Review

Status of Recovery of Strategic Metals from Spent Secondary Products

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Abstract: The need to drive towards sustainable metal resource recovery from end-of-cycle products cannot be overstated. This review attempts to investigate progress in the development of recycling strategies for the recovery of strategic metals, such as precious metals and base metals, from catalytic converters, e-waste, and batteries. Several methods for the recovery of metal resources have been explored for these waste streams, such as pyrometallurgy, hydrometallurgy, and biohydrometallurgy. The results are discussed, and the efficiency of the processes and the chemistry involved are detailed. The conversion of metal waste to high-value nanomaterials is also presented. Process flow diagrams are also presented, where possible, to represent simplified process steps. Despite concerns about environmental effects from processing the metal waste streams, the gains for driving towards a circular economy of these waste streams are enormous. Therefore, the development of greener processes is recommended. In addition, countries need to manage their metal waste streams appropriately and ensure that this becomes part of the formal economic activity and, therefore, becomes regulated.

Keywords: catalytic converters; electronic waste; batteries; precious metals; recovery; valorisation

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1. Introduction

The global demand for platinum group metals (PGMs), such as platinum (Pt), rhodium (Rh), and palladium (Pd), continues to increase. Against dwindling natural deposits, the demand for PGMs in the fine chemicals and petrochemicals industries, as fuel cells, for electrical and electronic products, for medical and dentistry applications, for jewellery, and in automobile industries requires the improvement of current recovery methods [1]. An alternative is to recover the important metals from end-of-cycle products, and in this way, the urban mining of metal waste can be advanced to contribute to the circular economy. Urban mining is becoming more attractive due to the high yields compared with extraction of strategic metals from primary ores. The importance of processing secondary sources of precious metals is remarkable given the decline of the supply of precious metals, especially PGMs, from primary mineral ores [2,3]. The primary supply can be complemented by the secondary supply that has been recovered from spent (end-of-cycle) products [4]. Precious metals are rare natural resources of high value and are concentrated in e-waste and other metal waste streams. Their unique properties, such as good thermal and electrical stability, resistance to corrosion, and low chemical reactivity, have resulted in their blossoming applications [5]. Recycling provides a highly efficient way of reintroducing valuable materials back into the economy, with huge benefits of saving energy and reducing environmental impacts, as opposed to the primary supply. The reprocessing of precious metals in successive life cycles reduces the environmental burden [6]. The PGMs have found usage in the automotive industry and are used in

catalytic converters to reduce harmful emissions [7]. It is mandatory for conventional vehicles to be fitted with catalytic converters to comply with air quality standards due to harmful emissions from combustion engines [8]. The combined catalytic properties of Pt, Pd, and Rh are very important in automotive emission control catalysts [9]. Toxic gases, such as nitrogen oxides (NO_x), carbon monoxide (CO), and unburnt hydrocarbons (HC), are converted to environmentally friendly gases in the catalytic converter to achieve low levels of harmful emissions [10]. Pd and Pt are used for oxidation reactions to convert CO into CO₂ and HC into CO₂ and H₂O, while Rh is used for reduction reactions to convert NO_x to N₂ [8]. There has been an increasing gross demand for the three metals, driven by the catalytic converter manufacturing sector from 2008 to 2018 [11,12]. Stricter emission acceptances announced in the Euro VI standards of 2014 are set to keep this demand trend on a growth trajectory. This provides motivation globally to consider increasing the recovery rates of valuable metals from waste by driving a “closed-loop” recycling system. Automotive catalytic converters may contain up to 2000 g per ton (g·t^{−1}) of PGMs in the ceramic catalyst brick [13], making them economically viable to be driven by the circular economy principle. Currently, automotive applications have reached a global average of 50–60% end-of-life recycling rates for PGMs [14], but there is room for improvement.

The treatment of e-waste prevents harmful effects on human health and the environment [15]. Mass-produced consumer components, such as computer motherboards, contain around 200–250 g·t^{−1} of gold and around 80 g·t^{−1} palladium, and mobile phone handsets contain up to 350 g·t^{−1} gold and 130 g·t^{−1} palladium [14]. This is significantly higher than the gold or PGM content in primary ores (on average < 10 g·t^{−1}). In addition to gold and palladium, there are significantly high quantities of silver, iron, and copper in e-waste. Recycling e-waste reduces the environmental burden of toxic agents from e-waste. Consumed batteries also contain hazardous materials, such as cadmium, lead, nickel, and zinc metals, at high-level concentrations [16,17]. Lead is a highly toxic metal with no known useful function in the human body, and of particular concern is the effect of relatively low-level exposure on cognitive and behavioural development in children, including the lowering of IQ [18]. Therefore, the high intrinsic metal values make recycling attractive from an economic point of view and due to the much higher concentration of metals compared to the mining of ores. The industrial applications of PGMs, such as catalysts used in the production of fine chemicals or in petrochemical processing and equipment used in the glass industry, are the current benchmark for platinum group metal (PGMs) recycling. Recycling rates of 80–90% have been reported for these industries, although catalyst lifetimes can reach several years before they enter the recycling chain [14]. Recycling in this case is solely market driven and is an integral part of the product life cycle, resulting in a limited need for the primary supply of PGMs in these industries. However, recycling rates are currently only 5–10% for precious metals in electronic applications. The largest use of PGMs in electronics is the palladium used in circuit boards, but gold, silver, and copper are also found in significant quantities in e-waste from end-of-cycle products, such as PCs, TVs, laptops, and mobile phones [19]. E-waste is made up of many different organic and inorganic compounds, which are determined by the age, manufacturer, and type of equipment, and it contains approximately 62% metal [20].

Technologies of precious metal recycling have progressed over the years, and challenges and opportunities have been acknowledged. Several reviews in the literature cover this topic from a technological approach and through national, continental, and global perspectives [14,21–23]. The literature search on this topic via Scopus also shows a steady increase in publications over the past decade (Figure 1) with China leading in this focus area of research (Figure 2). From the data obtained, the commonly used methods for purification are hydrometallurgy, pyrometallurgy, and biohydrometallurgy. Some studies are dedicated to the development of new chemistry towards the processing of these new feeds [24,25]. This review provides an overview of technologies, based on published research, for the recovery of platinum group metals (PGMs) from spent automotive catalytic

converters, e-waste, and other sources, such as batteries. It also covers the conversion of metal solutions of secondary products into important nanomaterials.

Figure 1. Research trends in the processing of secondary resources by the number of publications per year, 2010–2020. (Source: Scopus. Keywords: recovery of precious metals from spent secondary products and e-waste. The keywords were used to select the relevant articles and the search was limited to 2010 to 2020.).

Figure 2. Breakdown by country of the research trends in the processing of secondary resources from publications in Figure 1. (Source: Scopus. The same information obtained for Figure 1 was used, and the country breakdown was limited to countries with 20 or more published articles on the recovery of precious metals from spent secondary products and e-waste.).

2. Recovery of PGMs from Catalytic Converters

2.1. Composition of a Catalytic Converter

Catalytic converters consist of the catalytic material that contains a mixture of platinum group metals (PGMs), such as platinum (Pt), palladium (Pd), and rhodium (Rh). Figure 3 shows petrol and diesel catalytic converters. The composition of the catalytic carrier significantly varies depending on the engine capacity and the type of fuel used. The platinum metal content in car catalysts varies considerably depending on the manufacturer and performance of the catalyst [26,27]. The proportion of Pt, Pd, and Rh is dependent on the level of emission control, metals prices, and the type of car [28]. The amount of Pt, Pd, and Rh in a carrier differs: for a small car it can range from 1 to 5 g, for a truck from 12 to 15 g [29], and for a sports car from 6 to 30 g due to the large quantity of exhaust gases [30]. The Pt concentration is larger than Pd and Rh: the range for Pt is 300–1000 ppm, for Pd it is 200–800 ppm, and for Rh it is 50–100 ppm [30] and other authors suggest that the total content of PGMs is lower than 1000 ppm [31,32]. Yakoumis et al. reported that the data of a recycled quantity of more than 40,000 spent ceramic catalytic converters confirmed a PGM average concentration of 2596 ppm for catalytic converters [33]. The concentration of the PGMs' loading per metal in a spent ceramic catalytic converter was averaged as follows: Pt (720 ppm), Pd (1582 ppm), and Rh (294 ppm). Yakoumis et al. also reported on spent metallic catalytic converters using more than 4000 spent units, and an average concentration of about 7872 ppm was obtained, which was higher than what was obtained for spent ceramic catalytic converters [33].

Figure 3. (A) Spent diesel catalytic converter before dismantling. (B) Spent catalytic converter after dismantling. (C) Zoomed-in image of the ceramic catalytic converter. (D) Spent catalytic converters where (D) represents diesel and P represents petrol type.

The catalytic converter carrier (monolith) is mainly made up of a ceramic or metallic substrate. Metallic catalytic converters are an alternative to standard ceramic catalytic converters. The structural differences between metallic and ceramic catalytic converters carriers are detailed below and shown in Figure 4, and the typical metal carrier holes are triangular, while the structure of the ceramic carrier holes is square. The ceramic carriers are well studied, and there are many methods used for the recovery of PGMs from ceramic carriers compared to metallic carriers. In addition, the catalytic converters have some advantages and disadvantages as outlined in Table 1.

Figure 4. Structural differences between the metallic and ceramic monolithic substrate [33].

Table 1. Advantages and disadvantages of ceramic and metal carriers [26].

Carrier	Advantages	Disadvantages
Ceramic	Good surface development and low thermal expansion. A good distribution of catalytic active phase.	Lower thermal and mechanical strength of carriers.
Metal	Resistant to thermal shock and mechanical damage; thinner walls; heats up fast; able to create a variety of shapes to increase exhaust flow easily; heat.	The process of obtaining catalytic active coating is complicated.

2.1.1. Metallic Monolith

The metallic carrier is made up of FeCrAl steel with a porous surface deposited with Pt, Pd, and Rh. About 10% of automotive catalytic converters on the market contain a metallic carrier [33] that is made by employing corrugated FeCrAl foils, rolled into a spiralling cylinder shape [34]. The ferritic stainless steel is the typical alloy used for metallic catalytic converters, and it contains about 17–22% chromium (Cr) and 4–6% aluminium (Al). The metallic FeCrAl alloy consists of rare earth elements (REE) of about 0.01%. The commonly utilized REEs are lanthanum (La), cerium (Ce), praseodymium (Pr), neodymium (Nd), samarium (Sm), gadolinium (Gd), dysprosium (Dy), yttrium (Y), and erbium (Er) [35]. The metallic automotive catalytic converter carriers are also known as metal substrate converters (MSC) and were originally designed for sport and racing cars. The metallic monolith also comes in different components, such as Fe-Cr-Al-(Y), Fe, Fe-Al, or Fe-Cr-Al-Co. The addition of rare earth elements to FeCrAl alloys assists in improving the oxidation resistance of alumina at high temperatures by reducing oxidation rates and enhancing the scale of adhesion [29].

2.1.2. Ceramic Monolith

The ceramic carrier is the most used in catalytic converters (about 90%) [33]. The ceramic monolith is made of Al_2O_3 , and this type of monolith is known as honeycomb shape with numerous channels. The honeycomb cordierite skeleton-type ($2\text{MgO} \cdot 2\text{Al}_2\text{O}_3 \cdot 5\text{SiO}_2$) is coated with Pt, Pd, and Rh followed by a wash coat cover with high porosity (about 50–200 μm) that is made up of about 90% $\gamma\text{-Al}_2\text{O}_3$ and a mixture of oxides of cerium (Ce), zirconium (Zr), lanthanum (La), nickel (Ni), iron (Fe), and alkaline earths that assist in improving the stability of the catalytic converter [28,36]. The ceramic monolith comes in various compositions, such as Al_2O_3 , $2\text{MgO} \cdot 2\text{Al}_2\text{O}_3 \cdot 5\text{SiO}_2$, $3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$, $\text{MgO} \cdot \text{Al}_2\text{O}_3$, $2\text{MgO} \cdot 2\text{Al}_2\text{O}_3 \cdot 5\text{SiO}_2 \cdot 3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$, $3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot \text{Al}_2\text{O}_3 \cdot \text{TiO}_2$, SiO_2 , SiC,

Si_2N_4 , B_4C , ZrO_2 , and $\text{ZrO}_2\text{-MgO}\cdot\text{Al}_2\text{O}_3$. Different oxides (CeO_2 , NiO , or Li_2O) can be added to Al_2O_3 to increase its properties, such as its resistance to ageing at high temperatures. Table 2 represents the chemical analysis of a typical spent ceramic honeycomb catalytic converter reported by Yakumis et al. [33].

Table 2. Chemical analysis of a spent ceramic honeycomb catalytic converter [33].

Oxide/Metal	(ppm)
Ceramic Monolith—Cordierite	
MgO	85,100
Al_2O_3	371,500
SiO_2	353,300
Poisoning (Lubricants, Fuels)	
Na_2O	30,700
P_2O_5	18,100
CaO	11,600
Cr_2O_3	8000
Fe_2O_3	24,400
ZnO	3300
BaO	7700
S	4400
C	4300
Catalyst Support (Wash Coat)	
TiO_2	25,900
ZrO_2	45,500
La_2O_3	2900
CeO_2	35,100
Catalyst (Noble Metals)	
Pd	1364
Pt	754
Rh	257
PGMs Total	2375

After years of exposure to the harsh and severe conditions of a catalytic reaction process, spent catalysts no longer have homogeneous catalyst distribution. By the end of their life, either they have accumulated many different contaminants of various densities, organic compounds, oils, and water, or they are broken. Thus, once they are recycled, a suitable sampling procedure should be established to obtain a homogeneous representative sample for qualitative and quantitative analysis. This is a necessity due to other mixtures introduced from fuel and oil additives during operation.

2.2. Methods of Recovery of Precious Metals from Catalytic Converters

The process of the recovery of PGMs from spent catalytic converters can be carried out by pyrometallurgical [37] and/or hydrometallurgical processes [28]. Dismantling and preprocessing proceed by leaching and further purification using precipitation, solvent extraction, ion exchange, etc., in the hydrometallurgical route [38]. The methods involve energy-intensive processes and/or the generation of large amounts of effluents to be treated. However, the hydrometallurgical recovery process has been shown to be the most economical option, with low energy consumption and an average PGM recovery efficiency of about 90% [29]. Aberasturi et al. also demonstrated the recovery of platinum group metals from car catalytic converters using hydrometallurgical processing routes [39].

Hence, the recovery of PGMs from the spent catalytic converter can be broken into the following basic steps [40]: (1) dismantling, (2) preprocessing, (3) leaching process, (4) metal separation from concentrated leachate, (5) purification process, and then (6) pure PGMs are obtained.

2.2.1. Dismantling Process

The dismantling of spent catalytic converters was reported by Yakoumis et al. [33]. The precious-metal-bearing carrier material and the non-precious-metal-bearing steel casing are separated by decanning ceramic converters or shredding metallic converters. Firstly, the inlet and outlet exhaust extension tubes are cut, and the catalysts proceed to the sorting step. After the sorting of catalytic converters, the converters are decanned, and in this step, the metallic canister and the wool (fibre mat) are removed from the ceramic catalyst using a mechanical process. Once the steel casing is removed, the ceramic monoliths or the metallic catalytic converters are taken to a precrusher in order to downsize their dimensions and then to a mill in order to extract a fine standardized powder. The PGM-containing particles are downsized and ground in the analytical mill to about 160 μm . Several leaching processes have been developed based on the selective dissolution of the honeycomb unit consisting of PGMs after crushing and milling [40]. Coa et al. reported the preparation of spent catalytic converter powder, where the honeycomb unity is crushed, followed by grinding until a 0.3-mm size is reached using a cutting mill (IKA MF10) [41]. Spent catalytic converters are then processed in two ways: (1) pretreated and/or (2) the direct leaching of PGMs [32,41–43].

2.2.2. Preprocessing

The preprocessing of catalytic converter powder has been investigated under different conditions to remove organic substances and carbon deposition on the surface, followed by the hydrometallurgical treatment. PGMs are packed with special inert materials that lead to the low leaching efficiency of PGMs. Therefore, preprocessing steps, such as fine grinding, roasting, reduction, pressure leaching, etc., have been reported as necessary steps before leaching the PGMs. Many investigations have reported various pretreatment methods (oxidization roasting, reduction roasting, preleaching, etc.) to destroy organic substances on the surfaces of spent catalysts or to change the support forms [31].

In another study by Kim et al. [43], it was reported that the deposits of organics and carbon may absorb precious metals during leaching experiments and that calcination is necessary before leaching the spent catalysts. However, after calcination, $\alpha\text{-Al}_2\text{O}_3$ is formed which decreases the dissolution of alumina. The calcination temperature of 800 $^\circ\text{C}$ influenced the leaching behaviour of Pt from the spent catalysts with a leaching efficiency of 99.9%. The results demonstrated the significance of calcination temperature on Pt leaching [44]. It has also been reported that spent catalytic converters can be thermally pre-treated with hydrogen, oxygen, nitrogen, or air depending on the undesired organic constituents to be removed to produce ash-containing PGMs [31]. Another study reported spent automotive catalysts that were heated under a hydrogen atmosphere at 250 $^\circ\text{C}$, which improved metal recovery during leaching due to the elimination of the hydrocarbons and charcoal present on the catalyst surface and the reduction of oxidized PGMs [38]. Pretreatment of the catalytic powder also assists in stabilizing the PGMs in their metallic form, making them easy to dissolve with leaching solutions [45].

2.2.3. Leaching Process

The extraction of PGMs from catalytic converters is typically performed in a two-step process once dismantling, crushing, and milling have been done. The first step is leaching the spent catalytic converter into solution to dissolve the PGMs. This step is accompanied by leaching parameters, such as time, temperature, and solid–liquid (S/L) mass ratio [27]. The next stages are concentrating the solution, extracting metals, and recovering pure PGMs from the solution [28]. It has been reported that pregnant solutions (leachate solutions) can be treated using different treatment methods, such as thermal hydrolysis, electrowinning, or zinc cementation [46]. The metals contained in the catalytic converter's carrier are transferred to an aqueous solution. PGMs are recovered by dissolving in acids, such as hydrochloric acid, nitric acid, and sulfuric acid, in the presence of oxygen, iodine, bromine, chlorine, hydrogen peroxide, etc. The chloride medium is commonly utilized where HCl

serves as a chloride source for the leaching of PGMs. It provides a high oxidation potential and high concentration of the chloride ion, and the efficiency of chloride leaching has been evaluated and shown to be the main factor affecting the leaching process. HCl solutions and mixtures of HCl and chloride salts, such as MgCl_2 and NH_4Cl , have been investigated [44]. Oxidants have been employed to promote the dissolution of PGMs' metallic form. Oxidants, such as H_2O_2 , HNO_3 , NaOCl , NaClO_3 , FeCl_3 , and Cu(II) , among others, have been proposed [28,31,44,47,48]. Ding et al. [47] reported the effective leaching of palladium from spent catalysts through oxidation with Fe(III) . The chloride medium in the leaching system provides a high oxidation potential and a high concentration. As a result, PGMs are mostly in the form of chlorido complexes ($[\text{PtCl}_6]^{2-}$, $[\text{PdCl}_4]^{2-}$, and $[\text{RhCl}_6]^{3-}$) [47,48]. The leaching process or dissolution of PGMs in solution from spent catalytic converters has shown a leaching efficiency above 90%, as shown in Table 3. Other leaching processes of PGMs that have been reviewed by Jha et al. [38] were carried out using lixivants, such as sulfuric acid, sodium cyanide, chloride, iodide, or iodine solutions. The recovery of palladium and rhodium from spent catalysts by leaching in HCl/CuCl_2 media was studied by Nogueira et al. [49] and obtained 2.1 ± 0.1 M for Pd and 1.0 ± 0.3 M for Rh; with Cu^{2+} , the values were 0.42 ± 0.04 M for Pd and 0.36 ± 0.06 M for Rh. Yakoumis et al. reported on the hydrometallurgical process with HCl, NaCl, and H_2O_2 , and recovery rates of 100%, 92%, and 60% were obtained for Pt, Pd, and Rh, respectively [50]. In addition, the effect of ultrasound-assisted nitric acid pretreatment in enhancing the leaching of PGMs by cyanogenic bacteria is a promising biorecovery technique for the extraction of PGMs from spent automotive catalysts [51,52].

Table 3. Summary of the leaching process of PGMs from spent catalytic converters.

Reagents	Leaching Efficiency (%)	Results and Notes	Reference
3HCl:1HNO ₃ (aqua regia)	Pt (97.58%) Pd (93.23%)	The dissolution of Pt and Pd by aqua regia was (97.58%) and (93.23%), respectively, at a time of 120 min, temperature 100 °C, and S/L ratio 1/20.	[28]
1 M HCl 5 M NaCl 10% H ₂ O ₂	Pt (95.7%)	Calcination pretreatment enhanced the leaching efficiency of Pt at 800 °C calcination temperature. Pt leaching was 95.72% at 90 °C for 2 h with conditions of 1 M HCl, 5 M NaCl, S/L of 1:5, 10% H ₂ O ₂ : spent catalysts of 0.6 mL/g.	[46]
2 M HCl 1 M H ₂ O ₂	Pd (>90%)	The presence of H ₂ O ₂ showed improvement of Pd leaching by HCl to above 90%. Conditions were 2 M HCl, 1 M H ₂ O ₂ , at 25 °C, and reaction time 10–15 min.	[44]
2 M HCl 4 M NaCl 0.67 M Fe ³⁺	Pd (99.5%)	The mixture solution of NaCl and HCl was employed as the leaching agent and FeCl ₃ as the oxidant. The optimum leaching conditions were determined to be HCl concentration of 2.0 M, NaCl concentration of 4.0 M, Fe ³⁺ concentration of 0.67 M, temperature of 80 °C, and leaching time of 90 min with a leaching efficiency of Pd (99.5%)	[47]
6 M HCl 10% H ₂ O ₂	Pt (99.9%)	Calcination pretreatment at 800 °C removed the coke and organics, decomposed PtO ₂ , and enhanced the leaching efficiency of Pt (99.9%). The leaching conditions: 6 M HCl, L/S = 10, 10% H ₂ O ₂ : spent catalysts (0.6 mL/g), 90 °C for 2 h.	[46]
9 M HCl (0.8 Vol%) H ₂ O ₂	Pt (96%) Pd (98%) Rh (86%)	The use of 0.8 vol% H ₂ O ₂ and 9.0 M HCl mixture at 60 °C for a contact time of 2.5 h during the leaching process were considered the best conditions to be followed to save chemicals, energy, and time, and resulting efficiency was Rh (86%), Pt (96%), and Pd (98%).	[51]

Table 3. Cont.

Reagents	Leaching Efficiency (%)	Results and Notes	Reference
6M HNO ₃ 1% (<i>w/v</i>) and at pH 9, <i>P. fluorescens</i> and megaterium	Pt (38%)	Using two-step bioleaching at a pulp density of 1% (<i>w/v</i>) and at pH 9, <i>P. fluorescens</i> yielded a maximum recovery of Pt (38%), Pd (44%), and Rh (91%), and megaterium produced Pt (35%), Pd (41%), and Rh (82%).	[51]
	Pd (44%)		
	Rh (91%).	Ultrasound-assisted nitric acid pretreatment preceding bioleaching by cyanogenic bacteria is a promising biorecovery technique for the extraction of PGMs from spent automotive catalysts.	
	Pt (35%)		
	Pd (41%)		
	Rh (82%)		

2.2.4. Recovery and Purification of Leachates

Further processing or purification of PGMs has been done via solvent extraction (SX), precipitation, ion exchange, and cloud point extraction (CPE) [53–55]. Chromatographic separation methods [56–58] have also been investigated for PGM recovery from chloride leach liquors. The efficiency and selectivity of the extractant(s) are key factors for advancing a successful separation of the PGMs from other contaminant metals or between PGMs. In addition, the iron matte as a collector to extract PGMs from catalytic converters has been reported [28]. The process was studied at a relatively low temperature (950 °C), and the highest recovery percentages were 99% Pt, 99% Pd, and 97% Rh [28].

Purification of the leachates has been investigated utilizing several methods, as shown in Table 4, with a recovery rate greater than 80% [59]. The separations are based on selecting suitable separating agents to be successively applied to the PGM solution to separate them individually. Precipitation is one of the commonly used methods to recover and separate PGMs individually as insoluble substances which may be treated after filtration to obtain their pure metallic form. The cyanide extraction method, based on leaching utilizing sodium cyanide under pressure at a temperature about 120–180 °C, is an expensive method and produces cyanide waste which is dangerous to the environment [60]. Ruiu et al. reported the use of supercritical CO₂, and the extraction process included the addition of CO₂-soluble complexing polymers bearing pyridine units, which resulted in more than 70% of Pd extracted from an aluminosilica-supported catalyst [61].

High-temperature methods for the recovery of PGMs from spent catalytic converters have been reported. The use of smelting, where the catalytic converters are melted with iron at a temperature higher than 2000 °C, has been reported, but this method is very expensive [31]. The slag from the metallic phase is separated due to the difference between its density, the obtained metallic phase is leached in H₂SO₄, and, consequently, iron is removed from the platinum solution. A metal collector or whole catalytic converter carrier is melted with the addition of metal collectors, such as copper, magnesium, and calcium, resulting in an alloy of the metal collector (Cu, Mg, Ca) with PGM metals and slag being obtained. The slag is easily separated from an alloy of Pt and a metal collector. This alloy can be refined electrolytically, and Pt goes to the slime, whereas copper splits on the cathode [28]. The Pt-containing slime is then dissolved in aqua regia (a mixture of HNO₃ and HCl in the ratio 3:1), and H₂PtCl₆ is obtained, which is precipitated. The last stage is the platinum-refining process, and much liquid waste is produced which needs to be neutralized [42]. The chlorination method has also been reported to be done at high temperatures (higher than 1200 °C) to evaporate the metallic fraction. This process is expensive; uses chlorine, which is dangerous to the environment; and much liquid and solid waste are created [56]. Morcali reported the use of the classic iron smelting process at a higher temperature, and the recovery percentages were 99% Pt, 99% Pd, and 97% Rh [62].

Table 4. Summary of recovery of PGMs from spent catalytic converters.

Method of Extraction	Recovery (%)	Results and Notes	Reference
Solvent extraction	Pt (80%) Pd (98%)	Pt and Pd were coextracted using 0.01 M tri-isooctylamine (Alamine 308) dissolved in kerosene with a 5% <i>v/v</i> n-decanol used as a modifier, shaken for 30 min at 300 r/min, 1:1 O/A volume ratio at an optimum 5 M HCl. The recovery for Pt and Pd using Alamine 308 was 95% and 81%, respectively.	[46]
Cloud point extraction (CPE)	Pd (97%) Pt (96%) Rh (91%)	The recovery of Pd, Pt, and Rh using CPE was 97%, 96%, and 91%, respectively, after complexation with 2-mercaptobenzothiazole (complexing agent) in the presence of Triton X-100 (surfactant) and tin (II) chloride dihydrate as a reductant, at 90 °C with an incubation time of 120 min.	[46]
Precipitation	Pt (91.9%) Pd (98.9%)	The recovery efficiency of Pt and Pd was 92% and 99%, respectively, precipitated using ammonium chloride (NH ₄ Cl) and sodium chlorate (NaClO ₃) as precipitation agents at 50 °C for 1 h.	[27]
Precipitation/reduction	Pt (>98%)	Pt leaching efficiency was optimized by employing a response surface methodology followed by reduction. Pt leaching efficiency was over 98% under the optimized conditions: 1.45 M HCl, 4.55 M NaCl, 10% H ₂ O ₂ /spent catalysts of 0.66 mL/g, and 1:4.85 of S/L. The purity of Pt was over 90% by the reduction of iron powder.	[47]
Precipitation	Pt (99.5%) Pd (99.3%) Rh (95%)	The individual precipitation of Pt over Pd and Rh from each other was performed by adding NH ₄ Cl (290 g/L) at 40 °C to their solution, where a yellowish Pt precipitate of (NH ₄) ₂ [PtCl ₆] was obtained and calcined at 800 °C to obtain a fine pure Pt powder of 99.5%. A measurement of 3 g of sodium chlorate (NaClO ₃) was added to obtain a bright red precipitate of insoluble (NH ₄) ₂ [PdCl ₆] and calcined at 900 °C where a pure Pd powder of 99.3% was obtained. The rhodium species in the filtrate was precipitated using KOH as a lemon-yellow rhodium hydroxide (Rh(OH) ₃). The complete decomposition in the air for Rh(OH) ₃ to Rh ₂ O ₃ and then Rh metal was performed by ignition at 1150 °C to produce a grey Rh metal powder of 95.4% purity.	[45]
Solvent extraction and cementation	Pt (99.9%) Pd (99.9%) Rh (83.0%)	Tri-isooctylamine (Alamine 308) was used to selectively extract both Pt (99.9%) and Pd (99.9%). Rh was recovered by cementation using zinc metal powder; about 83% was recovered.	[63]

3. Recovery of Precious Metals from E-Waste

E-waste shows exponential growth every year due to the worldwide demand for technology. Waste printed circuit boards (PCBs) are rich in metal content (Figure 5). PCBs also contain polymers and ceramics, in addition to metals, that can all be recovered through effective recycling means. The precious metals present are palladium, silver, and gold in addition to copper and zinc [64]. The e-waste also contains hazardous substances, such as selenium, lead, hexavalent chromium, arsenic, mercury, and flame retardants. Some of the e-waste is shipped to developing countries, for instance, in Asia and Africa, and waste management is done manually under such unmonitored environments and without good handling equipment [65]. The concentration of toxic metals in the landfills of these countries increases the risk of exposure. China banned solid waste import in 2016, and

reforms were gradually introduced in 2017, and this was driven by the inherent problems downstream, even though a thriving precious metals recycling market was already set up.

The dismantling of e-waste can be performed in two ways, the first one being the manual picking of different components from the e-waste. This approach is very advantageous for reuse of the components but is time consuming. The second way is via simultaneous dismantling where the e-waste is heated and desoldered, followed by manual sorting. This approach is time efficient, but there is a risk of damaging the important components because of the heat involved (Figure 6). Over the years, some companies have tried robotics for the dismantling steps since they can be faster than human involvement [66]. Further separation can be achieved by mechanical treatment, which involves shredding, magnetic separation granulation, and magnetic separation, to form part of a pretreatment process to remove the iron scrap. The step is followed by a selection device, sieving separation, and fluid bed/electrostatic separation, resulting in plastics, copper grains, and aluminium grains [67].

Shredding is mainly the cutting down of electronic waste after dismantling to reduce the size of the e-waste for an effective leaching process. This is followed by the crushing and grinding to a desired size range. This step of pretreatment produces dust which reduces the number of metals to be salvaged due to the complex nature of e-waste. A significant amount of rare earth elements is lost. Separation steps can be simultaneously incorporated with shredding, such as size and shape sieving, magnetic separation, electric conductivity (eddy current, corona electrostatic, and triboelectric separation), and density separation. These can all be done depending on the desired size, although it is reported that the 2-mm size gives an output of high amounts of metal fraction compared with using a lower size [68].

Figure 5. Different types of printed circuit boards: (A) desktop computer, (B) laptop, and (C) television.

Figure 6. The pretreatment steps for e-waste.

Researchers have reported some harmful effects from exposure to e-waste due to metals, such as lead, mercury, and cadmium, and from brominated flame retardants, such as polybrominated diphenyl ether and hexabromocyclododecanes. Kwadwo reported on the recycling risks and opportunities in Africa [69], focusing on a place called Agbogloshie in Ghana where e-waste is handled by young men to gain some form of income. This is conducted by collection, recovery, and reuse companies. One example includes the recovery of aluminium from e-waste to produce cooking pots that are sold to nearby markets [70]. This type of recycling has a huge economic impact on developing countries such as Ghana but, on the other hand, produces a detrimental effect upon exposure to these metals without proper handling gear. The motivation behind the interest in metals contained in e-waste is due to the high metal content in the spent product when compared with their natural ores [71]. For example, printed circuit boards (PCBs) have 20 wt.% copper in most e-waste, but only ~0.62 wt.% is found in natural ores [31].

Most researchers have focused on PCBs because they are found in e-waste at an average of 10 wt.%. The metals in PCB composition are about 30 wt.%, and nonmetals are reported to be about 70 wt.%. Several methods are employed to recover these metals, such as hydrometallurgy, pyrometallurgy, biohydrometallurgy, etc. The leaching process has shown promising recovery yield (90–95%) for precious metals. Although the methods have their drawbacks, improvements have been conducted to decrease the level of waste generation and increase the recovered yield of the metals [72]. Table 5 shows evidence that scrap mobile phones and computers have a high content of precious metals.

Table 5. Composition of metals found in WEEE by weight [72].

Electronic Waste	Metal Content for Different WEEE							
	Weight (g/t)					Weight (%)		
	Ni	Ag	Au	Pd	Fe	Cu	Al	Pb
TV boards scrap	0.3 (5)	280 (6)	20 (25)	10 (6)	28 (4)	10 (39)	10 (13)	1 (2)
Mobile phone scrap	0.1 (0)	1380 (5)	350 (67)	210 (19)	5 (0)	13 (8)	1 (0)	0.3 (0)
PC board scrap	1 (1)	1000 (64)	250 (5)	110 (15)	7 (0)	20 (10)	5 (1)	1.5 (0)
PC mainboard scrap	1.1 (2)	639 (2)	566 (81)	124 (8)	4.5 (0)	14.3 (6)	2.8 (0)	2.2 (0)

Table 5. Cont.

Electronic Waste	Metal Content for Different WEEE							
	Weight (g/t)				Weight (%)			
	Ni	Ag	Au	Pd	Fe	Cu	Al	Pb
DVD player scrap	0.05 (1)	115 (4)	15 (33)	4 (4)	62 (17)	5 (35)	2 (4)	0.3 (1)
TV scrap (CRTs removed)	0.038 (4)	20 (3)	<10 (-)	<10 (-)	-	3.4 (82)	1.2 (9)	0.2 (2)
Printed circuit boards scrap	0.85 (6)	280 (3)	110 (66)	-	12 (1)	10 (19)	7 (4)	1.2 (1)
Portable audio scrap	0.03 (0)	150 (3)	10 (12)	4 (2)	23 (3)	21 (78)	1 (1)	0.14 (0)
Calculator scraps	0.5 (8)	260 (6)	50 (64)	5 (3)	4 (1)	3 (12)	5 (6)	0.1 (0)
Typical electronic device scrap	2 (2)	2000 (3)	1000 (87)	50 (2)	8 (0)	20 (5)	2 (0)	2 (0)

The precious metal recovery from e-waste has been under investigation for over two decades and was initiated by the value of precious metals contained in e-waste. Andreola et al. based their work on utilizing leaching processes, such as oxidative chlorination and cyanidation, to recover gold, copper, silver, and palladium. The yield from mobile phones was reported to be 93%, 95%, and 99% for silver, gold, and palladium, respectively [73]. Zhang et al. reported on leaching processes, such as cyanide, thiourea, thiosulfate, and halide leaching processes, comparing their efficiencies [74]. Zhang et al. reported that a cleaner method of leaching can be achieved for gold recovery through thiourea leaching when compared to iodide, aqua regia, and cyanide leaching methods. This report was motivated by the environmentally friendly method of thiourea against the conventional methods and the selectivity for gold [72]. Due to the disadvantage of utilizing extensive leaching reagents with high waste, Wu et al. reported on the use of polyaniline films and polyaniline-coated cotton to recover gold from an acid/halide solution during e-waste treatment in a hydrometallurgical process [75]. Akcil et al. proposed a soft leaching process to extract gold from e-waste utilizing chlorine leaching [76].

Bioleaching is a process that utilizes biological organisms to extract metals from their ore. This method has been utilized by several industries to extract metals from low-grade ores. Microorganisms have been utilized to recover about 20% of copper, 5% of gold, and a significant amount of nickel [77]. The main origin of bioleaching is based on the use of heterotrophic bacteria, chemolithotrophic prokaryotes, and fungi to recover precious metals from their ores. These can oxidize sulphur and iron (autotrophic bacteria) and are also able to produce cyanide (chemolithotrophic organisms), which is toxic to the environment, with extensive waste treatment required at the end of the process. Another drawback of the bioleaching process is the amount of time it takes for the microorganism to solubilize the metals. The factors affecting the process are pH, low acid generation, and medium sensitivity [77].

3.1. Pyrometallurgy

Pyrometallurgy is the most used method to recover precious metals and base metals from e-waste. The process incorporates smelting, incineration, and high-temperature roasting. Figure 7 shows a scheme of pyrometallurgy to recover metals from e-waste.

- Incinerators

The process is based on heating the e-waste after dismantling to decompose the organic components, resulting in carbon dioxide and water. The process utilizes high temperatures (over 1000 °C) to decrease the number of dioxins being released into the atmosphere. Incineration has been utilized amongst a wide range of industries due to the low operational cost and straightforward process. The emission of pollutants from this step can be monitored by varying parameters, such as temperature, rapid cooling of gaseous phase, and residence time. This process aims at producing metal-rich residue, gases, and oil.

- Pyrolysis

This process is similar to incineration; the difference is that it is conducted in the presence of inert gas and a temperature ranging from 350 to 900 °C. The process can avoid the generation of dioxin, and the resin can be further recycled. The oil obtained from the process can be recycled to form raw materials in the formation of porous carbon, carbon nanotubes, and phenolic resin, as it contains phenol derivatives. The advantage of pyrolysis over incineration is the ability to recycle most of the products formed, but it has the drawback of being difficult to scale up to industrial applications [78].

- Plasma

This is known to be a more efficient and less harmful practice of treatment. Plasma smelting can decompose all detrimental organic compounds simultaneously, producing fewer furans and dioxins. The molten slag produced from the furnace is more stable. The use of high temperatures may result in volatile metals escaping with the off gas when cooling the furnace, hence proper exhaust technology needs to be employed [79]. Szalatkiewicz et al. reported on the benefits of constructing a plasma-based technology for recycling e-waste in urban areas, focusing on the amount of e-waste being produced in Poland and sold to big companies such as Umicore (based in Belgium) [80]. Szewczyk et al. reported on the measurement and control system of the plasmatron plasma reactor for the recovery of metals from PCB waste to motivate the application of the plasma method as a tool of mining e-waste for metals [81].

Figure 7. A pathway for recycling precious metals from e-waste via the pyrometallurgy process [79].

3.2. Hydrometallurgy

The hydrometallurgical techniques involving leaching and purification have been reported to be useful in the field of precious metal recovery from e-waste [82]. Hydrometallurgy initially needs pretreatment steps followed by a leaching process utilizing suitable reagents and the recovery of the metals through purification either by precipitation, solvent extraction, adsorption, or ion exchange [83,84]. In the extraction of base and precious metals from e-waste, an oxidative step is necessary to solubilize the metal from e-waste to be part of a pregnant solution. This is mostly performed under acid conditions with an oxidizing reagent. The typical strong acids used are sulfuric acid, hydrochloric acid, nitric

acid, and aqua regia, as shown in Figure 8. However, within chemical leaching methods, cyanide, halide, thiourea, and thiosulfate leaching have also been used.

Figure 8. Different hydrometallurgical leaching processes depending on the metals of interest [69].

Thiosulphate has been explored due to highly toxic cyanide salts which pose detrimental effects. Although these have been employed on an industrial scale, they still pose a significant negative impact due to acid wastewater generation. Cyanide leaching is very efficient for gold leaching at very low grades (1–3 g/t), but there is concern around this process due to the highly toxic nature of cyanide. Thiourea, on the other hand, has a high leaching rate, but the complex formed has low chemical stability. Halide leaching has shown a healthier approach with high chemical stability but has challenges in application as it has not been fully developed in industries. Table 6 shows the mechanisms of the leaching agents that are most widely used to recover precious metals (in this case gold).

Table 6. Methods of leaching gold from e-waste [85].

Leaching Agent	Reagents and Conditions	Chemical Equation Involved
Cyanide	CN^- , Air (O_2) with pH > 10 at 25 °C	$4\text{Au} + 8\text{CN}^- + \text{O}_2 + 2\text{H}_2\text{O} \rightarrow 4\text{Au}(\text{CN})_2^- + 4\text{OH}^-$
Thiourea	$\text{CSN}(\text{NHS})_2$, Fe^{3+} with pH = 1–2 at 25 °C	$2\text{Au} + 4\text{CSN}(\text{NHS})_2 + 2\text{Fe}^{3+} \rightarrow 2\text{Au}[\text{CS}(\text{NH}_2)_2]_2^+ + 2\text{Fe}^{2+}$
Thiosulfate	$\text{S}_2\text{O}_3^{2-}$, NH_3 , Cu^{2+} with pH > 8–11 at 25 °C	$4\text{Au} + 8\text{S}_2\text{O}_3^{2-} + \text{O}_2 + 2\text{H}_2\text{O} \rightarrow 4[\text{Au}(\text{S}_2\text{O}_3)_2]^{3-} + 4\text{OH}^-$
Halide	Cl^-/Cl_2 , Br^-/Br_2 , I^-/I_2 with pH < 4 at 25 °C	$2\text{Au} + 11\text{HCl} + 3\text{HNO}_3 \rightarrow 2\text{HAuCl}_4 + 3\text{NOCl} + 6\text{H}_2\text{O}$

Table 7 represents some work done on the recovery of strategic metals with their recovery efficiency. Although hydrometallurgy has been proven to have great advantages, such as low capital costs, selectivity for metals, and no dust generation, improvements must still be made to reduce the use of harsh solvents such as cyanide. Figure 9 shows a diagram of the hydrometallurgy method for recovering precious metals from e-waste [69]. Hilal reported on the use of a composite material (polyacrylic acid/bentonite) to recover precious metals from e-waste processed in aqua regia. The results reported showed that the adsorption efficiency of Au^+ is higher than Ag^+ from adsorbent material, and the highest values are (53.238%, 43.3904%), (51.778%, 41.99%), and (51.278%, 39.595%) for (Au, Ag) [86]. Hong et al. reported a nanoporous polymer method to recover gold, and through a reductive capture mechanism, gold was recovered at 10 times the theoretical limit, reaching a record 1.62 g/g [24]. Arshadi et al. examined fax machine printed circuit boards, copy machine printed circuit boards, and a central processing unit. They adopted the optimal digestion method using aqua regia, hydrogen peroxide, hydrofluoric acid,

and boric acid. The results showed the recovery value of precious metals with gold and palladium accounting for the highest recovery among other precious metals [87].

Figure 9. A hydrometallurgy method for recovering precious metals from e-waste [69].

Table 7. Different leaching reagents and recovery yields.

E-Waste	Leaching Method	Metal(s)	References
Printed circuit boards	Thiourea and thiosulfate leaching	Au (40%) and (70%)	[85]
CPU	Slurry electrolysis (HCl, NaCl, and H ₂ O ₂)	Cu (67%) and Au (97.73%)	[88]
A mixture of electronic waste	Acid/halide leaching	Au (90%)	[89]
Random-access memory device	Iodine and hydrogen peroxide, ammonium persulfate	Ag (96.90%), Au (99.98%), and Cu (98.73%)	[90]
Mobile phone PCBs	HCl assisted iodine–iodide leaching	Au (63.3%), Ag (81.5%), and Pd (60.5%)	[91]
Mobile phone PCBs	Reduction smelting at a temperature of 1623 to 1698 K	Up to 95% of Au, Ag, Pt, Pd, and Rh at 1673 K	[38]

3.3. Biohydrometallurgy

Biohydrometallurgy is a technique used to manipulate microorganisms to produce the leaching agent or oxidative species for the leaching of metals. This technique has been applied in the recovery of metals from low grade ores. Table 8 shows a summary of the work done on bioleaching thus far. Bioleaching is ecofriendly, can be conducted at room temperature, uses less industrial equipment, does not require highly skilled workers, has less energy consumption, and is cost effective [92]. The most used microorganism for leaching base metals from PCBs is *C. violaceum* because of its ability to produce more cyanide due to the availability of its six HCN synthase enzymes. The recovery of Au by cyanogenic bacteria is less efficient when copper is present because of the ability of copper to form complexes more easily than gold. Hence, researchers have improved this by having a two-step method when recovering gold from PCBs. The first step is to remove base metals

by acidic leaching (hydrometallurgy) or the use of chemolithoautotrophy (bioleaching) followed by treatment with cyanogenic bacteria to recover precious metals [92].

Although bioleaching has promising results, the complex nature of PCBs does influence the mortality of the microorganisms being employed. Bhat et al. proposed a method of utilizing low-cost biomass laden with L-cysteine for the biosorption of gold/silver cyanide to obtain a more concentrated solution of gold/silver cyanide prior to the use of electroplating [83]. Chi et al. reported on the bioleaching of mobile phone printed circuit boards to recover copper and gold [93]. Liu reported on the recycling of electronic waste to recover gold utilizing *C. violaceum* with added nutrients to increase its efficiency [94]. Marra et al. reported the recycling of shredded dust from e-waste to recover gold and rare earth metals utilizing bioleaching [95]. Işildar et al. reported on a two-step bioleaching method to recovery gold and copper from discarded PCBs [96]. Jokar et al. reported the bioleaching of liquid crystal displays to recovery indium and strontium [97]. Table 8 shows the percentage values of the recovered metals together with the methods employed. Bioleaching has some drawbacks, such as the number of metals recovered and the time it takes for the metals to be leached.

Table 8. A summary of metal recovery from e-waste utilizing bioleaching processes.

E-Waste	Method	Metal(s) Recovered	Reference
Mobile phone PCBs	<i>C. violaceum</i> in yeast extract and polypeptone with glycine medium	Cu (24.6%) and Au (11.4%)	[93]
Electronic scrap	<i>C. violaceum</i> and nutritive salts	Au (70%)	[94]
Shredded dust from electronic waste	<i>A. thiooxidans</i> , and <i>P. putida</i>	Ce, Eu, Nd (>99%), La, Y (80%), and Au (48%)	[95]
Printed circuit boards	<i>Thiooxidans</i> , <i>At. ferrooxidans</i> ; <i>P. putida</i> and <i>P. fluorescens</i>	Cu (98.4%) and Au (44%)	[96]
Liquid crystal displays	<i>thiooxidans</i>	In (100%) and Sr (10%)	[97]

4. Recovery of Important Metals from Batteries

The global demand for batteries has rapidly increased over the past two decades due to the improvement of technology. This is a result of the low cost of, low maintenance of, and demand for portable electronic devices. In 2012–2013, 400 million batteries were purchased in Australia with 14,703 tons ending up in landfills and 404 tons recycled [98]. In 2015, 7 billion lithium-ion batteries were consumed globally [99]. Batteries can be divided into two groups, namely rechargeable (Ni-Cd, Ni-MH, LIBs) and nonrechargeable (Zn-Mn, Zn-C, mercury, and primary LIBs) batteries. The spent battery e-waste is made up of an anode (LiCoO₂, LiMn₂, LiNiO₂, or LiFePO₄), cathode (graphitic carbon), electrolyte (lithium salts: LiClO₄, LiBF₄, LiPF₆, and LiCF₃SO₃ dissolved in an organic solvent), separator, and collectors (Cu foil for the anode and Al foil for cathode and metal protective shell) [100].

The depletion and high rise of consumer consumption may lead to a shortage of lithium as raw material together with elements used in the manufacturing of batteries. The incorrect disposal of these batteries may cause a serious environmental impact especially to ecosystems situated close to landfills. Batteries present a secondary source for recovering important metals. Lead-acid batteries have been recycled to recover lead alloy, scrap plastic, and sulfuric acid for reuse (Figure 10). These lead-acid batteries are considered environmentally friendly since 60–80% of the materials used to manufacture them are recycled [101].

Figure 10. A systematic representation of the lead-acid battery process [102].

Figure 11 presents a typical pretreatment approach for lithium-ion batteries where the first step is to discharge the batteries to avoid a short circuit when they go to the dismantling process. The dismantling process is mainly done manually, although Li et al. made an automated prototype machine named the z-folded electrode separator [102]. The mechanical treatment follows, which involves the crushing and hammering of the electrodes to powder form to reach a size of approximately 5 mm. Sieving is mainly used to remove chunks of plastic material while the metal content sinks to the bottom (density separation). Pagnelli et al. reported on the optimization of preprocessing conditions and specifically studied the effect of physical treatment of the waste fraction of lithium-ion batteries (LIBs) [103]. They used two different physical treatment methods, the first one being crushing, milling, and sieving and the second one being granulation and sieving. For the first treatment, they recovered about 50% of the initial waste; for the second method, they recovered 37% of the powder. The size of the first sample (with 50% recovery) was <0.5 mm and the size of the second sample (with 37% recovery) was <1 mm [103].

Recently, Zhong et al. reported on an improved pretreatment method which does not involve the discharge step. The process of crushing and low-temperature volatilization of LIBs reduced secondary pollution produced by the discharging step [104]. Ebin et al. reported on the physical separation, mechanical enrichment, and recycling-oriented characterization of spent NiMH (nickel–metal hydride) batteries [105]. They used a Retsch SM 300 cutting mill at 2900 min^{-1} rotation speed with continuous feed inlet. The cutting mill had a sieve (4 mm) located underneath to collect the black mass obtained. They used 5–10 min milling time to compare the samples and sample loss. They noticed that with an increased grinding time, the weight of the <4 mm fraction increased (76.8% to 86.9%) and the >4 mm fraction decreased (9.7% to 1.0%).

Figure 11. The pretreatment steps for a lithium-ion battery [103].

Other elegant works demonstrate the approaches for recovery of metal value from spent batteries. Xiang et al. reported on the recycling of a Zn-Mn battery to zinc nanoparticles [101]. Zn-C battery recycling to zinc oxide film has been reported by Hassan et al. [106]. Naseri et al. reported on the recovery of lithium, cobalt, and manganese from coin cells [99]. Maroufi et al. reported on the recycling of a Zn-C battery to zinc oxide porous nanosheets [98]. Horeh et al. reported on the recovery of copper, lithium, manganese, aluminium, cobalt, and nickel from a lithium-ion mobile phone battery [107]. Li et al. reported on the recovery of metals (cobalt and lithium) from a lithium-ion battery [108]. The explanation of the literature above is shown in Table 9. The chemical leaching process for batteries includes sulphuric acid, which is a cost-effective reagent [109]. For the identification and influence of several parameters during leaching for the removal of metals from spent batteries, a Box–Behnken design was used in determining acid concentration, solid/liquid ratio, retention time, and cycles of leaching. The solid/liquid ratio and acid concentration influenced the solubilization of zinc, manganese, nickel, cadmium, and cobalt from spent batteries [110]. Pyrometallurgy and hydrometallurgy approaches have been employed to recycle metals from batteries, but their disadvantages have led most researchers to explore environmentally friendly approaches and cost-effective techniques, such as bioleaching, although this is still at the laboratory stage.

Table 9. A summary of work done in recycling batteries to metals and value-added materials.

Waste Product	Method	Metal(S) Recovered	Reference
Zn-Mn batteries	Vacuum separation and inert gas condensation	Zn nanoparticles	[105]
Zn-C batteries	Facile thermal route	ZnO nanoparticles	[106]
Coin cells	Two-step bioleaching utilizing <i>Acidithiobacillus thiooxidans</i>	Li (99%), Co (60%), and Mn (20%)	[99]

Table 9. Cont.

Waste Product	Method	Metal(S) Recovered	Reference
Zn-C batteries	Homogeneous precipitation-calcination route	ZnO porous nanosheets	[100]
Lithium-ion mobile phone batteries	Bioleaching using <i>Aspergillus niger</i>	Cu (100%), Li (95%), Mn (70%), Al (65%), Co (45%), and Ni (38%)	[107]
Lithium-ion batteries	Succinic acid leaching agent and hydrogen peroxide as an oxidant	Co (100%) and Li (96%)	[108]

5. Conversion of Waste Metals Directly to Nanomaterials

Nanomaterials are of great importance in the advancement of electronic devices. This is due to their intrinsic properties at the nanoscale compared to bulk material. These include a high surface area to volume ratio and improved optical, electrical conductivity, and photochemical properties. An alternative to recovering metals from e-waste would be to produce value-added materials such as nanomaterials [111]. Xu et al. reported on biowaste valorisation into valuable bio(nano)materials, showing the importance of adding value to waste materials as a necessity [112]. Devi et al. reported on the recovery of e-waste (Duracell AA discharged batteries) as nanostructures (carbon quantum dots and plasmonic carbon quantum dots) by employing pyrolysis in a hot air oven. The method produced ~5 nm and ~20 nm for carbon quantum dots and plasmonic carbon quantum dots, respectively [113]. Fang et al. reported the use of steel plant pickling waste material to iron nanoparticles by reducing the iron with sodium borohydride and removing excess reducing agent by rinsing the nanoparticles formed with a deoxygenated water and ethanol solution [114]. Samaddar et al. reviewed various pretreatment methods that have been developed and applied for the conversion of wastes into useful forms of nanomaterials (e.g., metallic/carbon nanoparticles, carbon nanotubes/nanosheets, and nanoactivated carbon) via chemical and thermal activation, arc discharge, vacuum evaporation-inert gas condensation, sodium borohydride reduction, and solvent thermal approaches [115]. The literature has shown that researchers have made advances in synthesizing nanoparticles from e-waste, and some of these nanoparticles have been reported with their potential applications. Figure 12 shows a schematic diagram of e-waste to silver nanoparticles. Extensive work has been done on producing copper nanoparticles from e-waste by using hydrogen peroxide as an oxidant. Chen et al. reported on the leaching of copper from e-waste utilizing five different ionic acids with hydrogen peroxide as an oxidant. They investigated the effects of temperature, particle size when shredding the e-waste, ionic liquid concentration (V/V in liquid solution), time, and the ratio of the ionic liquid acid amount to the oxidizing agent [116]. Zhang et al. reported on a green chemical and cost-effective way to recover copper from e-waste utilizing slurry electrolysis with ionic liquid acid (N-butylsulfonate pyridium hydrosulfate [BSO₄HPy]·HSO₄). The ionic liquid replaced sulphuric acid in the slurry electrolysis which showed an improved recovery rate (90.4%), 70.68% current efficiency, 81.69% purity, copper powder of 2.30 µm, and a change in morphology (scattered monocrystalline particles) [117]. Tatariants et al. reported on the recovery of copper from e-waste (motherboards, video cards, and random-access memory) to produce antimicrobial copper nanoparticles. The first step was to convert recovered copper to copper sulphate and reduce it by using ascorbic acid, employing β-cyclodextrins as a stabilizing agent to produce nanoparticles. They reported a 90% yield with a size distribution of 7 nm [118].

Nascimento et al. reported their work of synthesizing polymetallic nanoparticles from printed circuit boards for application in textile dye remediation [119]. Marouf et al. reported on the recycling of glass fraction of old computers as the silica source and computer plastic cover as the carbon source to synthesize silica carbide nanowires with a diameter of 30–200 nm and length of 10 µm through carbothermal treatment for electronic, optoelectronic,

and sensor application [120]. Majumder reported on the use of integrated circuit boards to synthesize nano-sized copper with a size of 20 nm, utilizing bacteria to obtain a size of 84–184 nm, and fungi producing a size of 93–115 nm [121]. Rajarao et al. reported on the use of compact discs as a carbon source to synthesize silicon carbide nanowires with a width of 40–90 nm [122]. Nayak et al. reported on the recycling of memory slots to synthesize ZnO/CuO nanocomposites for photocatalytic properties [123]. Moriwaki et al. utilized gold-containing electronic waste to produce gold nanoparticles [124]. Swain et al. reported the use of scrap low-temperature cofired ceramic to synthesize silver metal nanopowder [125]. Shokri et al. reported on the use of old PCBs to synthesize copper–tin nanoparticles [126]. Table 10 shows an example for the conversion of e-waste to silver nanomaterials. Recently, Nekouei et al. reported a review of value-added products from waste printed circuit boards. Their recommendation was on the valorisation of all materials in e-waste, including polymers, metals, and ceramics [127].

Figure 12. Conversion of e-waste to value-added nanomaterials (AgNPs as an example) [127].

Table 10. A summary of literature on conversion of e-waste to nanomaterials.

E-Waste Type	Method	Nanomaterial(s)	Reference
Glass fraction of obsolete computer monitors as a source of silica and computer plastic shell as a source of carbon	Uniaxial hydraulic press by applying 3 bars of pressure at 180 °C for 20 min and heated isothermally at 1550 °C in a tubular furnace (100 cm length × 5 cm diameter) under argon purge (1 L min ^{−1}) for 150 min	Silica carbide nanowires (30–200 nm in diameter) and a length of 10 μm) for electronic, optoelectronic applications, and sensors	[120]
Integrated circuits from electronic boards	Leaching was conducted using fungi and bacteria to obtain nano-sized copper leachate; the incubations were 21 days for fungi and 4–5 days for bacteria. The reduction of nano-sized copper was conducted utilizing a leaf extract (Lantana camara weed)	Nano-sized copper obtained was 84–130 nm for bacteria and 93–115 nm for fungi. The copper nanoparticles obtained were 20 nm after reduction	[121]

Table 10. Cont.

E-Waste Type	Method	Nanomaterial(s)	Reference
Compact disc as a carbon source	Carbothermal reduction at 1150 °C	Silicon carbide nanoparticles width size of 40–90 nm	[122]
Printed circuit boards (memory slots)	Leaching with aqua regia at 60 °C Nanoparticles synthesized by hydrothermal synthesis protocol	ZnO/CuO nanocomposites	[123]
Gold-containing electronic waste	Electrochemical extraction of gold	Gold nanoparticles	[124]
Scrap low-temperature co-fired ceramic	Leaching-precipitation-wet chemical reduction	Silver nanoparticles	[125]
Old computer printed circuit boards	Cryogenic grinding to 100 µm and followed by rapid heating and cooling to avoid oxidation	Cu–Sn based alloys	[126]
Old computer printed circuit boards	Thermal micronizing (heat treatment)	Copper–tin nanoparticles	[112]
Printed circuit boards	Leaching utilizing aqua regia and persulfate/sulfuric acid system, respectively; formation of nanoparticles in sodium hydroxide	Polymetallic nanoparticles	[120]

6. Summary and Perspectives

The development of methods for the recovery of precious metals from spent products has made significant progress over the past ten years. However, further improvements are constantly being made to increase the yields from the recycling process and to extend the range of metals that can be recycled [14]. The lack of uptake of current metal recycling technologies is due to a reliance on primary mining operations, where a significant amount of money has been invested, and the environmental concerns from processing. There is no question about the profitability of the secondary metal streams, and at this stage, much investment is required to set up commercial entities. The investment should possibly come from the profits of primary mining activities in order to contribute to sustainable mining. To address the technical and market challenges, there is a need to develop a well-tuned recycling chain, consisting of different specialized stakeholders, starting with the collection of old products, followed by the sorting/dismantling and preprocessing of relevant fractions, and finally, the recovery of the metals. In addition, to ensure the high recovery of precious metals from secondary waste, the optimization of technological refining operations and systems for organizing the collection and preparation of raw materials are necessary [128].

The recycling processes for catalytic converters have several stages, such as the pre-processing and homogenization of a substrate; processing of PGM content by pyro- and/or hydrometallurgical methods; refining PGM concentrate, which consists of dissolving and removing non-PGM elements; separation of PGM metals; and PGM purification, which produces a high-grade powder. However, the content of PGMs in catalytic converters depends on the catalytic converter construction and on the function of the catalytic converters and their manufacturer. This also makes it difficult to identify catalytic converters by age and type once removed from scrap cars. There needs to be a management of data during the car scrapping process to make the products useful for processing. There are concerns around chemical processes for the recovery of important metals from e-waste and other sources of spent metals [109], and the main challenge is the environmental pollution downstream of the processing units. The microwave method for the extraction of PGMs from solid waste reduces both the volume of acid and the time required to recover PGMs [129]. However, greener processes are being explored for leaching important metals [130]. The biorecovery

of PGMs from waste to produce high-value materials that are useful for several applications is recommendable [131–134].

Refining industries that are dedicated to processing these secondary sources should become important in the future. The current status and future perspectives of precious metal recovery from secondary production require the development of relevant methods, as traditional methods for primary ore solutions are time consuming and inefficient for simultaneous recovery of precious metals. Precious metal recycling from end-of-life products has become an increasingly important source of supply and is expected to continue increasing. The sector continues to strengthen the precious metal supply from primary production, but industries are gaining interest in precious metal recycling, and the sector is likely to see recycling close the gaps for demand. Precious metals are becoming depleted, and the current production struggles to meet demand; therefore, it is necessary to recognize that the primary and secondary production of precious metals are complementary and mutually dependent, with both required to meet the growing demand for the metals [38].

Globally, countries are seeking to recycle greater quantities of the components and materials currently being discarded to waste dumps, and recycling legislation is being implemented [135]. South Africa is one of the countries with recycling legislation and strives to control the waste produced from its mining industries and other sectors [136]. Other countries that have the processing power for waste streams, such as China, have also banned the import of solid waste to prevent the accumulation of further waste, and this necessitates other countries to process their waste and drive towards a circular economy. South Africa has embraced a circular economy with other global players, such as the UK, the EU, China, and the USA. A transition to a circular economy shifts the focus to reusing, repairing, refurbishing, repurposing, and recycling, and this means that businesses can redesign complete supply chains for resource efficiency and circularity, creating a new market responding to shifts in consumption patterns away from traditional ownership towards using, reusing, and sharing [135]. The proposed policy plan on resource efficiency to date has been the circular economy package, giving long-term guidance on increasing resource productivity while minimizing environmental impacts. Africa has mainly been focused on dismantling and reusing [69], but there is a concerted effort to understand the scope of waste management and opportunities for recycling, especially in South Africa [136–138]. There are currently few companies in South Africa that are involved in the preprocessing and processing stages.

Metals are not consumed and are only transferred from one form into another, with the major loss being the ionic form, which ends up in the environment. Urban mining needs to be viewed as contributing towards sustainable mining, especially if recycling rates can be increased. The nonrenewable character of minerals from primary mining may be less constraining than it may seem if considered in line with principles of sustainable development. Urban mining needs to be a compulsory activity for every country, and the illegal dumping of waste must be monitored by regulatory agencies.

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