste and being removed from the materials cycle [5]. Only in the Nordic countries, the quantity of critical and rare earth metals, such as Au, Ag, In, Ga, Nd, Pd in WEEE, accounted for a total of 148 tons (in 2015) [123,124]. As for the nonmetal materials, such as glass, latest value chain data show that the average collection for recycling rate for glass packaging grew to the record rate of 78% in 2019 in the EU [125]. The US Glass Packaging Institute (GPI) currently estimates that the container glass and flat glass industry in the US uses a total of 3.35 million tons of recycled glass [126]. The amount of the aforementioned critical raw materials that can be recovered and recycled from WEEE on a global scale and reused in sustainable new technologies, such as AM, can grow exponentially.

5. Conclusions and Future References

WEEE is a waste stream of different materials, including multiple hazardous constituents that can be released in the environment if not treated properly. Moreover, as the demand for raw material rises, more recovery and recycle operations, as well as illegal shipments of WEEE, take place in developed and developing countries, which do not always follow the strict regulations and requirements, increasing health and environmental risks [14]. The WEEE Directive has been adopted to reduce these risks by establishing requirements to ensure the safe collection and environmentally sound treatment of WEEE. The Directive of 2020 dictated that the minimum collection target was 4 kg/inhabitant/year of WEEE for countries-members (equal to 33% of WEEE arising per year). Inspection during recovery, recycle paths, and shipment of WEEE is mandatory, to ensure safe-by-design procedures. Finally, the WEEE collection points are increased, followed by the collection targets accordingly [14]. However, there are pitfalls, such as extra costs which are not matched by the increased recovery of valuable materials. For some types of WEEE, particularly those containing hazardous substances, these costs are very significant compared to the value of the materials themselves.

Furthermore, informative and educational campaigns can raise awareness of the advantages of WEEE reusability and lead to improved measures and tailor-made policies, for every country [127]. The logistics and closed-loop supply chains can be analyzed and optimized accordingly, to ensure a balanced, monitored stream of WEEE, based on a circular-economy, zero-waste, sustainable-by-design paradigm.

First, this review presented the main features and characteristics of the critical raw materials found in the main components of WEEE. Then, having narrowed them down to those that are widely used in sustainable energy sources and every-day appliances and which can be relatively easily isolated, removed, and recycled, this review can serve as a collection of thorough procedures, applied and theoretical, that can lead the way to future breakthroughs. It must be taken into account that general procedures and guidelines of complicated flow charts should allow for further experimentation for verification and validation. Each particular critical raw material requires thorough trials with its corresponding extraction and recycle method, in order for the exact steps of the procedure and the novelty of the composite material to be fully understood.

The plethora of available recycle techniques is one of the main targets of this review. However, not all of the technologies discussed will be of significant interest, as some will be economically and environmentally feasible. The progress made over the last years towards the sustainable-by-design recycling procedures, which can lead to novel materials with tremendous potential in industrial employment, has been the cornerstone of further development and optimization of composite materials.

As the need for sustainability during the recycle and reuse of raw materials is more significant than ever before, the state-of-the-art processes need to fulfill some major requirements. The flow-chart of the process needs to be composed of environmentally friendly techniques (such as mechanical removal of parts from WEEE) that are less energy- (and subsequently cost-) intensive, and ensure the use of non-hazardous chemicals during every stage. Furthermore, heat treatment of WEEE must be in compliance with safe-by-design protocols of capturing dangerous and toxic emissions. Most of the widely used REEs' extraction, recovery, and separation from minerals and ores are extremely costly, resulting in the increased price of the final product. The proposed recycle paths and the sustainability of the final novel composites are aimed at tackling this issue, while maintaining the environmental impact at a minimum.

The shift of technological progress towards sustainability, through the novel materials employed by AM applications, based on WEEE recycling, can be beneficial both environmentally, by decongesting landfills of potentially hazardous materials and toxins, and financially, as this can be the disengagement strategy from China and other major pioneers in extraction and monopoly of critical raw materials.

The originality of the review lies on the collection of relatively recent methods and patents on the recycle path of the mentioned CRM and their introduction, as additives to filaments used for the synthesis of composite materials with the ability to fabricate architectures that are impossible to fabricate through conventional processing techniques. By adding unique characteristics, tremendous potential for the performance and the commercialization of the final products can be achieved.

Further research will focus on testing some of the most sustainable and environmentally friendly, recent processes, followed by technoeconomic reports on the feasibility of each respective study and its applicability at a larger scale. Moreover, after the appropriate protocol of the critical raw material recovery is drafted, the existing expertise in the incorporation of critical raw material into composite materials and characterization of the final, novel 3D-printed composites for the modern industry, as well as the infrastructure with the latest technology equipment, will make the Physical Metallurgy Laboratory of the Department of Mechanical Engineering of AUTh an excellent candidate for the evaluation of the research and commercial exploitation of these new technologies, with further results to be published in scientific journals and conferences.

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Abbreviations

REE	Rare Earth Elements
WEEE	Waste Electric and Electronic Equipment
RM	Raw Materials
AM	Additive Manufacturing
FR	Flame Retardants
REO	Rare-Earth Oxides
PCB	Printed Circuit Boards
CP	Cell Phones
PV	Photovoltaic
WT	Wind Turbines
EOL	End of Lice Cycle
PWB	Printed Wired Boards
CdTe	Cadmium-Tellurium
CIGS	Copper-Indium-Gallium-Selenium
NdFeB	Neodymium-Iron-Boron
MW	MegaWatt
WPCB	Waste Printed Circuit Boards
GWP	Global Warming Potential
XRD	X-Ray Diffraction
CAD	Computer-Aided Design
3D	Three Dimensional
FDM	Fused deposition modeling
FFF	Fused Filament Fabrication
ABS	Acrylonitrile Butadiene Styrene
PLA	Polylactic Acid
M/MO	Metal/Metal Oxide
TE	Thermoelectric
ITO	Indium Tin Oxide
PET	Polyethylene Terephthalate
UV	Ultra Violet
PUA	Polyurethane Acrylate
OLED	Organic Light-Emitting Diodes
OFET	Organic Field Effect Transistors
NLO	Nonlinear Optical

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