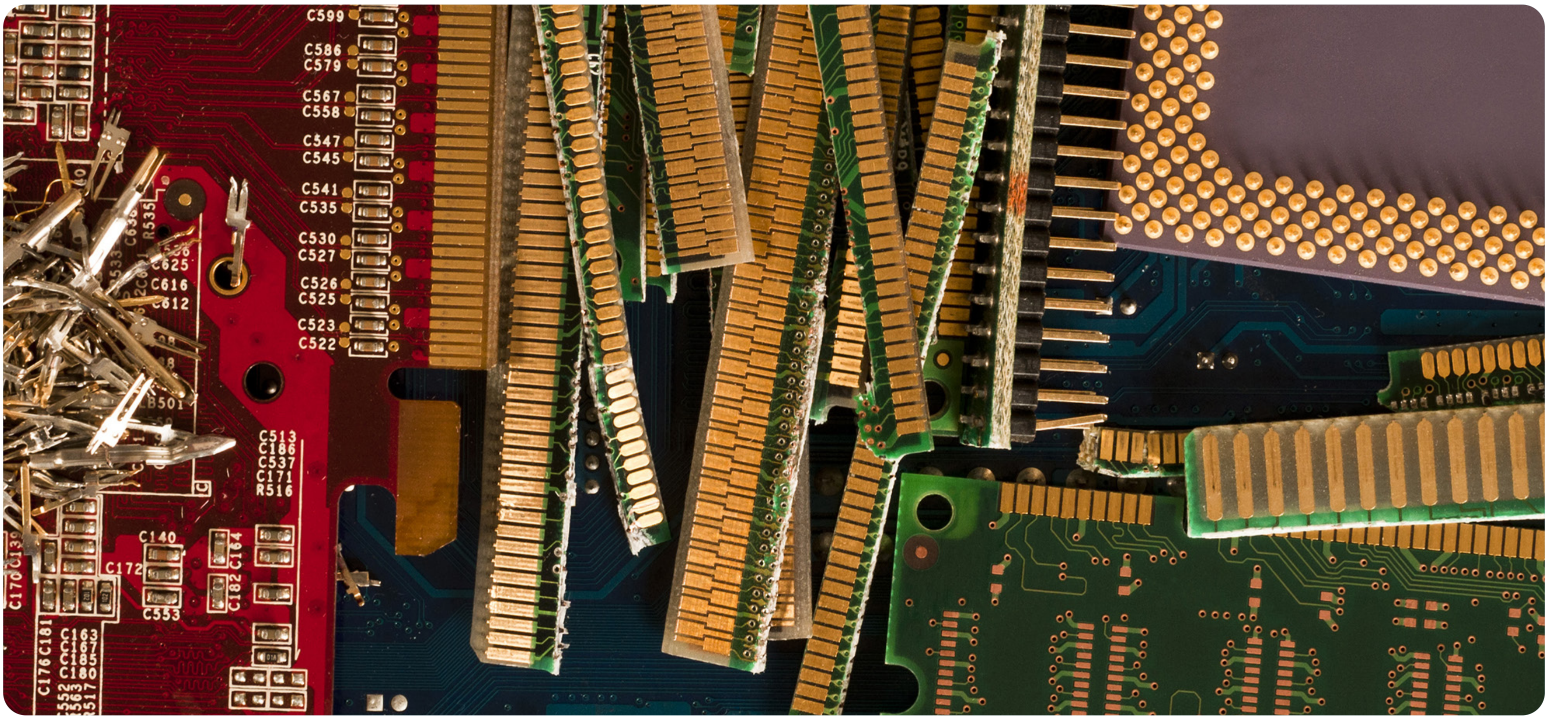
MATERIAL FOCUS

Research

# Recovering critical raw materials from waste electricals

July 2021



# Technology Roadmap and Taxonomy of Critical Raw Material (CRM) Recovery Technologies for Waste Electrical and Electronic Equipment (WEEE)

A report outlining emerging technologies with potential to increase recovery of CRMs from WEEE, in support of developing next generation recycling strategies.

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[1] DBA Design Effectiveness Award (Gold) with Virgin Atlantic Airways (VAA). Green Apple (Gold) awarded at House of Commons.

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# Glossary

## Acronyms

| | |
|---|---|
| AATF | Approved Authorised Treatment Facility |
| ATF | Authorised Treatment Facility |
| BIS | Department for Business, Innovation and Skills |
| BM | Base metal |
| BMS | Battery Management System |
| CRM | Critical Raw Materials |
| CEP | Circular Economy Package |
| DEC | Diethyl carbonate |
| DES | Deep Eutectic solvents |
| DMC | Dimethyl carbonate |
| EAF | Electric arc furnace |
| EC | Electrical/electronic component |
| EEE | Electrical and Electronic Equipment |
| EHF | Electrohydraulic fragmentation |
| EMC | Ethyl methyl carbonate |
| EoL | End-of-life |
| GHG | Greenhouse Gas |
| GWP | Global Warming Potential |
| HD | Hydrogen decrepitation |
| (H)EVs | Hybrid electric vehicles |
| HREE | Heavy rare earth elements |
| HDD | Hard disk drive |
| IL | Ionic liquids |
| IR | Infra-Red |
| ITO | Indium doped tin oxide |
| LCD | Liquid Crystal Display |
| LIB | Li-ion battery |
| LFP-C | Lithium iron phosphate battery with graphite anode |
| LFP-LTO | Lithium iron phosphate battery with lithium titanite anode |
| LFYP | Lithium-iron-yttrium-phosphate ($LiFeYPO_4$) battery |
| LME | London Metal Exchange |
| LMO-C | Lithium manganese oxide (spinel) battery with graphite anode |
| LREE | Light rare earth element |
| MFA | Material flow analysis |
| MLCCs | Multi-layered ceramic capacitors (SMCs) |
| NCA-C | Lithium nickel cobalt aluminium oxide battery with graphite anode |
| NCM-C | Lithium nickel cobalt manganese battery with graphite anode. |
| NIB | Neodymium-iron-boron (Nd-Fe-B) magnetic alloy |
| OTM | Other technology metals |
| PCB | Printed Circuit Board |
| PGM | Platinum Group Metals |
| PE | polyethylene |
| PMs | Precious metals e.g., gold (Au) and silver (Ag)[2] |
| PP | polypropylene |
| PVDF | Polyvinylidene fluoride (binder) |
| REE | Rare earth element (also known as REMs) |
| REMs | Rare Earth Metals |
| RoHS | Restriction of the Use of Certain Hazardous Substances in Electrical and Electronic Equipment (RoHS) EU Directive |
| SMCs | Surface mount components |

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[2] It should be noted, the terms precious metals (PMs) and platinum group metals (PGMs) are generally used interchangeably. However, the EU14 definition of PGMs excludes Au and Ag. To distinguish these sets of metals, further discussion uses the EU14 PGMs definition (Table 3), and PMs refers only to Au and Ag.

| | |
|---|---|
| sc$CO_2$ | Supercritical $CO_2$ |
| sc$H_2O$ | Supercritical water |
| TM | Technology metals |
| TRL | Technology readiness level |
| WFD | Waste Framework Directive |
| WEEE | Waste Electrical and Electronic Equipment |
| XRF | X-Ray Fluorescence spectroscopy |

## Chemical elements referred to in this report

| | |
|---|---|
| Ag | Silver |
| Au | Gold |
| Be | Beryllium |
| Bi | Bismuth |
| Ce | Cerium |
| Co | Cobalt |
| Cr | Chromium |
| Dy | Dysprosium |
| Er | Erbium |
| Eu | Europium |
| Ga | Gallium |
| Gd | Gadolinium |
| Ge | Germanium |
| Hf | Hafnium |
| Ho | Holmium |
| In | Indium |
| Ir | Iridium |
| La | Lanthanum |
| Lu | Lutetium |
| Mg | Magnesium |
| Nb | Niobium |
| Nd | Neodymium |
| Ni | Nickel |
| Os | Osmium |
| Pd | Palladium |
| Pm | Promethium |
| Pr | Praseodymium |
| Pt | Platinum |
| Rh | Rhodium |
| Ru | Ruthenium |
| Sb | Antimony |
| Sc | Scandium |
| Sm | Samarium |
| Sn | Tin |
| St | Strontium |
| Ta | Tantalum |
| Tb | Terbium |
| Tm | Thulium |
| V | Vanadium |
| Yb | Ytterbium |
| Y | Yttrium |

# Terminology

| | |
|---|---|
| **Technology metals** (speciality metals) | Any "rare" metal or a metal that is in demand, available, and used for the purpose of furthering technology and engineered system. i.e., those which impart special functionality. This includes CRM metals, PGMs, PMs, Nickel and Tin. |
| **CRMs** | Critical raw materials: Antimony (Sb), Baryte, Bauxite, Beryllium (Be), Bismuth (Bi), Borates, Cobalt (Co), Coking coal, Fluorspar, Gallium (Ga), Germanium (Ge), Hafnium (Hf), HREEs, Indium (In), Lithium (Li), LREEs, Magnesium (Mg), Natural graphite, Natural rubber, Niobium (Nb), PGMs, Phosphate rock, Phosphorous (P), Scandium (Sc), Silicon metal (Si), Strontium (Sr), Tantalum (Ta), Titanium (Ti), Tungsten (W) and Vanadium (V). |
| **REMs** | Rare earth metals (sometimes called 'Rare Earth Elements'; REEs) include LREEs and HREEs. |
| **LREEs** | Light rare earth elements: scandium (Sc), lanthanum (La), cerium (Ce), praseodymium (Pr), neodymium (Nd), promethium (Pm) and samarium (Sm). |
| **HREEs** | Heavy rare earth elements: yttrium (Y), europium (Eu), gadolinium (Gd), terbium (Tb), dysprosium (Dy), holmium (Ho), erbium (Er), thulium (Tm), ytterbium (Yb) and lutetium (Lu). |
| **PGMs** | Platinum group metals: platinum (Pt), palladium (Pd), ruthenium (Ru), iridium (Ir), rhodium (Rh) and osmium (Os). |
| **PMs** | Precious metals: includes gold (Au) and silver (Ag) (this term normally includes PGMs, however, to distinguish between Au and Ag and PGMs included in EU CRM list definition of PGMs, PMs for this report exclusively means gold (Au) and silver (Ag)). |
| **OTMs** | Other Technology Metals: Antimony (Sb), Beryllium (Be), Cobalt (Co), Gallium (Ga), Hafnium (Hf), Indium (In), Lithium (Li), Nickel (Ni), Niobium (Nb), Silicon (Si), Tantalum (Tn), Tin (Sn), Titanium (Ti), Tungsten (W) and Vanadium (V). This is the CRM metals plus tin and nickel. |
| **TRL** | Technology readiness level. This is a measure of the readiness of the technology to help management in making decisions concerning the development and transitioning of technology. This is broken down into 9 different levels:<br>TRL 1 – Basic principles observed<br>TRL 2 – Technology concept formulated<br>TRL 3 – Experimental proof of concept<br>TRL 4 – Technology validated in lab<br>TRL 5 – Technology validated in relevant environment (industrially relevant environment in the case of key enabling technologies)<br>TRL 6 – Technology demonstrated in relevant environment (industrially relevant environment in the case of key enabling technologies)<br>TRL 7 – System prototype demonstration in operational environment<br>TRL 8 – System complete and qualified (proven to work as specified)<br>TRL 9 – Actual system proven in operational environment |

## WEEE categories

1. Large household appliances (e.g., white goods less cooling equipment);
2. Small household appliances (e.g., vacuums, irons, toasters etc.);
3. IT and telecoms equipment (e.g., computers, printers, calculators, phones, answer machines but not displays);
4. Consumer equipment (e.g., radios, hi-fi equipment, electronic musical instruments excluding televisions);
5. Lighting equipment (excluding household lighting)
6. Electrical and electronic tools (e.g., drills, saws, sewing machines (excluding large stationary industrial tools);
7. Toys and leisure and sports equipment (e.g., train sets, video games, coin slot machines);
8. Medical devices (e.g., dialysis machines, ventilators) (Note that this category is not covered by the RoHS Directive at present);
9. Monitoring and Control instruments (e.g., smoke detectors, thermostats);
10. Automatic dispensers (e.g., ATMs, vending machines);
11. Display equipment (e.g., TVs and monitors);
12. Cooling equipment (e.g., refrigeration equipment);
13. Gas discharge lamps and LED light sources;
14. Photovoltaics.

# 1 Research Summary

The UK economy is highly reliant upon imports of technology metals (TMs), considered 'critical' due to their high relative economic importance and high relative supply risk and demand for which is forecast to increase by a factor of 20 for certain materials (EC JRC, 2016). TMs are defined as any "rare" metal or a metal that is in demand, available, and used for the purpose of furthering technology and engineered systems. TMs consist of Critical Raw Material (CRM) metals, platinum group metals (PGMs), precious metals (PMs), nickel and tin. TMs will be used to reference these materials throughout the report, unless the discussion is about specific materials or groups of materials.

Securing access to TM supply is an economic priority for governments and organisations around the world, particularly those that have resource hungry economies, and which rely heavily on imports due to lack of domestic availability.

Many technology metals are essential to the functionality of modern technologies, albeit in most cases used in trace amounts within products. Substitution in many applications is not feasible at this time, so measures to mitigate supply risks are urgently required. As demand for technology metals rises, supply from primary sources may struggle to keep up with demand. In fact, no rapid solutions to issues with primary supply exist. Raw material scarcity, market price fluctuations, geopolitics and heightened environmental consciousness compound the importance of securing a reliable supply of these valuable resources for the UK economy.

Future indispensable (critical) inputs of TMs are required for high-technology applications including electrical and electronic equipment (EEE), industrial systems (such as those which form a part of the fourth industrial revolution; Industry 4.0) as well as environmental technologies, including wind turbines, photovoltaics (PV), batteries and hybrid/electric vehicles ((H)EVs).

Despite almost a decade passing since the UK government first set strategy to increase domestic CRM supplies through recovery from Waste Electrical and Electronic Equipment (WEEE) to mitigate materials criticality issues for UK industry, TM/CRM recycling rates remain low generally, and barely exceed 50% even for the most economically valuable elements, such as gold (Au) and platinum group metals (PGMs). Currently less than 1% of all rare earth metals (REMs) are recycled with the vast majority lost in dust generated by the pre-processing of the WEEE or lost in the slag during metal recovery. In the UK there is very little in the way of TM recovery from WEEE, apart from the pre-processing of printed circuit boards (PCBs) and limited small-scale precious metal (PM) and platinum group metal (PGM) recovery.

WEEE is a potentially rich primary source of TMs. The Global E-waste Monitor 2020 reported UK WEEE generation to be ~1.6 Mt in 2019 or 23.9 kg/cap, one of the highest per capita WEEE generation rates in Europe and the World (Forti et al., 2020). This highlights the potential of domestic TM recovery from WEEE as a means to mitigate materials criticality issues for UK industry. Additionally, the valuable TMs contained in more than 500,000 tonnes of WEEE each year are wasted, as these end up in low-grade scrap processing, landfill, incineration, commercial and industrial waste, are stolen or are illegally exported (Material Focus, 2020a).

Even though WEEE legislation has resulted in an infrastructure for collection and treatment of WEEE, the rate of recovery of TMs from this waste stream remains low. Previous analysis of the flows of CRMs contained in UK WEEE through recycling process chains concluded that no CRMs are recovered from WEEE in the UK owing to the lack of domestic recovery infrastructure, and high losses of CRMs through dissipation in pre-processing carried out within the UK. However, the means to overcome these barriers have begun to emerge and an opportunity now exists for the UK to establish a circular economy of its own for TMs, capitalising on a high per capita rate of EEE consumption and WEEE generation to develop secondary TM supplies for reliant domestic industries.

This highlights the potential of domestic TM recovery from WEEE as a means to mitigate materials criticality issues for UK industry, improve environmental impacts of manufacturing, and create jobs in a new industry to facilitate recovery and recycling of TMs back into the UK manufacturing economy from end of life (EoL) products, or indeed in manufacturing sectors which can benefit from access to domestic sources of TMs.

This report reviews a wide range of different technologies that have been developed for the recovery of TMs. These include pre-processing methods that can be used to prevent the losses of TMs in the dust fractions and which allow for better segregation of the TMs to aid their recovery.

The technologies reviewed include:

- Solder removal as a pre-treatment process;
- Electrohydraulic fragmentation, steam gasification and super critical water for breakdown down of printed circuit boards;
- Pyrometallurgical methods using heat for metal recovery; and
- Hydrometallurgical methods using acids, ionic liquids, bio absorption and ion exchange for metal recovery.

## 1.1 Objectives

Material Focus commissioned Giraffe Innovation and Swansea University to undertake research with the key objectives:

- Examine the potential for securing supply of TMs through more effective recovery from WEEE- the opportunity;
- Conduct a Material Flow Analysis (MFA) - Quantify potentially recoverable amounts (weight) of TMs from UK WEEE and their value (£) and environmental benefit (carbon dioxide equivalent - $CO_2e$) to the UK economy; and
- Develop a 'Technology Roadmap' - Examine emerging recycling technologies and their potential for deployment in UK WEEE recycling to increase TM recovery rates – the means.

## 1.2 Scope

The material flow analysis (MFA) focussed on WEEE categories where quantitative data on the presence and amount of technology metals is available. This includes selected products within WEEE Category: 3: IT & Telecoms; Category 11: Display Equipment; and Category 13: Gas Discharge Lamps and LED Light Sources. The products in these categories account for an estimated 123,901 tonnes sent for recycling in the UK each year[3]. Specific TM rich and ubiquitous products are analysed including personal computers (PCs), laptop computers, tablets, TVs, monitors, smart phones and lighting. In addition to this there are also products that are not captured in the WEEE stream such as hoarded and stolen products, products arising in commercial and industrial waste and those being illegally exported, however these were not the focus of this report.

It is acknowledged that technology metals are present to varying degrees across other WEEE products. However, due to data paucity in the specific quantities present in these categories, further research and quantitative chemical analysis of these products would be required to establish technology metal content and reliably include these products within the material flow analysis (MFA).

The technology readiness level (TRL) of nascent, current state of the art, pilot plants and proprietary technology solutions (the means) was evaluated as was their potential for deployment in UK WEEE recycling to increase TM recovery rates. The processes and strategies for TM recovery outlined in this report are transferable across other WEEE categories and are suitable to unlock this additional unquantified value present in other products within the UK waste stream. Therefore, the informed results in this report should be considered as a minimum value of the UK opportunity.

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[3] https://www.gov.uk/government/statistical-data-sets/waste-electrical-and-electronic-equipment-weee-in-the-uk

## 1.3 Results

The recycling sector and material recovery technologies tend to focus on precious metal (PM) rich waste streams, which may be worth up to £126.5m (£119.7m of gold and £6.8m of silver; see Table 1) and disregards the other 'critical' elements (estimated to be worth £52.38m, made up of £11.37m of OTMs, £9.39m of REMs and £31.62m of PGMs; see Table 1) – hence the need to evaluate nascent and developed technology solutions where the selectivity of material recovery can target TMs to enhance recovery rates and therefore help recover these materials for the UK economy.

Within the 123,901 tonnes of PCs, laptops, mobile phones, tablets, televisions (TVs), monitors and lighting WEEE formally collected for recycling in the UK each year, 378.91t of TMs are 'lost' in processing (e.g., in the shredding dust) or exported for treatment each year (and either lost or recovered). These have a value of ~£146.84m to the UK economy with a carbon footprint equivalent of mining for these materials of 128,666t$CO_2$e.

The total mass (tonnes), value (£millions) and carbon footprint equivalent of mining for these materials (tonnes $CO_2$e) of technology metals (TMs) present in PCs, laptops, tablets, TVs, monitors, smart phones and lighting sent for recycling in the UK in 2017 is shown below (Table 1).

*Table 1: Total mass, value and carbon footprint of technology metals (TMs) present in PCs, laptops, tablets, TVs, monitors, smartphones and lighting sent for recycling in the UK in 2017.*

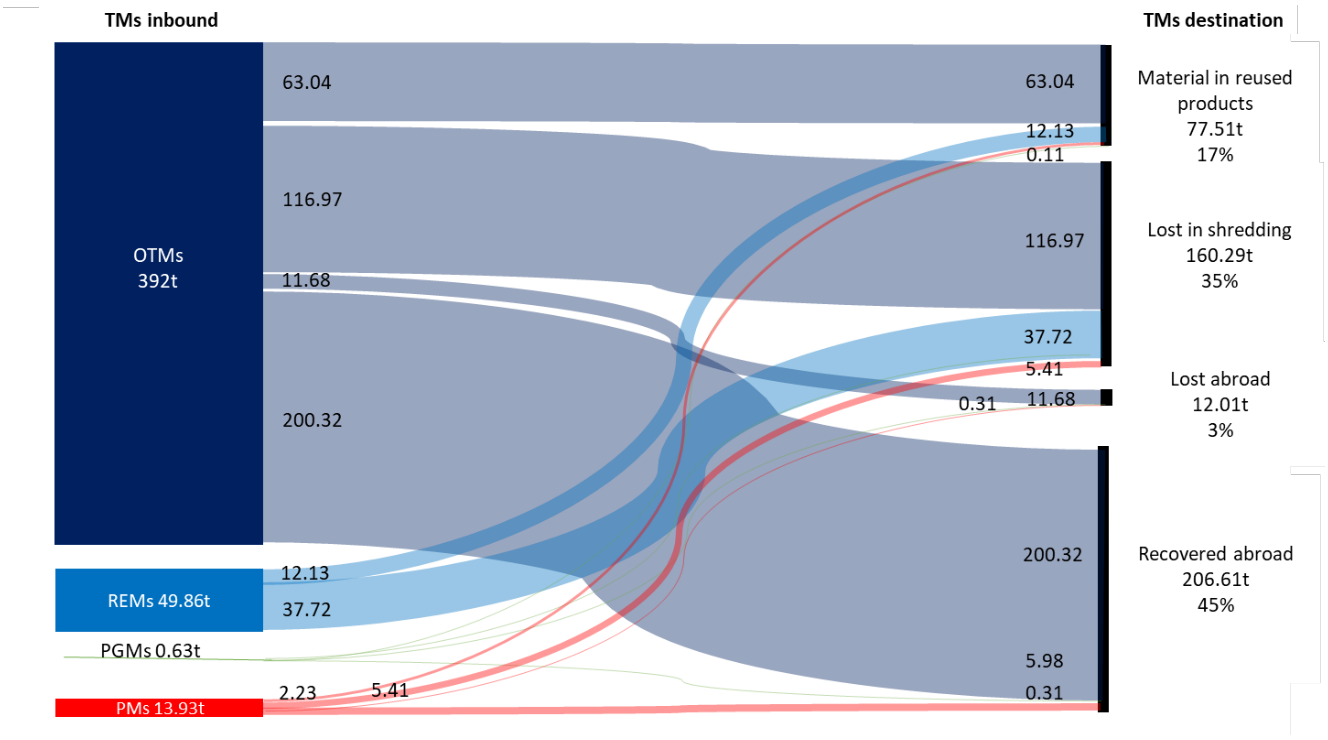
| Element | Mass (tonnes) | Value (£m) | Carbon footprint – GWP100a ($tCO_2e$) |
|---|---|---|---|
| OTMs | | | |
| Antimony | 43.55 | 0.20 | 381.96 |
| Beryllium | 0.02 | 0.01 | 2.37 |
| Cobalt | 0.07 | 0.0002 | 0.79 |
| Gallium | 0.35 | 0.04 | 53.51 |
| Hafnium | 2.84 | 1.86 | 105.26 |
| Indium | 6.34 | 1.05 | 836.29 |
| Nickel | 25.29 | 0.29 | 343.99 |
| Niobium | 0.67 | 0.04 | 24.77 |
| Silicon | 16.17 | 3.05 | 48.52 |
| Tantalum | 4.13 | 0.49 | 1,197.50 |
| Tin | 291.27 | 4.14 | 3,058.30 |
| Titanium | 0.71 | 0.01 | 22.75 |
| Vanadium | 0.60 | 0.19 | 136.97 |
| **Sub total** | **392.01** | **11.3702** | **6,212.98** |
| REMs | | | |
| Cerium | 4.31 | 0.01 | 32.78 |
| Dysprosium | 0.27 | 0.07 | 15.81 |
| Europium | 2.30 | 0.06 | 907.15 |
| Gadolinium | 0.47 | 0.01 | 22.12 |
| Lanthanum | 9.47 | 0.03 | 156.27 |
| Neodymium | 5.91 | 0.31 | 163.13 |
| Praseodymium | 0.84 | 6.82 | 21.73 |
| Terbium | 1.94 | 1.44 | 575.28 |
| Yttrium | 24.35 | 0.64 | 367.63 |
| **Sub total** | **49.86** | **9.39** | **2,261.9** |
| PGMs | | | |
| Palladium | 0.48 | 28.41 | 6,362.91 |
| Platinum | 0.15 | 3.21 | 10,453.47 |
| **Sub total** | **0.63** | **31.62** | **16,816.38** |
| PMs | | | |
| Gold | 2.53 | 119.69 | 124,989.28 |
| Silver | 11.40 | 6.81 | 6,078.50 |
| **Sub total** | **13.93** | **126.5** | **131,067.8** |
| **Total** | **456.42** | **178.88** | **156,359.04** |

In Table 1, OTMs refer to 'Other Technology Metals', REMs are 'Rare Earth Metals', PGMs are 'Platinum Group Metals' and PMs are 'Precious Metals'. See glossary for the materials contained within each of these groups.

### *1.3.1 Technology Metals Flows by Mass*

The fate of technology metals by weight contained within the selected WEEE (PCs, laptops, tablets, TVs, monitors, mobile phones and lighting products) is plotted as a Sankey diagram (Figure 1). Reuse of products originally sent for recycling retains 17% of the contained technology metals (~78 t) within the UK economy. It is estimated that 160 t (35%) of the TMs are dissipated and lost during shredding of WEEE in automated pre-processing within the UK. Less than half of the TMs contained in this WEEE (48%, ~218.6 t) are retained in the output fractions of pre-processing which are exported for recovery with ~206.6 t recovered and ~12 t lost during this recovery process.

*Figure 1: Mass flows of technology metals in assessed WEEE, grouped as precious metals (PMs), platinum group metals (PGMs), rare earth metals (REMs) and other technology metals (OTMs).*

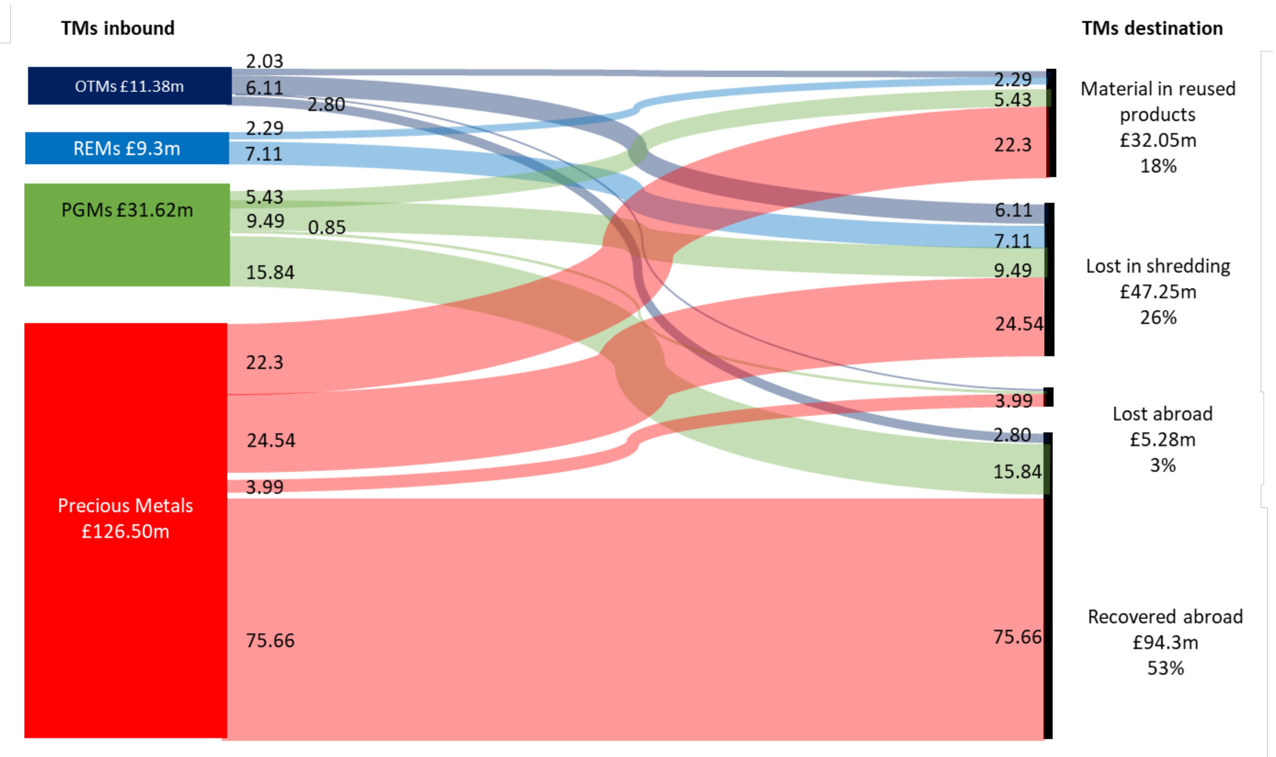


### *1.3.2 Technology Metals Flows by Material Value*

The economic (material) value (Figure 2) associated with the mass flow of TMs (Figure 3) shows the dominance of precious metals (PMs) which, despite having a low relative mass still account for ~71% of the economic value (£126.5m). Platinum Group Metals (PGMs) account for ~£32m (18%), Other Technology Metals (OTMs) account for ~£11m (6%) and Rare Earth Metals (REMs) ~£9m (5%).

The value of TMs currently retained in products that are reused instead of being recycled is approximately 18% of the value inherent in the WEEE collected in the UK. Meanwhile, £47 million worth of TMs are lost during UK pre-processing activities (e.g., mechanical shredding operations), and ~£100m worth of contained technology metals are exported for recovery, or 56% of the total inherent value of TMs in this WEEE. Of the exported WEEE 53% of the overall value is recovered primarily as PMs and PGMs, with the remaining 3% being lost during recovery processes.

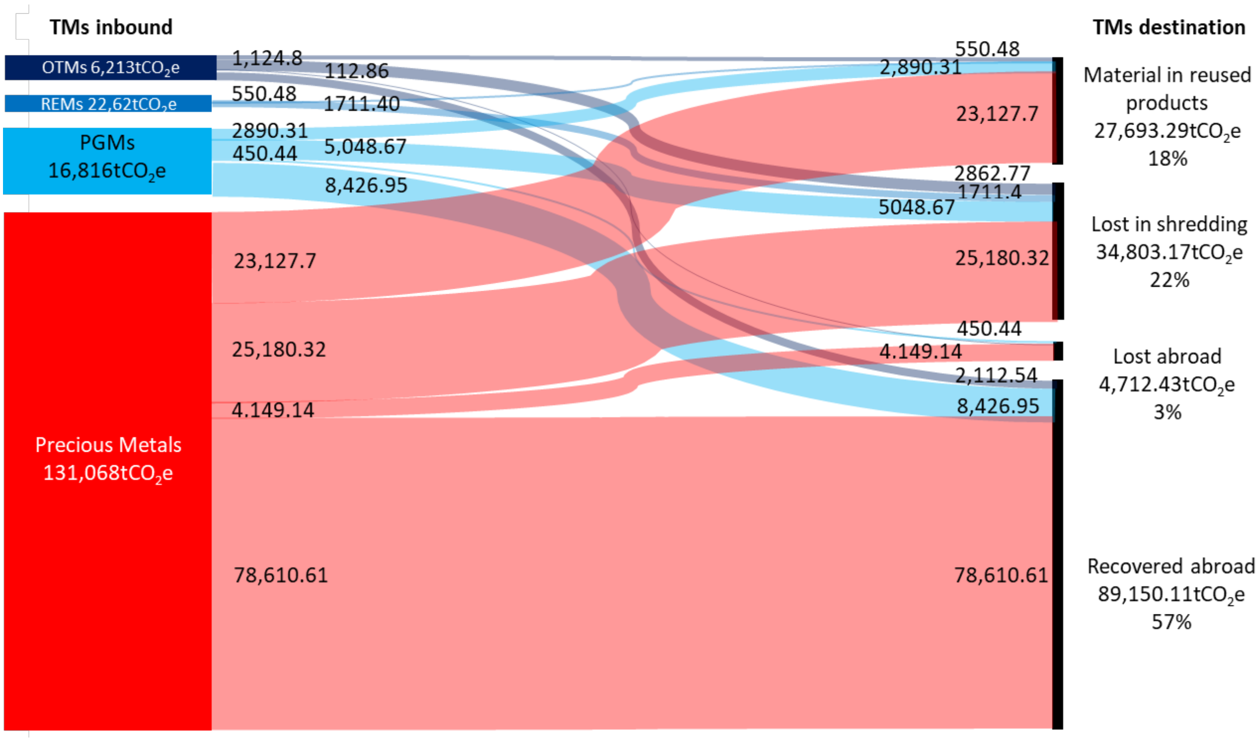
*Figure 2: Value flows of technology metals in assessed WEEE, grouped as precious metals (PMs), platinum group metals (PGMs), rare earth metals (REMs) and other technology metals (OTMs).*



### 1.3.3 Technology Metals Flows by Environmental Impact

The environmental impact (Figure 3) associated with the mass flow of TMs (Figure 1) is estimated using each TM's global warming potential (GWP) expressed as carbon dioxide equivalent ($CO_2e$) from primary production – i.e., the $CO_2$ emitted whilst extracting the same amount of these technology metals from ore. The dominant environmental impact is assigned to PMs, comprising ~ 84% of the total carbon footprint of the contained technology metals. PGMs account for 11%, OTMs 4% and REMs 1% of the environmental impact. Figure 3 also demonstrates how the $CO_2e$ of the TMs is split by their destinations i.e., that the equivalent of 25,180.32 tonnes of $CO_2e$ from PMs is lost in shredding.

*Figure 3: Flows of embodied carbon due to primary production of technology metals in assessed WEEE, grouped as precious metals (PMs), platinum group metals (PGMs), rare earth metals (REMs) and other technology metals (OTMs).*



### 1.3.4 *Summary of TM losses and potential recovery*

Pre-processing losses of TMs, such as through losses in shredding or within the slag during smelting processes (lost abroad), result in high levels of dissipation and therefore significant losses of TMs. Preventing these losses, in addition to if overseas recovery is instead moved to the UK, gives the potential to recover up to 378.9t (160.29t +12.01t + 206.61t - Figure 1) of technology metals from WEEE in the UK, with a market value of £147.83 and a carbon footprint equivalent of 128,666tCO$_2$e.

Based upon the mass of materials lost in shredding, along with those both lost and recovered abroad (see Table 13), the following is a short list of some of the TMs that could be recovered, utilising recovery technologies that appear to be commercially available (covered later in this report), and assuming a conservative 75% recovery efficiency:

- The 82.5kg of platinum recovered could help produce 11,785 new catalytic converters[4];
- The 1.56t of gold recovered could help produce 487,500 new wedding rings[5];
- The 7.72t of silver recovered could help produce 1.93m rings[6]; and
- The 3.3t of neodymium recovered could help produce 2,661 new wind turbines[7].

## 1.4 Recovery Technologies

The actual level of TM recovery is dependent upon numerous factors, particularly the concentration of TMs in WEEE (yield quality) as well as operational factors, including the pre-processing efficiency and losses of TMs therein in mechanical processing (comminution) shredding dust. Other important factors include throughput, yield and thermodynamic limitations to recovery efficiency.

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[4] https://www.specialtymetals.com/blog/2018/3/12/how-much-platinum-is-in-a-catalytic-converter

[5] Based upon 22 carat 3.5 g gold ring

[6] Based upon a weight of 4g per ring.

[7] 1.3MW turbines https://www.wrap.org.uk/sites/files/wrap/Wind%20Turbine%20Magnet%20Study.pdf

As is demonstrated (Figure 1), over 379 tonnes of TMs, including precious metals (PMs) such as gold, silver and Platinum Group Metals (PGMS) including palladium, are currently lost through existing recycling systems or exported for treatment.

The current commercially available TM recovery, extraction and recycling plants, such as those available from EMAK, Enviroleach and Mint Innovation, focus on the recovery of the PMs, PGMs, tin and copper from the WEEE (although copper was not covered within this research).

The analysis of the weight and value of the PMs, PGMs and tin currently lost in shredding, lost abroad during recovery processes and recovered abroad annually within the products covered by this report together total 258.09t (11.71t of PMs, 0.52t of PGMs and 245.88t of tin) with a value of £133.86m (£104.19m of PMs, £26.19m of PGMs and £3.48m of tin). See section 7 for more details.

Therefore, based upon a conservative recovery and recycling rate of 75% of this mass of TMs, a total of 204.5t of PMs, PGMs and tin, worth £100.40m, could potentially be recovered and recycled in the UK using currently commercially available TM recovery, extraction and recycling plants. Due to the energy intensive processes associated with raw material extraction for primary material, the amount of $CO_2$ that mining for these same TMs would emit is estimated to be ~96,499 tonnes of $CO_2e$, though the $CO_2$ emitted by these advanced recovery processes themselves is not known.

In order to recycle the remaining 120.81t of TMs, with a value of approximately £12.95m, that are lost during shredding, lost abroad and recovered abroad, additional processing of the WEEE would be required. Such additional processing could take the form of sequential leaching, which would enable more selective extraction of metals present to enable separate recovery. This process could use the current leaching acids, ionic liquids (ILs), deep eutectic solvents (DES) or biosorption (use of fungi/biological materials to extract the TMs). Biosorption in particular will be useful for extracting low concentrations of TMs in solution.

Once the TMs are in solution, they can then be recovered using various techniques, including electrowinning (which involves the electrodeposition of the TMs onto a cathode for recovery), cementation (precipitation of the metal from solution) or ion exchange (which uses a solid polymeric ion exchange resin to capture the TMs from the solution).

Additional value would also be recovered from the copper present in the WEEE, though again, this was not the focus of the research.

Furthermore, a turnkey development of existing recovery plants could be extended to increase selectivity of an extended range materials, such as copper, aluminium and steel, as well as TMs (e.g., Indium, Cobalt and Gallium) recovery using hydrometallurgical processes. In the pre-sorting of the materials the aluminium and steel can be easily segregated using existing technology.

### *1.4.1 Pre-processing technologies*

Losses in traditional recovery processes, such as pyrometallurgy (extraction and purification of metals via the application of heat followed by electrorefining which uses electrolysis to separate the metals) must be overcome with new approaches to recovery if the UK is to implement domestic infrastructure to increase secondary supply of technology metals from WEEE.

Installation of alternative pre-processing to mechanical shredding for the separation of TM-rich products and components is an important part in the efficacy of TM recovery technologies.

Alternative approaches to shredding for use in automated pre-processing, particularly electrohydraulic fragmentation (EHF) represent a high technology readiness level (TRL) alternative which avoids dissipation of technology metals, enabling cost-effective pre-processing, which therefore delivers greater quantities of technology metals in output fractions for subsequent recovery. The removal of solder and components from the PCBs using hydrometallurgy or thermal desoldering can also minimise the need for shredding and significantly reduce TM losses. Other approaches to removing polymeric components from WEEE components, such as supercritical water treatment, steam gasification or pyrolysis may also enable access to contained TMs for recovery without incurring the dissipative losses observed in shredding. If shredding is unavoidable, then using coarser grinding with control of rotor speed will limit technology metal dissipation in the shredding process.

### 1.4.2 Extraction and Recovery Technologies

Leaching of technology metals from pre-processing outputs using hydrometallurgical methods followed by recovery using electrowinning is considered the most promising method for high recovery rates of materials. Electrolytic refinement has a high TRL and is already widely adopted in recycling.

Proprietary systems for hydrometallurgical extraction of precious metals (e.g., using acid solutions to extract TMs from printed circuit boards (PCBs)) are already available, and in operation around the world. Examples include the systems of EMAK, Mint Innovation, and Enviroleach. Adoption of such technology for processing PCBs and lighting in the UK would enable domestic recovery, allowing the pre-processing outputs and their contained technology metals to be retained within the UK. The small scale of these plants compared to existing pyrometallurgical facilities and relatively low cost make them suitable for regional recovery hubs in the UK, potentially sited close to major WEEE recyclers and/or industries that can make use of recovered materials.

A typical EMAK plant, which is claimed to be capable of processing 1 tonne of TM-bearing WEEE per day (and therefore process approximately 250t per year), operational in the UK could treat ~5.8% of the estimated 4,500t of PCBs from the laptops, mobile phones, computers, TVs and monitors sent for recycling in the UK annually.

As well as acid solutions (such as those used by EMAK) being used to extract the TMs, ionic liquids (ILs), deep eutectic solvents (DES) and bioleaching (use of fungi/biological materials to extract the TMs) are also all proving particularly promising. The latter will be useful for selectively extracting low concentrations of technology metals in solution. Once the TMs are in solution, they can be extracted using electrowinning, which involves the electrodeposition of the TMs onto a cathode for recovery, or bacteria can be used to extract the TMs from solution, also known as 'Biosorption'.

Other factors affecting TM recovery yield include the efficiency of the leaching processes, concentration of the acids and selectivity, ionic liquids (ILs) and deep eutectic solvents (DES) used and their selectivity, length of time the parts are exposed to the leaching agents (also known as 'dwell time') and overall material throughput rate.

### 1.4.3 Cost of TM recovery plants

In general, setup and running costs for existing TM recovery facilities were not available. However, a capital expenditure of ~$2m to $2.5m (£1.4m to £1.8m) is thought to be indicative for a fully operational EMAK recovery and recycling plant (recovering PMs, PGMs, tin and copper) with a capacity to treat 1 tonne of PCBs a day (~250t per annum). Further investment will be required in the developing technologies identified by this research to recover the full range of TMs. At least 56% of the of £133.86m value of TMs in the WEEE covered in this research resides in the PCBs and an estimated 18 such plants would be needed to treat the estimated 4,500 tonnes of precious metal-bearing PCBs extracted from the computers, laptops, tablets, TVs and smart phones sent for recycling in the UK each year, with an estimated value in excess of £75m. This would require an approximate investment of ~£25m - £32m in TM recovery plant. Based upon approximate operational costs (as provided by EMAK) for chemicals, electricity and labour of £1,077 per tonne in a 1t a day EMAK plant, the cost to treat the 4,500t would be an estimated additional cost of £4.85m per year. Note that these costs do not include the cost of purchasing the WEEE to feed the plants, permitting costs and various other relevant overheads.

Although this would require significant investment by the WEEE recyclers or others, operational plants, such as that manufactured by EMAK (Table 22), are believed to be commercially viable where market conditions are favourable (e.g., gold bullion price).

As noted, additional specialised plant would be required to treat the other types of TM-bearing WEEE, such as lighting, display equipment and computer hard drives, in particular to focus on the recovery of the TMs beyond the PMs, PGMs and tin.

## 1.5 Summary

The research clearly shows the potential of developed and nascent technologies to recover increasing levels of TMs from WEEE. The extension of these technologies to the pre-processing and extraction processing of TMs from WEEE is highly dependent on economic factors, including geopolitics and market dynamics for material pricing. However, the criticality of materials, for which many ubiquitous technologies and emerging green technologies now depend, will increase, alongside the recognition of the importance of security of supply at a national level. The high environmental impact of producing prime materials will necessitate further development and deployment of TM recovery technologies.

This report considers the technologies that are suitable to augment recycling of WEEE arising today. Further validation of the various recovery technologies for distributed recovery, and evaluation of scaled up processes for those technologies currently at low TRL would further validate the potential outlined in this report. Moreover, further detailed analysis of TM presence and quantities within WEEE (e.g., X-Ray Fluorescence (XRF) and Inductively Coupled Plasma Mass spectrometry (ICP)) could be used to focus the recovery of TMs present in specific parts of WEEE assemblies and components.

Over the longer term, eco-design of products to facilitate the Circular Economy (CE) will require manufacturers to consider end of life (EoL) and facilitate ease of access to core assemblies that contain PGMs, REMs, PMs and other technology metals. This could enhance further levels of recovery, simplifying recycling strategies.

Furthermore, alternative approaches to automation of product disassembly could improve the efficiency and cost-benefit of pre-processing. This will reduce cut-off-grades, enabling greater quantities of WEEE containing TM-rich PCBs, for example, to be diverted from shredding, to prevent their dissipation and ensure they are delivered to recovery processes. However, this is a longer-term solution to materials criticality.

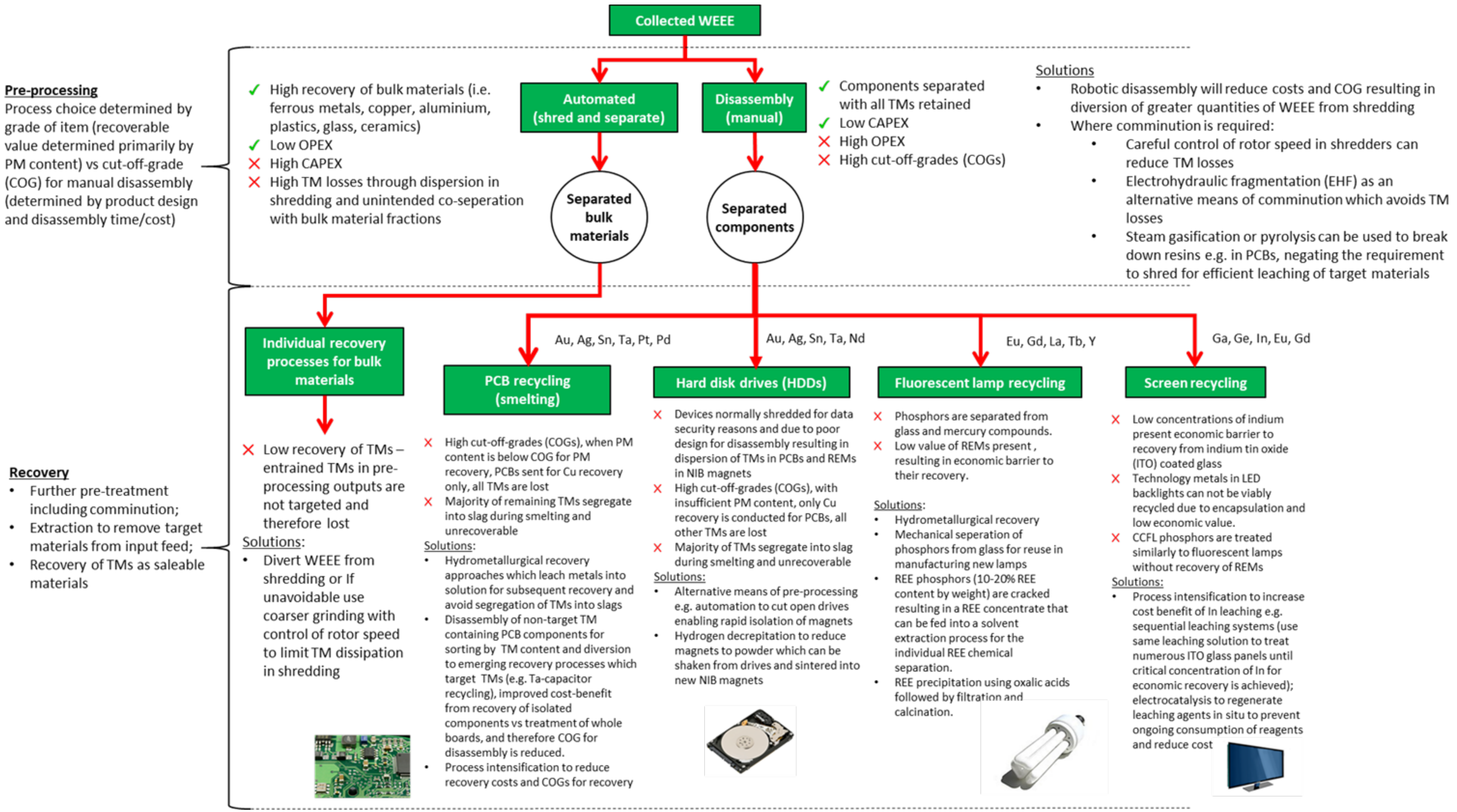
With the development of the national waste tracking system well-underway by a number of organisations on behalf of DEFRA, and the establishment of the National Materials Datahub, the UK is about to undergo a step-change in its ability to monitor flows of WEEE and contained TMs through the economy, and therefore prospect the urban mine to target viable domestic secondary TM sources.

The fate of technology metals in WEEE throughout the recycling process chain with recommended strategies to avoid losses and enhance recovery rates is shown in Figure 4.

A technology roadmap indicating the most promising technologies reviewed in this report, with their TRL levels, is shown in Figure 5. These technologies could be developed further and, as they reach higher TRLs, could potentially become commercialised. If the UK invested in these technologies, a significantly high portion of TMs could be recovered from WEEE. The technologies listed are described in detail throughout this report:

- Sections 2 to 6 gives an overview of TMs and how and where they are used;
- Section 7 identifies the quantities of TMs in WEEE; and
- Sections 9 to 18 review the different TM recovery methods for different types of WEEE.

*Figure 4: The fate of technology metals in WEEE throughout the recycling process chain with recommended strategies to avoid losses and enhance recovery rates.*



**Pre-processing**
Process choice determined by grade of item (recoverable value determined primarily by PM content) vs cut-off-grade (COG) for manual disassembly (determined by product design and disassembly time/cost)

Automated (shred and separate):

- ✓ High recovery of bulk materials (i.e. ferrous metals, copper, aluminium, plastics, glass, ceramics)
- ✓ Low OPEX
- ✗ High CAPEX
- ✗ High TM losses through dispersion in shredding and unintended co-seperation with bulk material fractions

Disassembly (manual):

- ✓ Components separated with all TMs retained
- ✓ Low CAPEX
- ✗ High OPEX
- ✗ High cut-off-grades (COGs)

<u>Solutions</u>

- Robotic disassembly will reduce costs and COG resulting in diversion of greater quantities of WEEE from shredding
- Where comminution is required:
    - Careful control of rotor speed in shredders can reduce TM losses
    - Electrohydraulic fragmentation (EHF) as an alternative means of comminution which avoids TM losses
    - Steam gasification or pyrolysis can be used to break down resins e.g. in PCBs, negating the requirement to shred for efficient leaching of target materials

**Recovery**

- Further pre-treatment including comminution;
- Extraction to remove target materials from input feed;
- Recovery of TMs as saleable materials

Individual recovery processes for bulk materials:

- ✗ Low recovery of TMs – entrained TMs in pre-processing outputs are not targeted and therefore lost

<u>Solutions</u>:

- Divert WEEE from shredding or If unavoidable use coarser grinding with control of rotor speed to limit TM dissipation in shredding

PCB recycling (smelting):

- ✗ High cut-off-grades (COGs), when PM content is below COG for PM recovery, PCBs sent for Cu recovery only, all TMs are lost
- ✗ Majority of remaining TMs segregate into slag during smelting and unrecoverable

<u>Solutions</u>:

- Hydrometallurgical recovery approaches which leach metals into solution for subsequent recovery and avoid segregation of TMs into slags
- Disassembly of non-target TM containing PCB components for sorting by TM content and diversion to emerging recovery processes which target TMs (e.g. Ta-capacitor recycling), improved cost-benefit from recovery of isolated components vs treatment of whole boards, and therefore COG for disassembly is reduced.
- Process intensification to reduce recovery costs and COGs for recovery

Hard disk drives (HDDs):

- ✗ Devices normally shredded for data security reasons and due to poor design for disassembly resulting in dispersion of TMs in PCBs and REMs in NIB magnets
- ✗ High cut-off-grades (COGs), with insufficient PM content, only Cu recovery is conducted for PCBs, all other TMs are lost
- ✗ Majority of TMs segregate into slag during smelting and unrecoverable

<u>Solutions:</u>

- Alternative means of pre-processing e.g. automation to cut open drives enabling rapid isolation of magnets
- Hydrogen decrepitation to reduce magnets to powder which can be shaken from drives and sintered into new NIB magnets

Fluorescent lamp recycling:

- ✗ Phosphors are separated from glass and mercury compounds.
- ✗ Low value of REMs present , resulting in economic barrier to their recovery.

<u>Solutions</u>:

- Hydrometallurgical recovery
- Mechanical seperation of phosphors from glass for reuse in manufacturing new lamps
- REE phosphors (10-20% REE content by weight) are cracked resulting in a REE concentrate that can be fed into a solvent extraction process for the individual REE chemical separation.
- REE precipitation using oxalic acids followed by filtration and calcination.

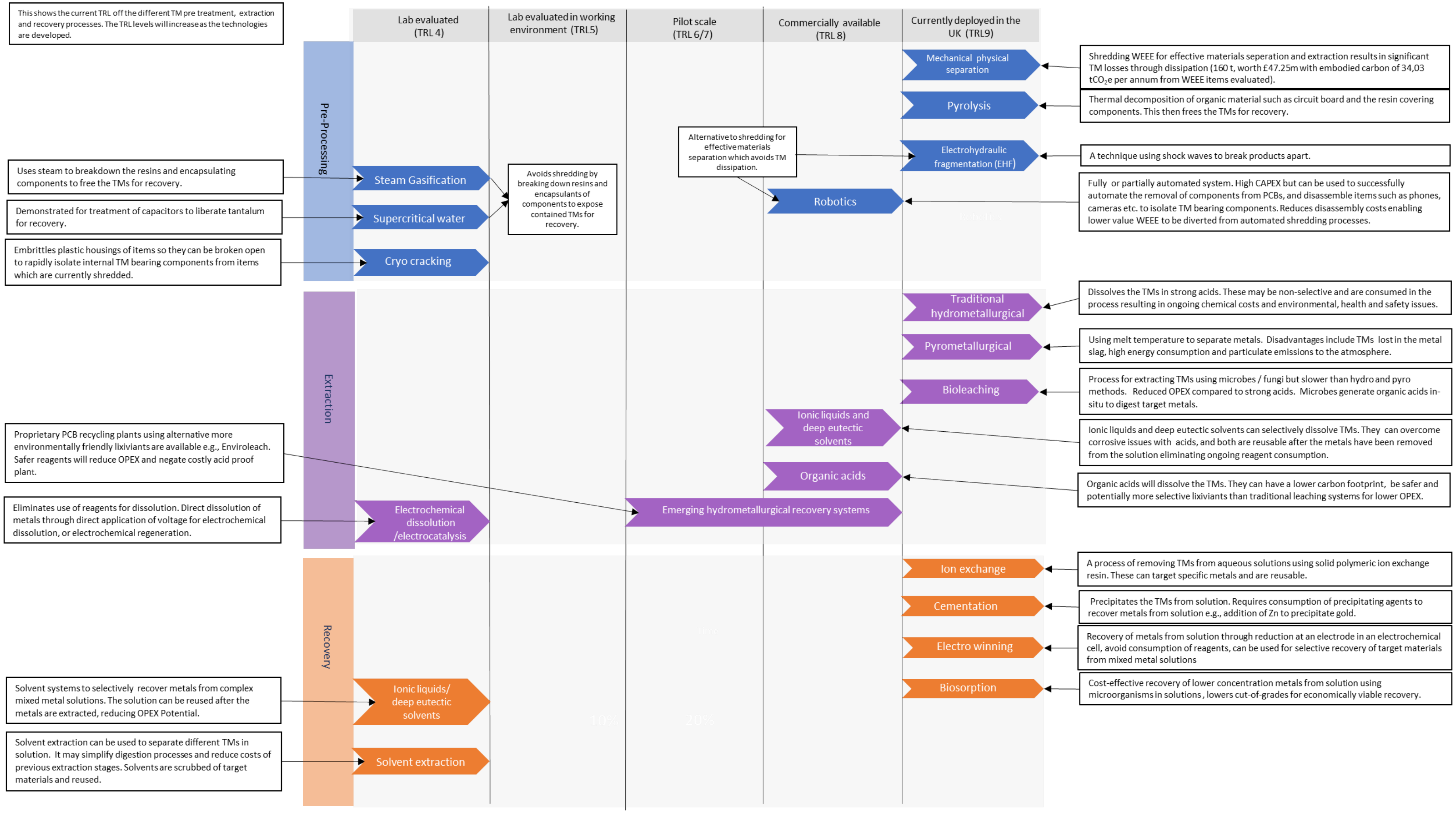
Screen recycling:

- ✗ Low concentrations of indium present economic barrier to recovery from indium tin oxide (ITO) coated glass
- ✗ Technology metals in LED backlights can not be viably recycled due to encapsulation and low economic value.
- ✗ CCFL phosphors are treated similarly to fluorescent lamps without recovery of REMs

<u>Solutions:</u>

- Process intensification to increase cost benefit of In leaching e.g. sequential leaching systems (use same leaching solution to treat numerous ITO glass panels until critical concentration of In for economic recovery is achieved); electrocatalysis to regenerate leaching agents in situ to prevent ongoing consumption of reagents and reduce cost

*Figure 5: Technology Metal (TM) recovery technology roadmap indicating the technology identified, and the stages to WEEE recycling to which they are applicable.*



This shows the current TRL off the different TM pre treatment, extraction and recovery processes. The TRL levels will increase as the technologies are developed.

Uses steam to breakdown the resins and encapsulating components to free the TMs for recovery.

Demonstrated for treatment of capacitors to liberate tantalum for recovery.

Embrittles plastic housings of items so they can be broken open to rapidly isolate internal TM bearing components from items which are currently shredded.

Avoids shredding by breaking down resins and encapsulants of components to expose contained TMs for recovery.

Alternative to shredding for effective materials separation which avoids TM dissipation.

Proprietary PCB recycling plants using alternative more environmentally friendly lixiviants are available e.g., Enviroleach. Safer reagents will reduce OPEX and negate costly acid proof plant.

Eliminates use of reagents for dissolution. Direct dissolution of metals through direct application of voltage for electrochemical dissolution, or electrochemical regeneration.

Solvent systems to selectively recover metals from complex mixed metal solutions. The solution can be reused after the metals are extracted, reducing OPEX Potential.

Solvent extraction can be used to separate different TMs in solution. It may simplify digestion processes and reduce costs of previous extraction stages. Solvents are scrubbed of target materials and reused.

Shredding WEEE for effective materials seperation and extraction results in significant TM losses through dissipation (160 t, worth £47.25m with embodied carbon of 34,03 $tCO_2e$ per annum from WEEE items evaluated).

Thermal decomposition of organic material such as circuit board and the resin covering components. This then frees the TMs for recovery.

A technique using shock waves to break products apart.

Fully or partially automated system. High CAPEX but can be used to successfully automate the removal of components from PCBs, and disassemble items such as phones, cameras etc. to isolate TM bearing components. Reduces disassembly costs enabling lower value WEEE to be diverted from automated shredding processes.

Dissolves the TMs in strong acids. These may be non-selective and are consumed in the process resulting in ongoing chemical costs and environmental, health and safety issues.

Using melt temperature to separate metals. Disadvantages include TMs lost in the metal slag, high energy consumption and particulate emissions to the atmosphere.

Process for extracting TMs using microbes / fungi but slower than hydro and pyro methods. Reduced OPEX compared to strong acids. Microbes generate organic acids in-situ to digest target metals.

Ionic liquids and deep eutectic solvents can selectively dissolve TMs. They can overcome corrosive issues with acids, and both are reusable after the metals have been removed from the solution eliminating ongoing reagent consumption.

Organic acids will dissolve the TMs. They can have a lower carbon footprint, be safer and potentially more selective lixiviants than traditional leaching systems for lower OPEX.

A process of removing TMs from aqueous solutions using solid polymeric ion exchange resin. These can target specific metals and are reusable.

Precipitates the TMs from solution. Requires consumption of precipitating agents to recover metals from solution e.g., addition of Zn to precipitate gold.

Recovery of metals from solution through reduction at an electrode in an electrochemical cell, avoid consumption of reagents, can be used for selective recovery of target materials from mixed metal solutions

Cost-effective recovery of lower concentration metals from solution using microorganisms in solutions, lowers cut-of-grades for economically viable recovery.

# 2 Background

## 2.1 Status of TM Recovery from UK WEEE and Scale of the Opportunity

It is almost a decade since government first set strategy to increase domestic Technology Metals (TM) supplies through recovery from waste electrical and electronic equipment (WEEE) to mitigate materials criticality issues for UK industry. TMs are all defined as any "rare" metal or a metal that is in demand, available, and used for the purpose of furthering technology and engineered systems. These are CRM metals, Platinum group metals (PGMs), Precious Metals (PMs), Nickel and Tin. Further details are given in section 5.

Despite this, TM recycling rates remain low generally, and barely exceed 50% even for the most economically valuable e.g., gold (Au) and platinum group metals (PGMs). Currently less than 1% of all rare earth metals (REMs) are recycled as the vast majority is lost in dust generated by the pre-processing of WEEE or is lost in the slag during pyrometallurgical metal recovery. In the UK there is very little in the way of TM recovery from WEEE, apart from the pre-processing of printed circuit boards (PCBs) and limited small-scale precious metal (PM) and platinum group metal (PGM) recovery.

Even though WEEE legislation has resulted in an infrastructure for collection and treatment of WEEE, the rate of recovery of TMs from this waste stream remains low. This is due to factors such as collection rates of WEEE being low compared with that generated overall for post-consumer products, dissipation of TMs during recycling, lack of available recovery processes that target the wide range of technology metals present within WEEE and economic barriers to recovery presented by the relatively low economic value that can be recouped from recovery of trace amounts of TMs within individual products, despite the net value afforded to economies by access to technology metals. These issues must be addressed to avoid permanent loss of the vast quantities of TMs present in collected WEEE in the future.

Without this, growing UK demand for TMs will necessitate further primary resource development resulting in destruction of precious natural capital and environmental degradation, leaving the UK economy increasingly vulnerable to global resource security issues. To-date, the UK has been entirely reliant upon foreign recovery infrastructure for TM recycling, with efforts to mitigate materials criticality driven by the European Commission at an EU level. With the prospect of becoming an 'economic competitor' to the EU, the UK may be more vulnerable than ever to resource criticality issues.

However, the means to overcome these barriers have begun to emerge and an opportunity now exists for the UK to establish a circular economy of its own for TMs, capitalising on its high per capita rates of EEE consumption and WEEE generation to develop secondary TMs supplies for reliant domestic industries (Material Focus, 2020a).

Recently, Material Focus released findings which estimated that UK WEEE generation in 2017 was ~1.4 Mt (Material Focus, 2020a). In 2019, ~499,000 t of consumer (household) WEEE was collected and treated in the UK, according to government data reported for the WEEE Regulations (Environment Agency, 2020), and the Global E-waste Monitor 2020 reported UK WEEE generation to be ~1.6 Mt in 2019 or 23.9 kg/cap, one of the highest per capita WEEE generation rates in Europe and the World (Forti *et al.*, 2020). This highlights the potential of domestic TM recovery from WEEE as a means to mitigate materials criticality issues for UK industry, improve environmental impacts of manufacturing, and create jobs in new industry to facilitate recovery and cycling of TMs back into the UK manufacturing economy from End of Life (EoL) products, or indeed in manufacturing sectors which are able to benefit from access to domestic sources of TMs.

The Material Focus report also highlighted issues with different disposal fates and stockpiling, which prevents WEEE from being collected for reuse and recycling and therefore limits the size of the secondary TMs reserve and TM recycling potential from UK WEEE. If acted upon, recommendations made to increase capture rates of WEEE for reuse and recycling will enable access to greater quantities of domestic WEEE for EoL treatment, and potentially access to the contained technology metals (Material Focus, 2020a).

The UK economy is highly reliant upon imports of TMs, demand for which is forecast to increase considerably into the future. WEEE has been identified as one of the fastest growing waste streams and due to the extensive use of TMs in electronics, WEEE is a rich potential domestic secondary source of TMs for the UK economy.

Accelerating technology innovation cycles and growth in emerging economies has steadily increased raw materials demand with predictions showing that global resource use could double between 2010 and 2030. To meet climate and energy targets in the EU, critical raw material (CRM) demand, in particular, is expected to increase by a factor of 20 for certain materials (EC JRC, 2016), and access to TMs may limit technology deployment and our ability to combat climate change. TMs enable essential functionality in a wide range of products and technology applications.

# 3 Policy Context

The rare earths trade dispute in 2010 resulting from reductions in export quotas for rare earth metals (REMs), molybdenum (Mo) and tungsten (W) by China sparked global efforts to ensure access to the most important materials for economies. This began with a wave of resource criticality assessments to determine which raw materials are of greatest economic importance to economies which rely on them and face highest risks to future supply.

The EU Raw Materials Initiative identified a list of critical raw materials at EU level, in close cooperation with Member States and stakeholders. A report was released in 2011 which puts forward a concept of "criticality", where the raw material is labelled critical when the risks of supply shortage and their impacts on the economy are higher than compared with most of the other raw materials (EC, 2014). This report concluded that the 14 raw materials were to be considered "critical" due to their high relative economic importance and high relative supply risk. This was revised to 20 CRMs in 2014 (EC, 2014), and 27 CRMs in 2017 (EC, 2017). The most recent evaluation in 2020 concluded that 30 materials are critical for the EU (the 'EU30') (Figure 6, Table 2), following an investigation of import reliance, economic significance, main global producers of these materials and supply risks associated with those supplies.

The primary purpose of CRM lists such as these are to identify the raw materials with a high supply-risk and a high economic importance to which reliable and unhindered access is a concern for European industry and value chains, in order to inform innovation and attempts at mitigation of primary supply issues. Although the list includes numerous elements which are not 'technology metals', technology metals are often found to form the core of criticality assessments globally and for different sectors, despite varying priorities for different governments and organisations conducting these assessments, owing to the importance of technology metals for advanced and emerging technologies in all aspects of our lives and across all countries and sectors.

*Figure 6: Results of the most recent EC materials criticality evaluation depicting the 'EU30' critical raw materials in red in the top right of the plot as those with highest supply risk and economic importance (EC, 2020).*

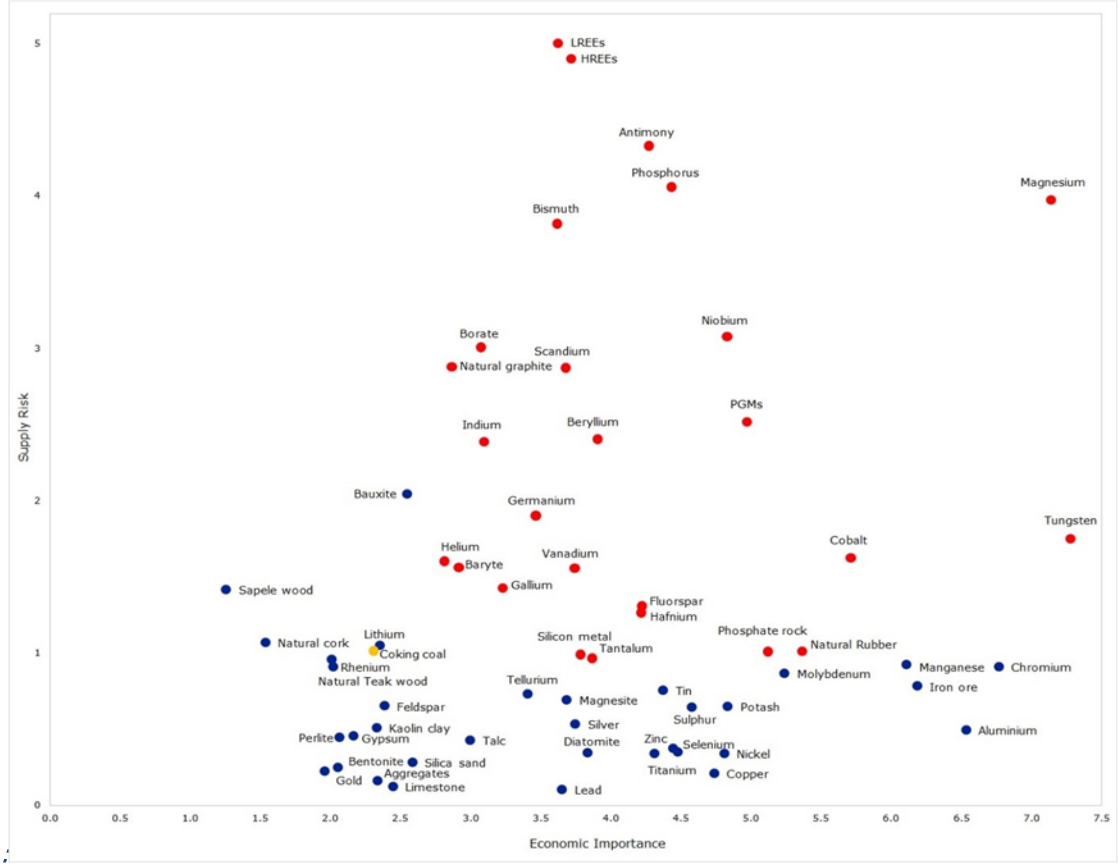


*Table 2: The 2020 EU list of critical raw materials (CRMs) – the 'EU30' (EC, 2020), TMs are emboldened.*

| | | | | |
|---|---|---|---|---|
| **Antimony (Sb)** | **Cobalt (Co)** | **HREEs**[1] | Natural rubber | **Silicon metal (Si)** |
| Baryte | Coking coal | **Indium (In)** | **Niobium (Nb)** | |
| Bauxite | Fluorspar | **Lithium (Li)** | **PGMs**[4] | **Tantalum (Ta)** |
| **Beryllium (Be)** | **Gallium (Ga)** | **LREEs**[2] | Phosphate rock | **Titanium (Ti)** |
| **Bismuth (Bi)** | **Germanium (Ge)** | Magnesium (Mg) | Phosphorous (P) | **Tungsten (W)** |
| Borates | **Hafnium (Hf)** | Natural graphite[3] | **Scandium (Sc)** | **Vanadium (V)** |

*[1] The heavy rare earth elements (HREEs) are yttrium (Y), europium (Eu), gadolinium (Gd), terbium (Tb), dysprosium (Dy), holmium (Ho), erbium (Er), thulium (Tm), ytterbium (Yb), and lutetium (Lu); [2] The light rare earth elements (LREEs) are scandium (Sc), lanthanum (La), cerium (Ce), praseodymium (Pr), neodymium (Nd), promethium (Pm), and samarium (Sm); [3] Although not a technology metal, this form of carbon is vital for numerous EEE technologies including Li-ion batteries for portable devices and stationary renewable energy storage. [4] platinum group metals (PGMs) include platinum (Pt), palladium (Pd), ruthenium (Ru), iridium (Ir), rhodium (Rh), osmium (Os).*

In response to private sector concerns about availability of CRMs, the Resource Security Action Plan (RSAP) was developed jointly by DEFRA and BIS in 2012 (DEFRA and BIS, 2012). This strategy took the view that the picture of material criticality is temporal due to innovation, technological advancement and changes in world markets impacting upon the factors which determine a material's 'critical' status.

Therefore, the results of all criticality assessments are valid, and that due to the common prevalence of technology metals qualifying as critical in assessments, technology metals are critical for the UK economy.

The current Resources and Waste Strategy for England aims to reinvigorate this plan as the major component of its national resource strategy (DEFRA, 2018). The Clean Growth Strategy seeks to 'green' industry and grow the UK's clean energy and electric transport sectors, amongst other heavily CRM-reliant industries (Weil and Ziemann, 2014; Government, 2017; HM Government, 2017b). However, despite almost a decade passing since government first set its strategy to increase domestic TM supplies through recovery from WEEE, TM recycling rates remain very low, due to various factors, including low collection rates for post-consumer products, dissipation of TMs during recycling, and economic barriers to recovery presented by the relatively low financial value that can be recouped from recovery of trace amounts of TMs within individual products, despite the net value afforded to economies by access to technology metals overall.

In light of this, the UK Government's approach with regard to mitigating criticality of technology metals would be to facilitate business action where there is the greatest scope to reduce risk and environmental impact and to capture value for the UK economy, and that this response would also be crucial to managing supply chain risks resulting from impacts of climate change. This is to be achieved through increased domestic supply by recovery from wastes, including WEEE.

With one of the highest per capita EEE consumption rates and therefore per capita WEEE generation rates in the world, the UK has significant potential to utilise WEEE as a secondary supply of TMs domestically. The RSAP also indicated that >£350 million worth of gold was present in the EEE purchased annually in the UK, and that this information would form the basis of a material flow analysis (MFA) study for WEEE to identify opportunities and risks for business and waste companies in the WEEE sector.

Table 3 shows the materials deemed insecure or at risk by recent reports.

*Table 3: Materials deemed insecure or at risk by recent reports – presented in the Resource Security Action Plan (DEFRA and BIS, 2012).*

| Material | Materials criticality evaluation | | | | | | |
|---|---|---|---|---|---|---|---|
| | EU | TSB | Defra | SEPA (AEA Technology plc, 2010) | STC | BGS* | US |
| Aggregates | | | ✓ | ✓ | | | |
| Antimony | ✓ | ✓ | | | ✓ | ✓ | |
| Beryllium | ✓ | | | | ✓ | | |
| Bismuth | | | | | | ✓ | |
| Bromine | | | | | | ✓ | |
| Chromium | | | | | ✓ | | |
| Cobalt | ✓ | | | ✓ | ✓ | | |
| Copper | | | | ✓ | | | |
| Fish | | | ✓ | ✓ | | | |
| Fluorspar | ✓ | | | | | | |
| Gallium | ✓ | | | | ✓ | | |
| Germanium | ✓ | | | | ✓ | | |
| Gold | | ✓ | | | ✓ | | |
| Graphite | ✓ | | | | | | |
| Hafnium | | | | | ✓ | | |
| Indium | ✓ | | ✓ | ✓ | ✓ | | ✓ |
| Lithium | | | ✓ | ✓ | ✓ | | ✓ |
| Lead | | | | ✓ | | | |
| Magnesium | ✓ | | | | ✓ | | |
| Mercury | | ✓ | | | | ✓ | |
| Nickel | | | | | ✓ | | |
| Niobium | ✓ | | | | ✓ | ✓ | |
| Palm oil | | | | ✓ | | | |
| Phosphorous | | | ✓ | ✓ | | | |
| PGMs | ✓ | ✓ | | | ✓ | ✓ | |
| REMs | ✓ | | ✓ | ✓ | ✓ | ✓ | ✓ |
| Rhenium | | | | | ✓ | | |
| Silver | | ✓ | | | | | |
| Strontium | | ✓ | | | | ✓ | |
| Tantalum | ✓ | | | | ✓ | | |
| Tellurium | | | | | ✓ | | ✓ |
| Thorium | | | | | | ✓ | |
| Timber | | | | ✓ | | | |
| Tin | | ✓ | | ✓ | | | |
| Tungsten | ✓ | | | | | ✓ | |

- Note: the above table provides a full list of materials identified as critical by a range of organisations and reports, which extend beyond the scope of this study.
- *The BGS list was updated in 2015, data from the 2011 list is taken from "*Hislop and Hill, 2011*". (Hislop and Hill, 2011)
- EU: Raw Material Supply Group, chaired by EC 2010, *"Critical raw materials for the EU: report on ad-hoc working group on defining critical raw materials"*

- TSB: Technology Strategy Board: Resource Efficiency Knowledge Transfer Network
- Defra: *"Review of the future resource risks faced by UK business and an assessment of future liability"*
- SEPA: Scottish Environment Protection Agency: "*Raw Materials Critical to the Scottish Economy".*
- STC: Science and Technology Committee, House of Commons. "*Enquiry into strategically important metals*".
- BGS: British Geological Survey, 2001, "*Risk List 2011"*
- US: US Department of Energy 2010*: "Critical Materials Strategy"*

A 'Green' second industrial revolution has been stated by the Prime Minister as the means of economic recovery from the recession caused by the ongoing coronavirus pandemic. Secondary CRM supplies from WEEE will be essential to achieve these ambitions (HM Government, 2017c). The UK and devolved governments recognise the benefits of shifting to a more resource efficient and circular economy as a means of reducing industrial emissions; ensuring ongoing competitiveness, resilience and growth of UK industry. All four nations have committed to enhance environmental protections, recognising the need to accelerate action to address the climate emergency. The recent transposition of the EU Circular Economy Package (CEP) into national legislation provides the policy and regulatory framework to support the UK's transition towards a circular economy. This will keep resources in use as long as possible, extracting maximum value from them, minimizing waste and promoting resource efficiency (DEFRA et al., 2020). This will also build upon the Resources and Wastes strategy for England (HM Government, 2018), the UK Government's 25-year Environment Plan, and the Circular Economy Strategies of the Welsh and Scottish Governments (Scottish Government and Natural Scotland, 2016; Welsh Government, 2019).

The Resources and Wastes strategy for England is supporting further investment and innovation in resource efficiency, working with UK Research and Innovation (UKRI) on their Areas of Research Interest and supporting further investment in resource efficient technologies, including through the Industrial Strategy Challenge Fund.

Development of healthy markets for secondary raw materials and increased uptake by industry is a key component of the UK Industrial Strategy to increase the resilience of UK industry to global resource security issues, reduce emissions (as recycling materials is possible with considerably lower environmental impact than developing primary resources), and create jobs in new industries to facilitate circular flows of materials within the UK's economy (HM Government, 2017a).

It is clear that technology metals are extremely important to the existing economy and underpin ambitions to grow new industries which will rely on ongoing supplies. This issue has been explored in the House of Commons Science and Technology Committee report 'Strategically important metals' (House of Commons Science and Technology Committee, 2012). Contributions by the IOM3 revealed that major UK manufacturing companies rely on technology metals as raw materials, and the Minor Metals Trade Association (MMTA) confirm this in a statement which declared the UK to be "one of the world's leaders of advanced technologies that consume strategic metals".

Wales and Scotland have in the past conducted their own materials criticality evaluations (SEPA, 2011; Harfield, 2020). Wales concluded that many of its economic priority sectors are vulnerable to materials criticality issues. This includes a vast number of organisations within the advanced materials and manufacturing sector including Rolls Royce; and the ICT sector including IQE, a world leader in the manufacturing of advanced semiconductor wafers; as well as the major metals producers such as Tata Steel. The additional reliance of other sectors including pharmaceuticals, aircraft and chemicals organisations was also highlighted. The Scottish report further highlighted the importance of ongoing technology metal supply for the automotive sector, particularly in light of the ongoing transition to EVs, as well as the chemicals sector that requires numerous technology metal-based catalysts to synthesise its products, and also the electronics sector which requires numerous technology metals to manufacture its products.

As discussed, the Government's ambitions to build a green manufacturing sector are underpinned by access to technology metals, an issue that may be more important than ever in light of the 'green recovery' plan to regrow and restructure our economy post COVID. For example, the availability of lithium and other CRMs such as cobalt and graphite will be vital if the UK is to secure inward investment from organisations such as Nissan who have expressed interest in establishing battery manufacturing in the UK to support the EV revolution. Other emerging organisations in this sector, such as Oxford PV, likely to become the first organisation in the world to commercialise the use of perovskite solar cells in tandem photovoltaic (PV) devices, will require access to many of the technology metals that can be recovered from WEEE, including indium for its Indium Tin Oxide (ITO) substrates, and gold and silver for use as contacts in its devices.

A material flow analysis (MFA) study (Parker and Arendorf) investigated flows of CRMs contained in UK WEEE through recycling process chains and concluded that no CRMs are recovered from WEEE in the UK, owing to the lack of domestic recovery infrastructure, and high losses of CRMs through dissipation in pre-processing carried out within the UK. However, the means to overcome these barriers have begun to emerge and an opportunity now exists for the UK to establish a circular economy of its own for CRMs (and TMs), capitalising on a high per capita rate of EEE consumption and WEEE generation to develop secondary TM supplies for reliant domestic industries.

With the development of the national waste tracking system well-underway on behalf of DEFRA, and the establishment of the National Materials Datahub, the UK is about to undergo a step-change in its ability to monitor flows of WEEE and contained TMs through the economy, and therefore prospect the urban mine to target viable domestic secondary TM sources.

# 4 The Importance of Technology Metals and their Critical Status

As noted, there is increasing global concern over the security of global supply of a wide range of strategically important TMs, which are essential to the functionality of modern technologies, albeit in most cases used in trace amounts within products. Substitution in many applications is not feasible at this time so measures to mitigate supply risks are urgently required, particularly by resource-hungry nations whose economies rely heavily upon imports, such as the UK. The global availability of TMs is increasingly under pressure due to growing demand and the increasing likelihood of supply bottlenecks from primary sources. Securing access to TMs has become a priority issue for governments around the world with TMs linked to all industries and stages of supply chains and economies.

## 4.1 The importance of Technology Metals

As discussed, TMs enable essential functionality in many products and technologies, including ceramics; catalysts for fuel, chemicals and pharmaceuticals production; and electrical and electronic equipment (EEE) including consumer electronics and green technologies which are vital for climate change mitigation such as wind turbines, photovoltaics (PV), batteries, hybrid/electric vehicles ((H)EVs), electrolysers and fuel cells.

Accelerating technology innovation cycles and growth in emerging economies has steadily increased raw materials demand with predictions showing that global resource use could double between 2010 and 2030. Penetration of electrical and electronic equipment (EEE), in which TMs are vital to functionality, is accelerating in industrial and domestic markets. This is partly due to the deployment of emerging technologies for renewable energy generation and storage, electrification of transport, and adoption of industry 4.0 and Internet of Things (IoT) technologies to facilitate efficiency in industry and the UK's energy networks. Therefore, UK's ability to decarbonise its energy networks, transport, reduce energy and materials demand through efficiency savings in industry and facilitate an advanced state of circular economy all rely on access to TMs.

Furthermore, to meet climate and energy targets in the EU, CRM demand is expected to increase by a factor of 20 for certain materials (EC JRC, 2016), and access to TMs may limit technology deployment and the UK's ability to combat climate change (Kleijn *et al.*, 2011; Elshkaki and Graedel, 2013; Roelich *et al.*, 2014; de Koning *et al.*, 2018).

## 4.2 Sources of CRMs and issues with primary supply

For economies such as the UK, EU, and USA which rely heavily on imported CRMs, owing to limited domestic production, rising global demand creating potential supply bottlenecks is of great concern. Primary reserves of these metals are finite and increasing supply to meet demand risks exhausting natural reserves, with many technology metals at risk of exhaustion within the next century, and many being supplied from conflict minerals, introducing ethical issues around consumption from these sources (Figure 7).

The UK, Europe and rest of the world are heavily dependent on CRM supply from China and several other countries, in which primary production is concentrated (Figure 8). These countries tend to have resource hungry economies of their own and are more frequently adopting policy measures to conserve reserves that may limit supply to the rest of the world, a key example being the rare earths trade dispute with China in 2010, which sparked global efforts to ensure security of supply for the most important, or 'critical' materials for nations' economies, as mentioned earlier.

*Figure 7: The 90 natural elements that make up everything. Area of element indicates natural abundance on Earth, colour indicates degree of supply risk and conflict mineral issues. Elements present in smartphones are indicated by a smartphone icon (European Chemical Society, 2018).*

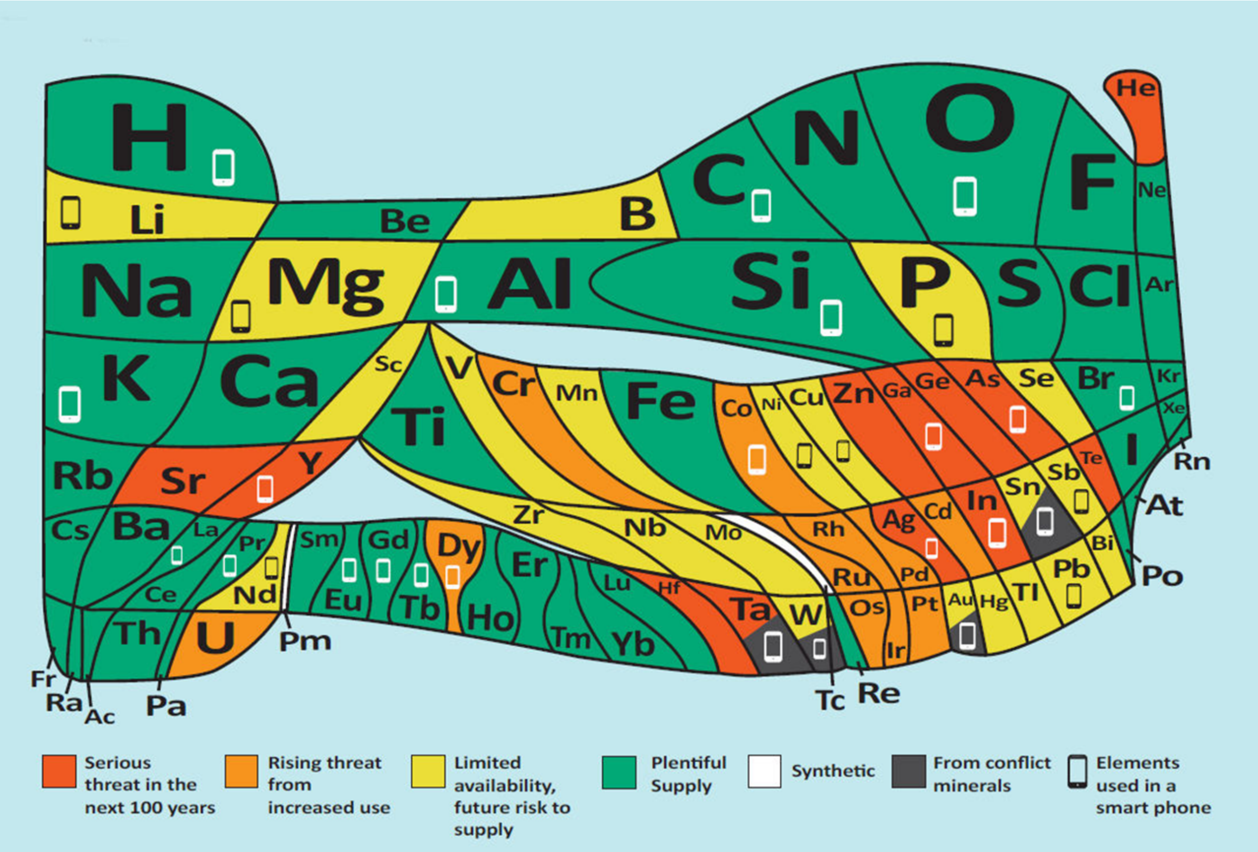
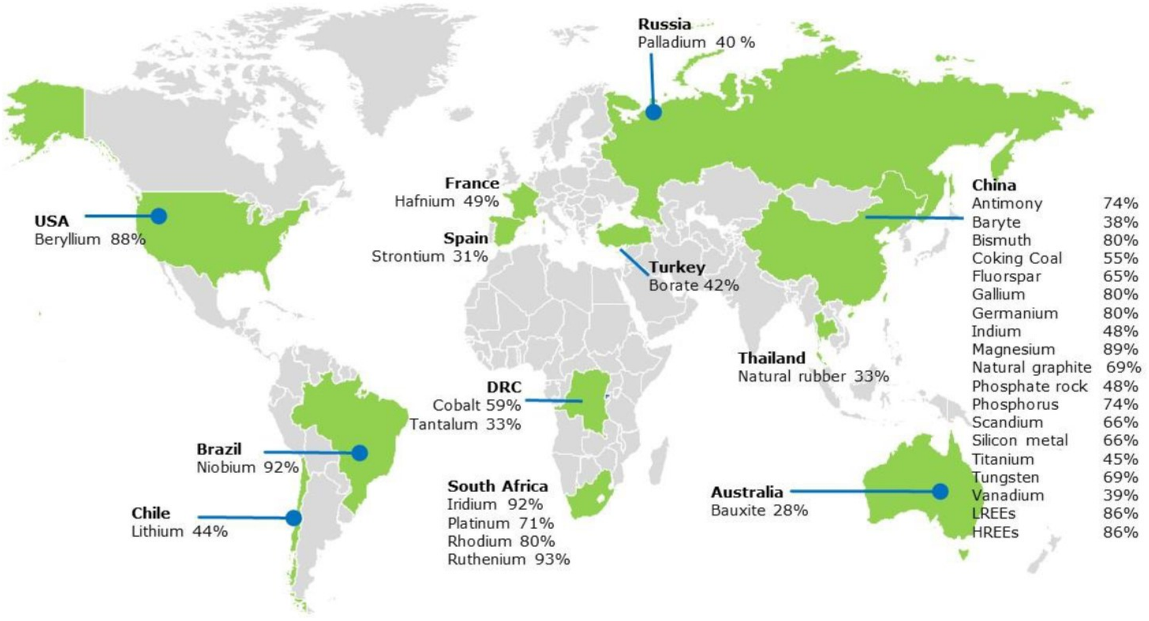


*Figure 8: Countries accounting for largest share of global CRM supply (EC, 2020).*

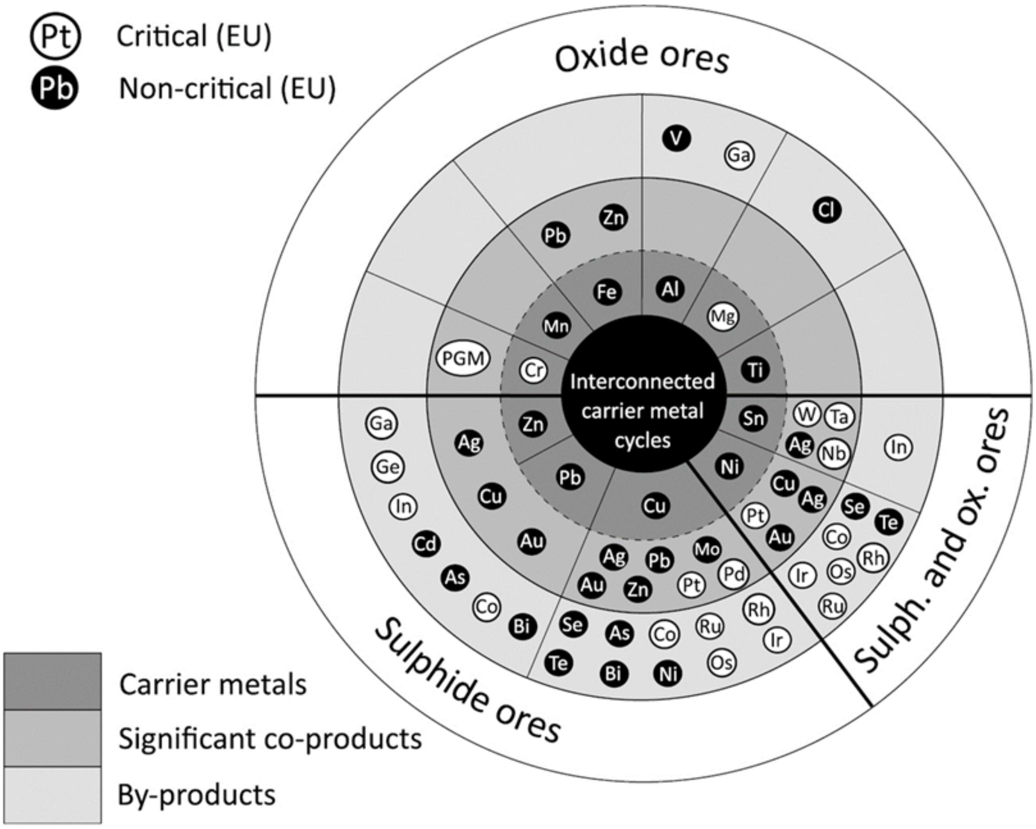


As demand for technology metals rises, supply from primary sources may struggle to keep up with this demand. In fact, no rapid solutions to issues with primary supply exist. There are a number of reasons for this. Firstly, the majority of CRMs are produced as minor by-products of the mining and refining of other major metals (Figure 9). This means that supply of CRMs depends largely on market conditions for the major metals in the ores from which they are produced. For example, germanium (Ge), an important metal used as a semiconductor in numerous applications, is produced as the minor by-product from zinc (Zn) and lead (Pb) production.

If global demand for these metals and market values are insufficient to warrant increasing production, then it is economically unviable to increase production for the sake of the minor amounts of germanium produced in the process. For some CRMs, despite high demand, conservative policy measures adopted by nations where production is concentrated may limit export and global availability.

Despite the fact that alternative primary reserves may exist elsewhere in the world, bringing new mines online is a process that takes significant time and investment, and so this is by no means a suitable solution to growing CRM demand in the short-term. In addition, CRMs suffer from price volatility, which introduces high levels of risk to any investment in new sources of these materials. In light of the growing environmental crisis, the impacts of increasing primary production to meet demand must also be considered, and the additional risk that the most significant impacts of climate change (degradation of ecosystems, land system change, biodiversity loss, and disruption to the Earth’s natural bio-geochemical flows) will be unavoidable.

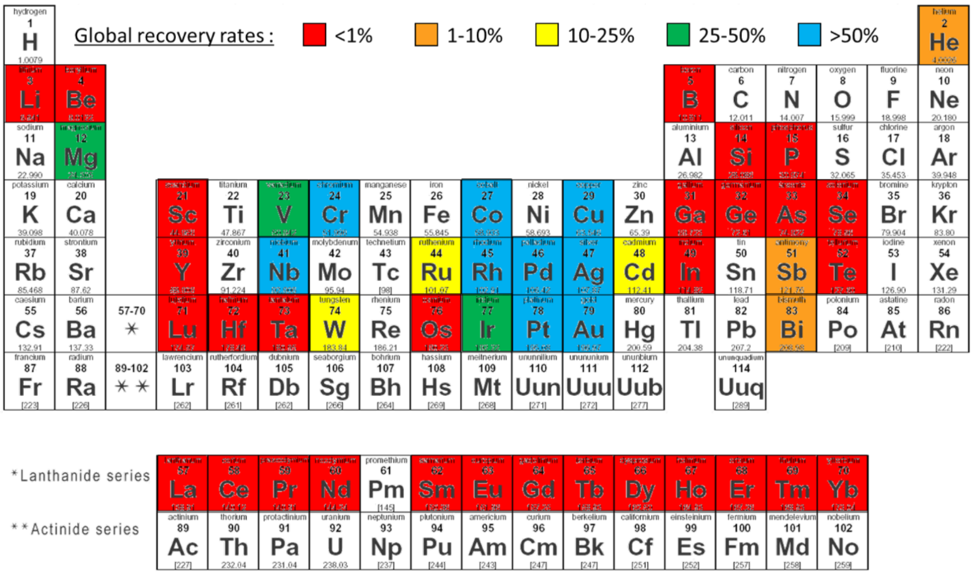
*Figure 9: The metal wheel—a schematic illustration of the geological relationships between different industrial metals and their co- and by-products; criticality is assigned according to the EU20 (Frenzel et al., 2017).*



## 4.3 Secondary Supply Issues

As increasing global consumption results in increasing rates of extraction of TMs from the Earth, the concentrations of these elements in the 'urban mine' is also increasing, improving the potential EoL products have as domestic secondary reserves of technology metals. Primary supply constraints leave importer nations with little option to mitigate materials criticality issues when access to primary sources cannot be secured, other than to develop domestic secondary supplies from wastes, comprising products that contain technology metals. To-date however, post-consumer recovery rates of CRMs from EoL products including WEEE (a rich potential secondary source of TMs), are generally very low, often <1% (Figure 10), resulting in a considerable barrier to developing domestic secondary resources (Schaeffer *et al.;* 2018a; Charles *et al.*, 2019).

*Figure 10: Global end-of-life functional recycling rates (EoL-RR) of elements highlighted as critical in recent assessments (Rhys G. Charles et al., 2018).*



Reasons for low recovery rates of CRMs, even for valuable technology metals like the precious metals (PMs) and platinum group metals (PGMs)[8], include the use of technology metals in dissipative applications, i.e., where they are present in low concentrations and the consequent high costs of collection and recovery, coupled with the low recoverable economic value of these metals, make traditional recycling routes unfeasible, resulting in loss of these elements from the economy (Reller *et al.*, 2009; Rotter and Chancerel, 2012; Zimmermann and Gößling-Reisemann, 2013; Richter and Koppejan, 2016).

Collection rates of TM-bearing items, particularly from the general public, tend to be low, with many countries around the world yet to develop sufficient regulatory systems and infrastructure to achieve high capture rates of TM-bearing products at EoL.

## 4.4 Example – Platinum

Consider the recycling rates of platinum (Pt) from various applications (Table 4). Platinum is a TM with high economic value (£770/Oz, £29,298kg)[9] reflecting its low abundance in the Earth's crust, as well as the economic and environmental costs of its production (13,954 t $CO_2$/t)[10] (Schluep *et al.*, 2009).

The high value of platinum has driven efficient collection from applications such as industrial chemical catalysts and bullion, where use and management of these materials is carefully controlled and where recycling rates are very high, ~95%. On the other hand, recycling rates from electronics are very low, 0-5%, mainly due to the dissipative use of platinum in electronics, low collection rates for WEEE, and poor product design for recycling & recovery (Hagelüken, 2012). This exemplifies the issue at hand for recovery of technology metals in general, most of which are of lower economic value, exasperating cost-benefit issues of recovery.

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[8] It should be noted, the terms precious metals (PMs) and platinum group metals (PGMs) are generally used interchangeably. However, the EU14 definition of PGMs excludes Au and Ag. To distinguish these sets of metals, further discussion uses the EU14 PGMs definition (see Table 2), and PMs refers only to Au and Ag. See section 5 for more details.

[9] LBMA 17/05/21

[10] 4,100 times greater $CO_2$ emitted per unit mass of metal produced than copper.

*Table 4: Global demand for platinum by end use sector in 2015 and estimated end-of-life (EoL) recycling rates (Rhys G Charles et al., 2018). 'Industrial Total' is the sum of the italicised subcategories below it.*

| **Application** | **Global demand** | | **Recycling Rate** (%) |
|---|---|---|---|
| | (tonnes) | % of demand | |
| Autocatalyst | 937 | 40 | 50-55 |
| Jewellery | 764 | 32 | 90-100 |
| Industrial Total | 507 | 22 | 80-90 |
| *Chemical* | 154 | 7 | ≤90 |
| *Electronics* | 47 | 2 | 0-5 |
| *Glass* | 51 | 2 | - |
| *Petroleum* | 31 | 1 | ≤90 |
| *Other Industrial* | 224 | 10 | 10-20 |
| Retail Investment | 147 | 6 | 90-100 |
| **Total** | **2,861** | | **60-70** |

## 4.5 Industry Context UK

The automotive, aerospace and defence manufacturing all depend on a number of strategic metals, as do steel companies such as Tata that use many technology metals as alloying elements in high-grade steel alloys.

**Automotive**
Almost 1.6 million vehicles were produced in the UK in 2018, comprising 1.5 million cars and 85,000 commercial vehicles. The UK motor vehicle manufacturing industry contributed £16.6 billion to the UK economy in 2018, 0.8% of the whole economy's output, and 8.5% of manufacturing output. The industry employed 166,000 people across Great Britain in 2018.[11]

The UK is a world leader in the manufacture and development of autocatalysts, which require platinum, palladium and rhodium (PGMs). The first autocatalysts were manufactured on a commercial scale by Johnson Matthey at Royston, UK. More than 90% of new passenger vehicles (cars, vans and light trucks) are now fitted with catalysts, one third of which have been supplied by Johnson Matthey[12], an estimated 480,000 vehicles.

Additionally, Niobium, a TM, is used in high strength low alloy steel, for car bodies, wheels, and structural members.

**Aerospace**
The aerospace sector directly employs 110,000 people in the UK. UK aerospace output has grown by 45 per cent since 2010, with annual turnover of £35.9bn and exports of £34.2bn in 2019[13].

Domestic companies with a large presence in the British aerospace industry include BAE Systems (the world's seventh-largest defence contractor)[14,15], Britten-Norman, Cobham, GKN, Hybrid Air Vehicles,

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[11] https://commonslibrary.parliament.uk/research-briefings/sn00611/#:~:text=1.6%20million%20vehicles%20were%20produced%20in%20the%20UK,employed%20166%2C000%20people%20across%20Great%20Britain%20in%202018.

[12] https://matthey.com/en/markets/automotive/emissions-control

[13] https://www.adsgroup.org.uk/wp-content/uploads/sites/21/2019/05/ADS-Industry-Facts-and-Figures-2019.pdf

[14] https://people.defensenews.com/top-100/

[15] https://www.sipri.org/publications/2018/sipri-fact-sheets/sipri-top-100-arms-producing-and-military-services-companies-2017

Meggitt PLC, QinetiQ, Rolls Royce (the world's second-largest maker of defence aero engines) and Ultra Electronics[16].

Nickel-based superalloys for jet engines all contain cobalt, niobium and tantalum; all TMs. Cobalt used in super-alloys represents 22% of world cobalt consumption. Additionally, Yttrium, a REE, is used as a coating for turbine blades to increase thermal resistance (Parker, 2013).

Other uses of TMs include Beryllium and beryllium alloys, which are used in vehicle landing gear and brakes (Parker, 2013).

**Electrical and electronic equipment (EEE)**
The estimated turnover of the UK EEE industry was £13.9b in 2019[17]. The TM requirements for the sector include antimony, beryllium, gallium, graphite, indium, magnesium and REEs. One example application for TMs in the UK EEE industry is in semi-conductor wafers, such as those produced by IQE, who are based in Wales. IQE is a leading supplier of gallium nitride (GaN) epitaxial wafers for radio frequency (RF) and RF power applications.

**Photonics**
The manufacturing of optical fibres in the UK was worth over £600m[18] in 2019. Germanium, a TM, is the main critical material used in optical products, where 30% of its production is used in fibre optics (Parker, 2013).

**Solar cells**
Solar cells require the use of TMs, including tellurium, indium and gallium. Over 91.6% of the world supply of tellurium, 46.8% of indium and 3.9% of gallium are used in solar cells. However, there are currently only a few manufacturing operations for solar cells in the UK[19], but this may grow with the UK development in Oxford (Oxford PV), utilising perovskite-silicon solar cells[20] which contain the TM titanium. The cells have achieved a certified 28% conversion efficiency, which is higher than most other forms of tandem panels.

# 5 Which are the key technology metals (TMs)?

As noted, TMs have remained the core of the EU CRM list through two revisions, with expansions of the range of materials evaluated (EC, 2010, 2014b, 2017), and are consistently found critical by other assessments (Jin, Kim and Guillaume, 2016; Frenzel et al., 2017; Hayes and McCullough, 2018). In addition to these materials being deemed critical by the EU, the strategic importance of securing the supply of the ‘precious metals’ (PMs), Gold (Au) and Silver (Ag), (vital elements used in electronics) has been well-explored, with PMs found to be critical by numerous stakeholders, including the UK Government. Lithium (Li) demand, as a vital component of Lithium-Ion Batteries (LIBs), required for consumer electronics and green technologies for climate change mitigation such as hybrid and electric vehicles ((H)EVs) and renewable energy systems, is also rising, with great risks to primary Li supply anticipated due to soaring global demand. Although not all technology metals are deemed critical within the context of the EU assessment, mitigation of global resource security issues for all these metals is deemed to be important.

This report provides details on the various materials deemed insecure or at risk by recent reports and those that can be classified as technology metals (TMs or speciality metals) defined as any “rare” metal or a metal that is in demand, available, and used for the purpose of furthering technology and engineered systems, i.e., those which impart special functionality. A full list of these materials, broken

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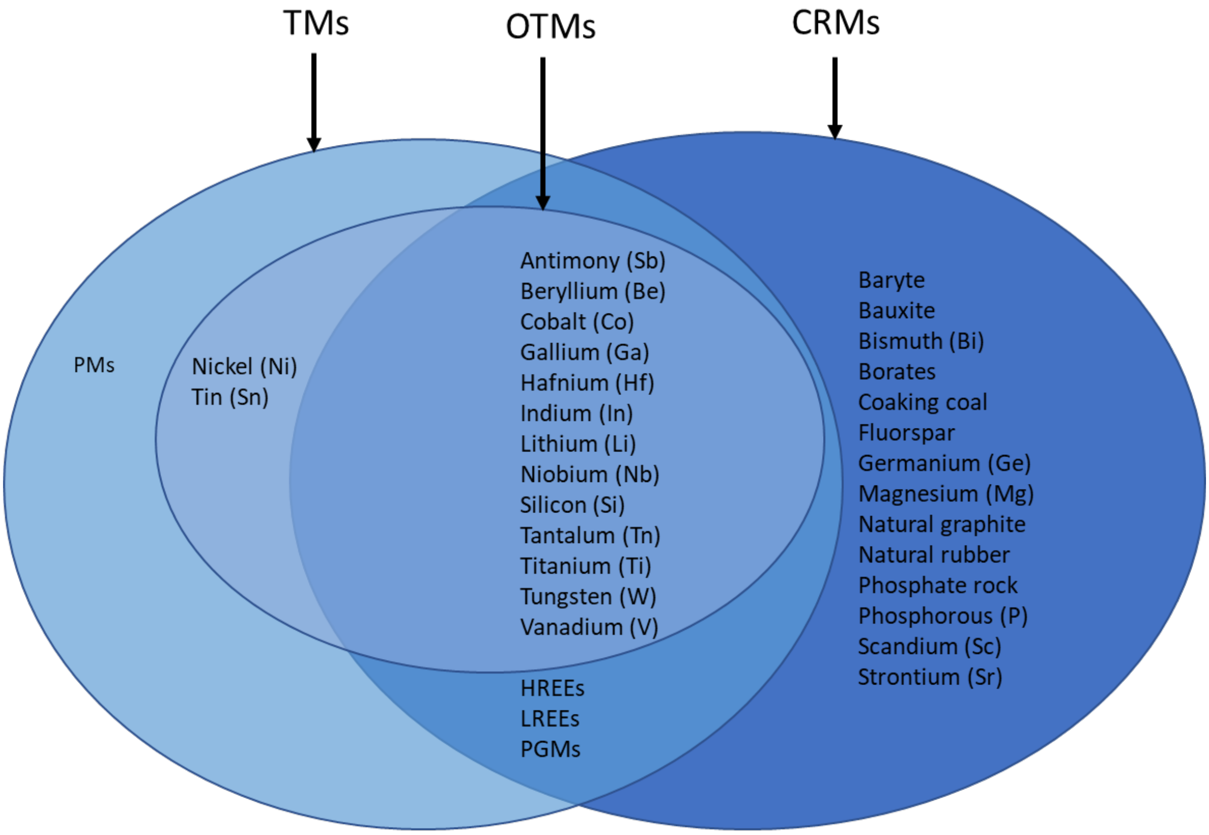
[16] https://www.rolls-royce.com/about.aspx
[17] https://www.statista.com/study/42052/manufacturing-electrical-equipment-in-the-uk/
[18] https://www.ibisworld.com/united-kingdom/market-research-reports/fibre-optic-cable-manufacturing-industry/
[19] https://www.exeoenergy.co.uk/solar-panels/solar-panels-uk/
[20] https://www.oxfordpv.com/perovskite-pv-transform-global-solar-market

down under the various categories, are given below. Specific CRM metals (metals found within CRM lists) highlighted in bold are included in this research due to their presence within PCBs, lighting monitors and TV screen. As is visible below, the materials falling under some of these groupings overlap.

| | |
|---|---|
| CRMs | Critical raw materials: **Antimony (Sb),** Baryte, Bauxite, **Beryllium (Be),** Bismuth (Bi), Borates, Cobalt (Co), Coking coal, Fluorspar, **Gallium (Ga),** Germanium (Ge), **Hafnium (Hf), Indium (In), Lithium (Li),** Magnesium (Mg), Natural graphite Natural rubber, **Niobium (Nb), PGMs**, Phosphate rock, Phosphorous (P), Scandium (**Sc), Silicon metal (Si),** Strontium (Sr), **Tantalum (Ta), Titanium (Ti), Tungsten (W), Vanadium (V), Light Rare Earth Elements (LREEs) and Heavy Rare Earth Elements (HREEs) – detailed below.** |
| REMs | Rare earth metals (also referred to as 'Rare Earth Elements'; REEs) includes LREEs and HREEs. |
| LREEs | Light rare earth elements: Scandium (Sc), Lanthanum (La), Cerium (Ce), Praseodymium (Pr), Neodymium (Nd), Promethium (Pm) and Samarium (Sm). |
| HREEs | Heavy rare earth elements: Yttrium (Y), Europium (Eu), Gadolinium (Gd), Terbium (Tb), Dysprosium (Dy), Holmium (Ho), Erbium (Er), Thulium (Tm), Ytterbium (Yb) and Lutetium (Lu). |
| PGMs | Platinum group metals: Platinum (Pt), Palladium (Pd), Ruthenium (Ru), Iridium (Ir), Rhodium (Rh) and Osmium (Os). |
| PMs | Precious metals: Gold (Au) and Silver (Ag). |
| OTMs | Other technology metals: Antimony (Sb), Beryllium (Be), Cobalt (Co), Gallium (Ga), Hafnium (Hf), Indium (In), Lithium (Li), Nickel (Ni), Niobium (Nb), Silicon (Si), Tantalum (Tn), Tin (Sn), Titanium (Ti), Tungsten (W) and Vanadium (V). |

To separate REMs and PGMs from the other CRM metals, the remaining CRM metals, nickel and tin are grouped as OTMs. The partitioning of these materials is shown below (Figure 11). Technology Metals as a whole include rare earth elements (light and heavy), PGMs, PMs, some CRM metals, plus nickel and tin because these are found within EEE at various dissipation and concentration levels.

*Figure 11: Relationship between the different material groupings found in this report.*



It is acknowledged that additional technology metals are present to varying degrees across other products covered by the WEEE Directive. However, due to data paucity in the specific quantities present in these categories, further research and quantitative chemical analysis of these products would be required to establish technology metal content and reliably include these products within the material flow analysis (MFA) contained in this report. Moreover, further detailed analysis of TM presence and quantities within WEEE (e.g., by using X-Ray Fluorescence (XRF) and Inductively Coupled Plasma Mass spectrometry (ICP)) could be used to focus the recovery of TMs present in specific parts of WEEE assemblies and components.

Therefore, the results in this report should be considered as a minimum value of the UK opportunity. The processes and strategies for technology metal recovery outlined in this report are transferable across other WEEE categories and are suitable to unlock this additional unquantified value present in other products within the UK waste stream.

## 5.1 Technology Metals Applications

A list (non-exhaustive) of technology metals and their applications in electrical and electronic equipment (EEE) is given in the table below (Table 5). For instance, Gallium (Ga) is used in multiple applications in integrated circuits, particularly smartphones, laser diodes, LEDs, photodetectors, and solar cells.

*Table 5: List (non-exhaustive) of technology metals and their applications in electrical and electronic equipment (EEE).*

| Technology metals | Applications |
|---|---|
| Other Technology Metals (OTMs) | |
| Antimony (Sb) | Flame retardants as $SbO_3$, hardener for lead electrodes in lead acid batteries, used for decolourising and fining agent for cathode ray tube (CRT) glass, optical glass used in cameras, photocopiers, binoculars. |
| Beryllium (Be) | Used in electronics and electrical components, mainly in the form of beryllium copper alloys, widely used in telecommunications and the automotive sector. |
| Bismuth (Bi) | Many bismuth alloys have low melting points and are found in specialty applications such as solders. Many automatic sprinklers, electric fuses, and safety devices in fire detection and suppression systems contain a eutectic alloy containing bismuth. |
| Cobalt (Co) | Mainly used in portable and rechargeable batteries, e.g., Nickel-metal hydride (NiMH) or lithium-ion (Li-ion) battery packs of laptops, and as an alloy in superalloys (jet engines and turbines) and magnets. |
| Fluorspar | Acid-grade fluorspar is used as a source of hydrofluoric acid (HF). HF is a precursor in the synthesis of almost all fluoro chemicals in the production process of electronics, computer chips, printed circuit boards, and thermal insulation. Also used in smaller quantities for processing of metals including aluminium, stainless steel, and uranium for nuclear fuel. |
| Gallium (Ga) | Major use in integrated circuits, particularly mobile phones, also laser diodes, LEDs, photodetectors, and solar cells. |
| Germanium (Ge) | Used in fibre optics, infrared optics and in semiconductors for mobile phones, LEDs and solar panels (photovoltaics – PV). |
| Hafnium-(Hf) | Hafnium-based compounds are employed in gate insulators in the 45 nm generation of integrated circuits. |
| Indium (In) | Used in the form of indium tin oxide (ITO) as electrode material in flat screens (e.g., TVs, mobile phones, laptops). It is also used in infrared detectors, high-speed transistors and high-efficiency photovoltaic (PV) cells. |
| Nickel (Ni) | Used as a substate for gold plating and in the production of stainless steel. |
| Magnesium (Mg) | Used in laptop housings, automotive, and construction as an alloy due to its mechanical properties - helps light weighting of metal products due to its low density. |
| Niobium (Nb) | Niobium-titanium-tin alloy is used in medical applications in the construction of the magnetic coils for magnetic resonance imagery (MRI) equipment. Pure niobium metal is used in corrosion resistant equipment, sputtering targets, and cathodic protection systems. Lithium niobate, which is a ferroelectric, is used extensively in mobile telephones and optical modulators, and for the manufacture of surface acoustic wave devices. Niobium capacitors are available as an alternative to tantalum capacitors. |
| Scandium (Sc) | High-intensity discharge lamps. |
| Tantalum (Ta) | Used widely in capacitors in electronics and in superalloys. Also used in medical applications such as hearing aids and pacemakers. |
| Tin (Sn) | The main component of solder. |
| Vanadium (V) | Approximately 85% of the vanadium produced is used as ferrovanadium or as a steel additive. |

| Technology metals | Applications |
|---|---|
| Rare Earth Metals (REMs) | |
| Cerium (Ce) | Used in cold-cathode fluorescent lamps (CCFLs), auto catalyst, petroleum refining, metal alloys, batteries and as a glass polishing agent. |
| Dysprosium (Dy) | Permanent magnets. |
| Erbium (Er) | Laser applications – pink fluorescence. |
| Europium (Eu) | Used in CCFLs and LEDs – Provides red colour. |
| Gadolinium (Gd) | Used in CCFLs and LEDs, also in magnets. |
| Holmium (Ho) | Used in lasers. |
| Lanthanum (La) | Used in CCFLs, hybrid engines, metal alloys, batteries and as a glass polishing agent. |
| Lutetium (Lu) | No electrical and electronic use identified. |
| Neodymium (Nd) | Used in neodymium iron boron (NdFeB) magnets, hard drives in computers and laptops, headphones and batteries. |
| Praseodymium (Pr) | Used in magnets, batteries and as a glass polishing agent. |
| Samarium (Sm) | Used in NdFeB magnets, cobalt magnets (SmCo), also in batteries. |
| Scandium (Sc) | No electrical and electronic use identified. |
| Terbium (Tb) | Used in CCFLs, phosphors and in permanent magnets. |
| Thulium (Tm) | Used in medical x-ray units. |
| Ytterbium (Yb) | Used in lasers and steel alloys. |
| Yttrium (Y) | Used in CCFLs and LEDs. |
| Platinum Group Metals (PGMs) | |
| Iridium (Ir) | Used in spark plug tips and iridium crucibles to produce high-quality crystals for electronics. |
| Osmium (Os) | Used in alloy with platinum for electrical contacts, filaments in light bulbs and medical implants. |
| Palladium (Pd) | Used in multi-layer ceramic capacitors (MLCCs) and hybrid ICs). |
| Platinum (Pt) | Used in electronics and electrical contacts. |
| Rhodium (Rh) | Used in liquid crystal display (LCD) glass manufacture and electrical applications. |
| Ruthenium (Ru) | Used in hard disks, chip resistors, flat screen displays, plasma display panels and as a conductive paste used in resistor chip components. |
| Precious Metals (PMs) | |
| Gold (Au) | Many applications – electrical contacts, electric wiring and coatings. |
| Silver (Ag) | Many applications – electrical contacts, conductors and coatings |

## 5.2 Technology Metals in Printed Circuit Boards (PCBs)

Although printed circuit boards (PCBs) only represent a small proportion of the total mass of the WEEE waste stream (~3%), currently the major economic driving force for WEEE recycling comes from recovery of copper and PMs from PCBs, in which gold accounts for over 80% of the total intrinsic value (Figure 12). Gold, silver, PGMs and copper are widely recovered from PCBs, although this vast majority of this recovery occurs outside of the UK. It is also by virtue of the value inherent in these metals, in particular gold, that it becomes viable to recycle PCBs, and recover the lower value and lower quantities of technology metals present, of which there are an enormous variety. The majority of these are present in trace amounts within individual surface mount devices (SMDs), components but are also dispersed throughout the entire PCB (Figure 13 and Table 6).

The historic value of gold in relationship to other materials in WEEE is shown below (Figure 15). Although this breakdown is now over 20 years old, gold, palladium, copper, silver and tin are still likely to provide the highest revenues from recycling PCBs at present.

*Figure 12: Historic revenue breakdown by material from electrical waste (Sodhi and Reimer, 2001).*

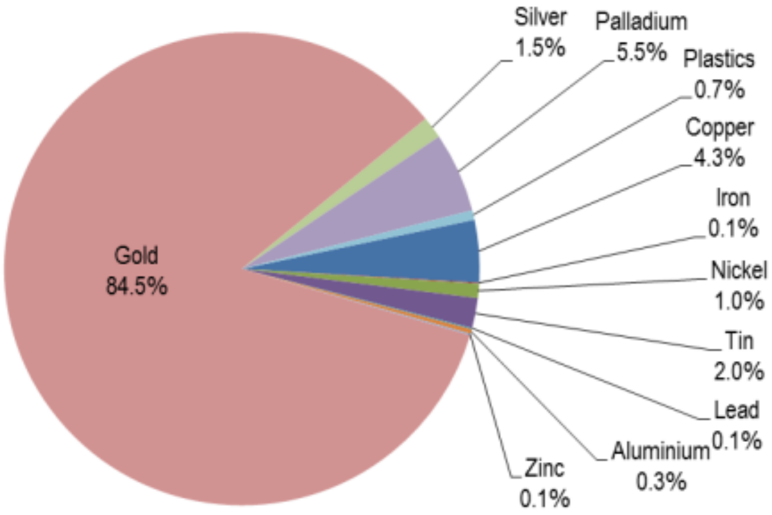
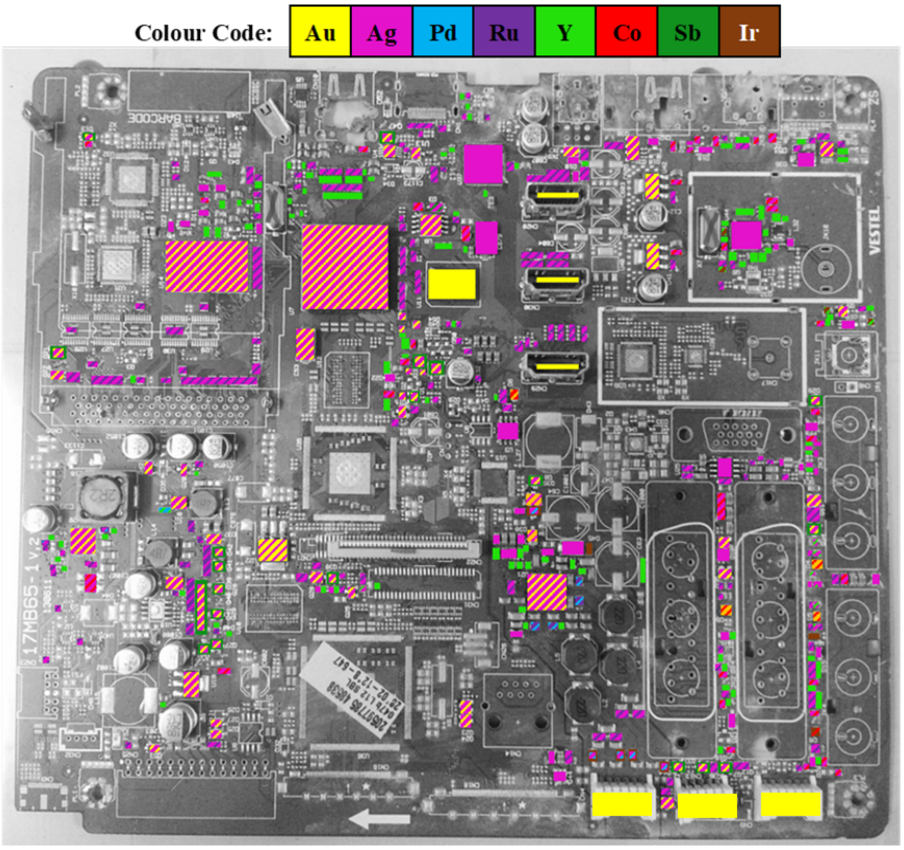


*Figure 13: Colour coded CRM distribution in TV main PCB (Charles, Douglas, Dowling and Davies, 2020).*



A list of common technology metals found in PCBs is given below (Table 6). This is a non-exhaustive list of those present and represent only those identified in the most commonly occurring components of PCBs. This demonstrates the wide range of TMs present in these commonly used components, that can be found in products across the different WEEE categories. Examples of gold found in WEEE are shown in Figure 14 and Figure 15.

*Table 6: Technology metals identified by XRF spectrometry in common PCB components (Charles, Douglas, Dowling and Davies, 2020). Technology metals identified are a non-exhaustive list of those present and represent only those identified in the most commonly occurring components of PCBs.*

*✓ - consistently present in component; (✓) – sometimes present in component*

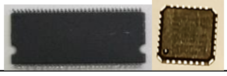
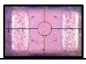
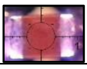
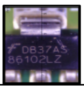
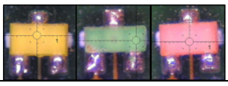
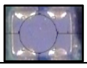
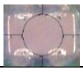
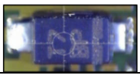
| **Component** | | **PMs** | | **PGMs** | | | **REMs** | **Other technology metals** | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | Au | Ag | Pd | Ru | Ir | Y | Co | Ga | Nb | Sb | Ta | W |
| Gold contacts | Figure 14 | ✓ | | (✓) | | | | | | | | | |
| Surface gold | Figure 15 | ✓ | | (✓) | | | | | | | | | |
| Solder | Connects all SMDs to PCBs | | ✓ | | | | | | | | | | |
| Chip resistors |  | | ✓ | (✓) | ✓ | | | (✓) | | | ✓ | | |
| Integrated circuits (ICs) |  | ✓ | ✓ | (✓) | | | | | | | (✓) | | ✓ |
| Multi-layer ceramic capacitor MLCCs |  | | | | | | ✓ | | | | | | |
| |  | | | ✓ | | | | | | ✓ | | | |
| |  | | | | | ✓ | | | | ✓ | | | |
| |  | | ✓ | | | | | | | ✓ | | | |
| |  | | | ✓ | | | | | | ✓ | | | |
| Transistors |  | ✓ | ✓ | | | | | | | | ✓ | | |
| |  | ✓ | ✓ | | | | | | | | ✓ | | |
| |  | ✓ | ✓ | | | | | | ✓ | | ✓ | | |
| Ta-capacitors |  | | ✓ | | | | | | | | ✓ | ✓ | |
| Chip arrays |  | (✓) | ✓ | (✓) | (✓) | | | | | | | | |
| Inductors |  | | ✓ | | | | | ✓ | | | | | |
| |  | | ✓ | | | | | | | | | | |
| |  | | ✓ | ✓ | | | | | | ✓ | | | |
| Polarized diodes |  | | | | | | | | | | ✓ | | |

*Figure 14: Gold (Au) coated mounted contacts of HDMI ports on a TV PCB (left) and coated edge contacts of a PCI graphics card (right).*

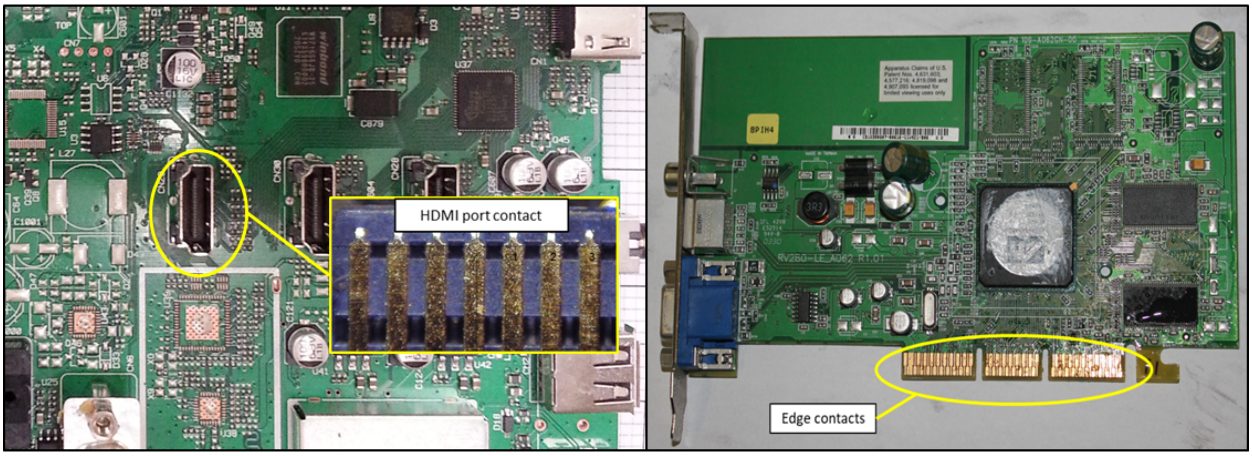
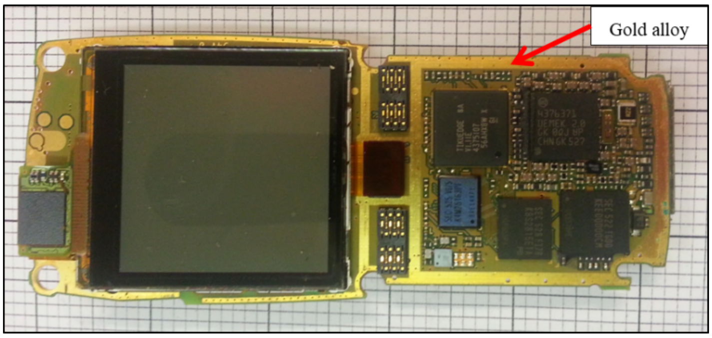


*Figure 15: Photograph of the coated Au on the surface of a mobile phone PCB.*



**Hard disk drives (HDDs)**
Hard disk drives (HDDs) are common components of numerous EEE products including PCs, laptops, servers and set-top boxes. In addition to the TMs contained in the PCBs of these devices, rare earth metals (REMs, specifically neodymium (Nd) and dysprosium (Dy)) are used in the neodymium-iron-boron (NIB) voice coil accelerator magnets. To-date, very little recovery of this alloy or the REMs contained occurs.

**Screens**
Screens are also commonly encountered components of numerous types of EEE, including monitors, TVs, tablets and smartphones. These rely heavily on technology metals for their functionality. Indium (In) is used in indium doped tin oxide (ITO) thin films, which act as transparent conducting layers in flat screens. The backlights of such screens are today one of two types: i) cold compact fluorescent lamps (CCFLs) which contain a variety of REMs in their phosphors, or ii) light emitting diodes (LEDs) which also contain REMs, and other semiconductor metals such as Ga. These light emitting technologies are also commonly encountered in other forms in the waste stream. Fluorescent lighting for example uses the same REM based phosphors present in CCFLs, and LEDs are commonly found not only in lighting, but within numerous other products. Although these are well-known applications of technology metals in EEE, numerous other technology metals are used, all with considerable barriers to their recovery from WEEE.

# 6 Methodology

The objectives and scope of this research is detailed in this section, along with the methodologies used to identify the TMs in the WEEE covered by this research.

## 6.1 Objectives

Material Focus commissioned Giraffe Innovation and Swansea University to undertake research with the key objectives:

- Examine the potential for securing supply of TMs through more effective recovery from WEEE- the opportunity;
- Conduct a Material Flow Analysis (MFA) - Quantify potentially recoverable amounts (weight) of TMs from UK WEEE and their value (£) and environmental benefit (carbon dioxide equivalent - CO2e) to the UK economy; and
- Develop a 'Technology Roadmap' - Examine emerging recycling technologies and their potential for deployment in UK WEEE recycling to increase TM recovery rates – the means.

## 6.2 Scope

The research outlined in this report includes a material flow analysis (MFA) to quantify the amounts of TMs present in WEEE and their material flows along with their value (£) and environmental impact. The technology readiness level (TRL) of nascent, current state of the art, pilot plants and proprietary technology solutions (the means) was evaluated as was their potential for deployment in UK WEEE recycling to increase TM recovery rates.

The material flow analysis (MFA) focussed on WEEE categories where quantitative data on the presence and amount of technology metals is available. This includes selected products within WEEE Category: 3: IT & Telecoms; Category 11: Display Equipment; and Category 13: Gas Discharge Lamps and LED Light Sources. The products in these categories account for an estimated 123,901 tonnes sent for recycling in the UK each year. Specific TM rich and ubiquitous products are analysed including personal computers (PCs), laptop computers, tablets, TVs, monitors, smart phones and lighting. In addition to this there are also products that are not captured in the WEEE stream such as hoarded and stolen products, products arising in commercial and industrial waste and those being illegally exported, however these were not the focus of this report.

It is acknowledged that technology metals are present to varying degrees across other WEEE products. However, due to data paucity in the specific quantities present in these categories, further research and quantitative chemical analysis of these products would be required to establish technology metal content and reliably include these products within the material flow analysis (MFA).

The technology readiness level (TRL) of nascent, current state of the art, pilot plants and proprietary technology solutions (the means) was evaluated as was their potential for deployment in UK WEEE recycling to increase TM recovery rates. The processes and strategies for TM recovery outlined in this report are transferable across other WEEE categories and are suitable to unlock this additional unquantified value present in other products within the UK waste stream. Therefore, the informed results in this report should be considered as a minimum value of the UK opportunity.

The processes and strategies for TM recovery outlined in this report are transferable across other WEEE categories and are suitable to unlock this additional unquantified value present in other products within the UK waste stream. Therefore, the informed results in this report should be considered as a minimum value of the UK opportunity.

In this report, emerging and proprietary recycling technologies are evaluated, which have the potential to increase the recovery rates of TMs from WEEE. This will support those seeking to develop next generation recycling strategies to enable a circular economy for TMs through efficient recovery from WEEE to create viable secondary resources for UK industry.

The report details:

- Material flow analysis (MFA) to quantify the scale of the opportunity for the UK which details the masses (tonnes), value (£) and embodied environmental impacts (carbon dioxide equivalent - $CO_2e$) of TMs contained in UK WEEE, those lost (e.g., to shredding dust) within the current WEEE recycling process chain and those which are ultimately recovered or retained through product reuse (the opportunity);
- Overview of emerging WEEE recycling technologies and their application for CRM recovery (the means);
- Current state of the art TM recovery technologies, pilot plants and proprietary solutions (the means);
- Technology readiness level (TRL) of the different developing technologies;
- Commentary on the likely capital expenditure (CAPEX) and operational expenditure (OPEX) characteristics; and
- The likely environmental implications of materials recovered from the examined processes.

*Table 7: Products and kt sent for recycling in 2017 (Material Focus, 2020a).*

| Product | WEEE category | Kilotonnes (Kt) collected for recycling |
|---|---|---|
| Desktop PCs | 3. IT & Telecoms | 33.591 |
| Laptop computer & tablets | | 3.713 |
| Mobile phones | | 1.035 |
| **Sub total** | | **38.339** |
| CRT monitors | 11. Display equipment | 9.612 |
| Flat screen monitors | | 5.580 |
| CRT TVs | | 44.437 |
| Flat screen TVs | | 19.951 |
| **Sub total** | | **79.58** |
| Compact fluorescent tubes | 13. Gas Discharge Lamps and LED Light Sources | 1.862 |
| Straight tube lamps | | 2.552 |
| LED lamps | | 1.568 |
| **Sub total** | | **5.982** |
| **Total** | | **123.901** |

The rationale for the focus on these WEEE categories includes:

- These categories contain high concentrations of PMs, PGMs, REMs and at least 13 other TMs;
- There is a high level of confidence in the TM, PM, PGM and REM content in these devices, with high quality compositional data available;
- Display equipment is the third highest by weight WEEE category, collected through the official system;
- 'IT and telecoms' is the fourth highest WEEE category by weight, collected through the official system;
- PMs and PGMs are concentrated in the medium or high-grade PCBs of these products and are therefore more attractive for recovery due to the high market price of these materials e.g., Gold (AU);
- The majority of REMs are contained in lighting and hard disk drives (HDD);
- Gas discharge lamps and LED light sources contain up to 7 different REMs; and

- Indium is found predominantly in screens as the transparent conductive indium doped tin oxide (ITO).

This report examines nascent technologies, pilot plants and proprietary solutions (the means) and their potential for deployment in UK WEEE recycling to increase technology metal recovery rates. Based upon previous amounts of WEEE collected and the statutory collection targets imposed by legislation in the UK, it is anticipated that generation of WEEE in the IT and Telecoms category will continue to rise, increasing the viability of this WEEE as a domestic technology metal source. Over the long term, lamp collections are declining as the move from fluorescent (typical lamp life 6-10,000 hours) to LED (typical lamp life 25,000 hours) gathers pace. Sales of Liquid Crystal Display (LCD) and Light Emitting Diode (LED) screens have long since replaced longer-lived Cathode Ray Tube (CRT) TVs and monitors (Kalmykova *et al.*, 2015), which are still appearing in the waste stream, although these too will eventually disappear.

## 6.3 Stage 1: Identify the TMs present in WEEE

Secondary research was carried out through published documentation as well as 'grey literature' to identify the levels of TMs in products. Over 250 published papers were reviewed, and data collated on the TMs present in a range of products. Each paper was also assessed for the following:

- Relevance to this research;
- Products and TMs covered by the paper;
- Weight of the product examined;
- Weight of TMs in the product;
- Sources, quality, quantity and date of the data used in the report;
- If the paper was peer reviewed; and
- Review any potential bias in the report e.g., sources of funding;

For each product where sufficient accurate data was available, the TM content per product was calculated.

## 6.4 Stage 2: TMs lost at different processing stages

From stage 1 the weight of TMs in a range of products recovered as WEEE was calculated based upon the concentration of the TM per product, average weight of product and tonnes of products recovered and treated as WEEE (Material Focus, 2020a), using the following formula:

$$TMr = (TMp/Pw) * Ww$$

Where:

- TMr=weight of TMs in product
- TMp= Weight of TM in product
- Pw =average weight of product
- Ww= total weight of products recovered as WEEE

This was repeated for each TM present in each product.

Details of the TM losses per material or material group was calculated by determining the levels of TMs potentially recoverable and those lost from WEEE as per the previous calculation, and calculating their losses using data obtained from the report "Mapping consumption and waste of raw materials in electrical products in the UK" published in 2012 (Parker and Arendorf, 2012a). This identified the percentage of each of the materials that were used, lost or recovered at the different WEEE processing stages. The weight of TMs lost was calculated using the following formula.

$$TMl = TMr * TMl\%$$

Where TMl% (is the percentage of TMs lost based upon the published data (Parker and Arendorf, 2012a). The weight of each TM recovered is the difference between TMr and TMl. This calculation was repeated for each TM present in the product.
Using this formula, the TMs recovered in products, lost in shredding, lost abroad and recovered abroad were calculated. Data on the losses of some REEs was not available and it was assumed that the fate of these was the same as the other REEs in the research (Parker and Arendorf, 2012a).

The value of the materials was determined by first quantifying the levels of these materials based upon the tonnes sent for recycling and multiplying it by the value as posted on the following web sites: London Metals Exchange (www.LME.com) and Shanghai Metals Market (SMM) (www.metal.com). The latter is the leading metals information provider for China with prices on the majority of the REMs[21].

### *6.4.1 Environmental Impact*

The carbon footprint of a material is the total greenhouse gas (GHG) emissions caused directly and indirectly by the extraction of the material and processing of it into a usable form. The carbon footprint in kilograms of carbon dioxide equivalents (kg$CO_2$e) per kg of TM was determined using Ecoinvent Swiss Lifecycle Inventory database v.3.5 to the International Planet Protection Convention (IPPC) standard. This dataset is recognised worldwide as one of the leading databases, containing over 10,000 materials. There were some data gaps, in particular the rare earth elements, and the footprint of these was determined from the following report: 'Life Cycle Assessment of Metals: A Scientific Synthesis' carried out by Nuss and Eckleman (Nuss and Eckelman, 2014).

Reliable data on TM concentration was available for laptops, desktop computers including hard disc drives (HDD), mobile phones, display equipment (TVs and monitors), fluorescent tubes and LED lighting. This is due to data paucity on TM content in other WEEE categories as previously noted. Although the presence of TMs in known for some products, the exact type and quantities has not yet been determined in items such as set top boxes, games consoles, baby alarms and other monitoring devices including Internet of Things (IOT) (modern washing machines/fridges) containing integrated circuits (ICs), inductors and chip arrays, which contain TMs. Further research using analysis equipment (e.g., X-Ray Fluorescence (XRF) and Inductively Coupled Plasma Mass spectrometry (ICP)) to determine TM concentrations in WEEE assemblies and components is required to validate TM presence and quantities in these products.

## 6.5 Stage 3: Identify different technologies used to recover the TMs from WEEE

Secondary research was carried out through published documentation as well as 'grey literature' to identify the different technologies suitable for recovering TMs from WEEE. This was supported by expert interviews with selected commercial and academic organisations developing the most promising technologies. Details on the following were captured whenever possible:

- Type of TM recovery technology;
- TMs the technology could recover;
- TM recovery efficiency and operational costs (£);
- Equipment and chemicals needed for TM recovery;
- Identification of any potential high health and safety risks;
- Geographic location and originating organisation;
- Technology readiness level (TRL); and
- Potential for adaptation and scalability for the UK market.

All recovery processes that were successful in recovering TMs, are reported on, along with the TMs they could recover, theoretical recovery potential (percentage of TMs recoverable from WEEE), technology readiness level, a brief process description and the technology's advantages and disadvantages.

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[21] Prices dated Nov 2020

# 7 Identified TMs in specific WEEE

The total mass (tonnes), value (£millions) and carbon footprint equivalent (tonnes $CO_2$e) of technology metals (TMs) present in PCs, laptops, tablets, TVs, monitors, smart phones and lighting sent for recycling in the UK in 2017 is shown below (Table 8).

*Table 8: Total mass, value and carbon footprint of technology metals (TMs) present in PCs, laptops, tablets, TVs, monitors, smartphones and lighting sent for recycling in the UK in 2017.*

| Element | Mass (tonnes) | Value (£m) | Carbon footprint – GWP100a ($tCO_2e$) |
|---|---|---|---|
| OTMs | | | |
| Antimony | 43.55 | 0.20 | 381.96 |
| Beryllium | 0.02 | 0.01 | 2.37 |
| Cobalt | 0.07 | 0.0002 | 0.79 |
| Gallium | 0.35 | 0.04 | 53.51 |
| Hafnium | 2.84 | 1.86 | 105.26 |
| Indium | 6.34 | 1.05 | 836.29 |
| Nickel | 25.29 | 0.29 | 343.99 |
| Niobium | 0.67 | 0.04 | 24.77 |
| Silicon | 16.17 | 3.05 | 48.52 |
| Tantalum | 4.13 | 0.49 | 1,197.50 |
| Tin | 291.27 | 4.14 | 3,058.30 |
| Titanium | 0.71 | 0.01 | 22.75 |
| Vanadium | 0.60 | 0.19 | 136.97 |
| **Sub total** | **392.01** | **11.3702** | **6,212.98** |
| REMs | | | |
| Cerium | 4.31 | 0.01 | 32.78 |
| Dysprosium | 0.27 | 0.07 | 15.81 |
| Europium | 2.30 | 0.06 | 907.15 |
| Gadolinium | 0.47 | 0.01 | 22.12 |
| Lanthanum | 9.47 | 0.03 | 156.27 |
| Neodymium | 5.91 | 0.31 | 163.13 |
| Praseodymium | 0.84 | 6.82 | 21.73 |
| Terbium | 1.94 | 1.44 | 575.28 |
| Yttrium | 24.35 | 0.64 | 367.63 |
| **Sub total** | **49.86** | **9.39** | **2,261.9** |
| PGMs | | | |
| Palladium | 0.48 | 28.41 | 6,362.91 |
| Platinum | 0.15 | 3.21 | 10,453.47 |
| **Sub total** | **0.63** | **31.62** | **16,816.38** |
| PMs | | | |
| Gold | 2.53 | 119.69 | 124,989.28 |
| Silver | 11.40 | 6.81 | 6,078.50 |
| **Sub total** | **13.93** | **126.5** | **131,067.8** |
| **Total** | **456.42** | **178.88** | **156,359.04** |

The differences in monetary value per unit weight for the TMs present in the WEEE examined by this report are outlined in Table 9. Whilst some TMs appear to be of relatively low economic value (e.g., 382t of antimony is worth ~£200,000) the PGMs and PMs, which are found in very small quantities, have a very high relative value (e.g., 0.48t of palladium is worth ~£28.41m). Additionally, whilst some of these elements may appear common and of low monetary value at-present, due to potential future supply scarcity and a lack of substitution options, their future value could rise significantly. This reinforces the urgent need for domestic recycling of these TMs to mitigate future potential supply risk.

The fate of technology metals by weight contained within the selected WEEE (PCs, laptops, tablets, TVs, monitors, mobile phones and lighting products) is plotted as a Sankey diagram (Figures 16 and 17). Reuse of products originally sent for recycling retains 17% of the contained technology metals (~78 t) within the UK economy. It is estimated that 160 t (35%) of the TMs are dissipated and lost during shredding of WEEE in automated pre-processing within the UK. Less than half of the TMs contained in this WEEE (48%, ~218.6 t) are retained in the output fractions of pre-processing which are exported for recovery with ~206.6 t recovered and ~12 t lost during this recovery process.

Further work is needed to determine whether this reuse rate is in fact accurate. If the report has overestimated the reuse rate, then potentially there will be a higher amount of materials in the recycling stream available for TM recovery.

The annual TM losses from collected WEEE in laptops, desktop computers, mobile phones, display equipment (TVs and monitors), fluorescent tubes and LED lighting in the UK, including those dissipated in automated pre-processing, are given in Table 9 below (Material Focus, 2020a).

*Table 9: Annual TM losses from collected WEEE in the UK and those dissipated in automated pre-processing (Material Focus, 2020a).*

| Technology metals | Total **mass (t)**, *value (£m) and* embodied emissions contained in UK WEEE ($tCO_2e$) | % retained by **mass**, *value* and embodied emissions in product reuse | % losses by **mass**, *value* and embodied emissions in UK pre-processing | % losses by **mass**, *value* and embodied emissions in foreign recovery processes | **% Mass**, *value* and embodied emissions recovered abroad |
|---|---|---|---|---|---|
| PGMs[1] | 0.63 t | 17% | 30% | 3% | 51% |
| | £31.62m | | | | |
| | 16,816 $tCO_2e$ | | | | |
| REMs[2] | 49.86 | 24% | 76% | 0% | 0% |
| | 9.39 | | | | |
| | 2,261 | | | | |
| Antimony | 43.55 | 17% | 68% | 3% | 12% |
| | 0.204 | | | | |
| | 381.96 | | | | |
| Beryllium | 0.019 | 24% | 76% | 0% | 0% |
| | 0.013 | | | | |
| | 2.37 | | | | |
| Cobalt | 0.35 | 23% | 15% | 16% | 46% |
| | <0.01 | | | | |
| | 0.79 | | | | |
| Gallium | 474.63 | 17% | 83 % | 0% | 0% |
| | 0.041 | | | | |
| | 53.51 | | | | |
| Tantalum | 4,129 | 26% | 74 % | 0% | 0% |
| | 0.49 | | | | |
| | 1,197 | | | | |
| Tungsten | 0.11 | 16% | 74% | 4% | 7% |
| | 0.017 | | | | |
| | 8.90 | | | | |

*[1] Platinum group metals (PGMs) include platinum (Pt), palladium (Pd), ruthenium (Ru), iridium (Ir), rhodium (Rh), osmium (Os);*
*[2] Rare earth metals (REMs) include LREEs and HREEs (Table 5)*

## 7.1 Technology metal content

Research to determine the technology metal content in the products in scope for this report was carried out. This required reviewing the different TM recovery 'claims' in research papers and how they calculated the original TM content in the products. The level of detail in these research papers varied considerably but some sources did give detailed information (Table 10) for compact fluorescent tubes (CFLs).

*Table 10: Assumed average content of CFLs (Punkkinen, H. et al. (2017).*

| CRM | Average content (wt%) | Low range (wt%) | High range (wt%) |
|---|---|---|---|
| Cerium | 0.1456 | 0.00096 | 0.36585 |
| Europium | 0.05661 | 0.02195 | 0.08232 |
| Gadolinium | 0.00006 | - | - |
| Gallium | 0.00006 | - | - |
| Lanthanum | 0.1549 | - | - |
| Yttrium | 0.05514 | 0.03427 | 0.09146 |
| Lanthanum | 0.62734 | 0.25400 | 1.13415 |

From compiling the data, the weight of the TMs per tonne of product and therefore the total weight of TMs in the products sent for "recycling" was determined. For Desktop PCs the TM content was based upon an average product weight of 10kg and the assumption that each product contained one hard drive (HDD, not solid-state drive SSD). The TM content of laptops, flat screen monitors and TVs content covered both LCD and LED screens. It is likely that the levels of CFLs will gradually reduce as they are replaced with longer life LEDs. The weight of CRTs and LCD displays collected for recycling will also reduce significantly over time as they are replaced with lighter LED displays.

### *7.1.1 Technology metals material flow.*

The technology metals material flows were based upon the values for bulk collection of WEEE for processing ("Mapping consumption and waste of raw materials in electrical products in the UK" 2012). This research identified the percentage of each of the materials that were reused, lost or recovered in the different WEEE processing stages:

1. Reused products: this relates to products recovered for reuse after consignment to waste disposal;
2. Lost in shredding;
3. Lost abroad; and
4. Recovered abroad.

The processing of the WEEE in the UK has not changed significantly since this report was published, however, as previously noted, it is acknowledged that the percentage of WEEE arriving at recycling facilities that can be diverted for reuse may be lower than what has been modelled in this research and therefore the distribution of TMs between end destinations may require further work. Changes to the material content of the WEEE has been accounted for all the products covered in this report. As noted, the technology metal content for each product was calculated based upon data from a wide range of published sources.

Based upon this information, the technology metals reused, lost and recovered at each stage were modelled, including the associated financial losses and gains and the carbon footprint of the materials contained within each stage.

From the reviewed research papers, the kgs of technology metals per tonne of product was calculated and these are listed alphabetically in the table below (Table 11). This also shows the REMs, PGMs, PMs and other technology metals (OTMs) found in the products. A dash (-) in the table shows either no material was found or data paucity. For example, there was no data on germanium or ytterbium in any of the products. Strontium is used as the carbonate in special glass for television screens and visual display units, but no data was available on its concentration in any of the products. Lithium is missing from all of the products as the batteries, which are the main source of this material, have been removed prior to treatment of the WEEE, as required under the WEEE directive see Appendix C for more information on lithium battery recycling technologies).

Bismuth was also missing from the analyses reviewed, and this material is found in low melting point alloys and in specialty applications such as solders. Many automatic sprinklers, electric fuses, and safety devices in fire detection and suppression systems contain a eutectic alloy containing bismuth, therefore it is unlikely to be present in the products covered in the report, except for a trace amount in electric fuses. The weights of the TMs were therefore calculated as per the stated methodology.

*Table 11: Technology metal content of TM-rich WEEE items (kg/tonne).*

| Element | Laptops & tablets | PCs | Mobile phones | CRT TVs | Flat screen TV | CRT monitors | Flat screen monitors | Compact tube lighting | Straight tube lighting | LED lighting | Total (kg) |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Antimony (Sb) | 0.2200 | 0.4731 | 0.5250 | 0.5600 | 0.0710 | - | - | - | - | - | 1.85 |
| Beryllium (Be) | - | - | 0.0188 | - | - | - | - | - | - | - | 0.02 |
| Bismuth (Bi) | - | - | - | - | - | - | - | - | - | - | - |
| Cerium (Ce) | - | - | - | - | 0.0003 | - | - | 1.4560 | 0.6041 | 0.0350 | 2.10 |
| Chromium (Cr) | 0.0200 | 0.0847 | - | 0.0012 | - | - | - | - | - | - | 0.11 |
| Cobalt (Co) | 0.0186 | - | 0.0514 | - | - | - | - | - | - | - | 0.07 |
| Dysprosium (Dy) | 0.0171 | 0.0060 | - | - | - | - | - | - | - | - | 0.02 |
| Europium (Eu) | - | - | - | - | 0.0004 | - | - | 0.5661 | 0.3240 | 0.2600 | 1.15 |
| Gadolinium (Gd) | - | - | - | - | 0.0001 | - | 0.0002 | 0.0006 | - | 0.3000 | 0.30 |
| Gallium (Ga) | - | 0.0017 | - | - | 0.0003 | - | 0.0006 | 0.0600 | - | 0.1090 | 0.17 |
| Germanium (Ge) | - | - | - | - | - | - | - | - | - | - | - |
| Gold (Au) | 0.0629 | 0.0399 | 0.3125 | - | 0.0110 | 0.0194 | 0.0400 | - | - | - | 0.49 |
| Hafnium (Hf) | - | 0.0847 | - | - | - | - | - | - | - | - | 0.08 |
| Indium (In) | 0.0011 | 0.0050 | - | - | 0.0003 | - | 0.0161 | - | - | 3.8700 | 3.89 |
| Lanthanum (La) | - | - | - | - | 0.0004 | - | - | 1.5549 | 2.5740 | - | 4.13 |
| Lithium (Li) | - | - | - | - | - | - | - | - | - | - | - |
| Neodymium (Nb) | 0.6000 | 0.1000 | 0.3125 | - | - | - | - | - | - | - | 1.01 |
| Niobium (Nb) | - | 0.0199 | - | - | - | - | - | - | - | - | 0.02 |
| Nickel (Ni) | - | 0.7530 | - | - | - | - | - | - | - | - | 0.75 |
| Palladium (Pd) | 0.0114 | 0.0053 | 0.1219 | - | 0.0044 | - | 0.0080 | - | - | - | 0.15 |
| Platinum (Pt) | 0.0011 | 0.0035 | 0.0250 | - | - | - | - | - | - | - | 0.03 |
| Praseodymium (Pr) | 0.0783 | 0.0145 | 0.0625 | - | - | - | - | - | - | - | 0.16 |
| Silicon (Si) | - | - | 15.6250 | - | - | - | - | - | - | - | 15.63 |
| Silver (Ag) | 0.0714 | 0.1220 | 4.6500 | - | 0.0450 | 0.0781 | 0.1040 | - | - | - | 5.07 |
| Strontium (St) | - | - | - | - | - | - | - | - | - | - | - |

| Element | Laptops & tablets | PCs | Mobile phones | CRT TVs | Flat screen TV | CRT monitors | Flat screen monitors | Compact tube lighting | Straight tube lighting | LED lighting | Total (kg) |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Tantalum (Ta) | 0.4857 | 0.0548 | 0.4688 | - | - | - | - | - | - | - | 1.01 |
| Terbium (Tb) | - | - | - | - | 0.0001 | - | - | 0.5514 | 0.3559 | - | 0.91 |
| Tin (Sn) | - | 4.4647 | 9.3750 | 1.2800 | 1.8000 | 1.2500 | 4.8000 | - | - | - | 22.97 |
| Titanium (Ti) | - | - | - | - | - | - | 0.1266 | - | - | - | 0.13 |
| Tungsten (W) | - | - | - | - | - | - | 0.1266 | - | - | - | 0.13 |
| Vanadium(V) | - | - | - | - | - | 0.0625 | - | - | - | - | 0.06 |
| Ytterbium (Yb) | - | - | - | - | - | - | - | - | - | - | - |
| Yttrium (Y) | 0.0006 | - | - | - | 0.0058 | 0.0625 | 0.0016 | 6.2734 | 4.5808 | 0.1584 | 11.08 |
| *References* | Buchert, M. Manhart, A., Bleher, D., Pingel, D. (2012)<br>Batinic, B. et al. (2018a)<br>Chancerel, P., Meskers, C. E. M., Hagel, C., et al. (2009)<br>Chancerel, P. and Rotter, V. S. (2016)<br>Cucchiella, F. (2014)<br>Cui, J. and Forssberg, E.<br>Dowling, M. (2014a)<br>Parker, D. and Arendorf, J. (2012a)<br>Cucchiella et al., (2015) | | | Batinic, B. et al. (2018a)<br>Cui, J. and Forssberg, E. (2003)<br>Parker, D. and Arendorf, J. (2012a)<br>Cucchiella *et al.*, (2015) | | | | Binnemans, K. et al. (2013)<br>Batinic, B. et al. (2018a)<br>Cui, J. and Forssberg, E. (2003)<br>Parker, D. and Arendorf, J. (2012a)<br>Punkkinen, H. et al. (2017)<br>Cucchiella et al., (2015) | | | |

The total weight, value and carbon footprint of the technology metals present in PCs, laptops, mobile phones, tablets, TVs, monitors and lighting sent for recycling in the UK in 2017 is shown in Table 12. This is based upon data presented in the 'Electrical Waste – challenges and opportunities', Material Focus report, 2020 and were calculated using the stated methodology.

*Table 12: Total mass, value and carbon footprint of technology metals present in PCs, laptops, tablets, TVs, monitors, mobile phones and lighting sent for recycling in the UK in 2017.*

| Element | Mass (tonnes) | Value (£m) | Carbon footprint – GWP100a ($tCO_2e$) |
|---|---|---|---|
| OTMs | | | |
| Antimony | 43.55 | 0.20 | 381.96 |
| Beryllium | 0.02 | 0.01 | 2.37 |
| Cobalt | 0.07 | 0.0002 | 0.79 |
| Gallium | 0.35 | 0.04 | 53.51 |
| Hafnium | 2.84 | 1.86 | 105.26 |
| Indium | 6.34 | 1.05 | 836.29 |
| Nickel | 25.29 | 0.29 | 343.99 |
| Niobium | 0.67 | 0.04 | 24.77 |
| Silicon | 16.17 | 3.05 | 48.52 |
| Tantalum | 4.13 | 0.49 | 1,197.50 |
| Tin | 291.27 | 4.14 | 3,058.30 |
| Titanium | 0.71 | 0.01 | 22.75 |
| Vanadium | 0.60 | 0.19 | 136.97 |
| **Sub total** | **392.01** | **11.3702** | **6,212.98** |
| REMs | | | |
| Cerium | 4.31 | 0.01 | 32.78 |
| Dysprosium | 0.27 | 0.07 | 15.81 |
| Europium | 2.30 | 0.06 | 907.15 |
| Gadolinium | 0.47 | 0.01 | 22.12 |
| Lanthanum | 9.47 | 0.03 | 156.27 |
| Neodymium | 5.91 | 0.31 | 163.13 |
| Praseodymium | 0.84 | 6.82 | 21.73 |
| Terbium | 1.94 | 1.44 | 575.28 |
| Yttrium | 24.35 | 0.64 | 367.63 |
| **Sub total** | **49.86** | **9.39** | **2,261.9** |
| PGMs | | | |
| Palladium | 0.48 | 28.41 | 6,362.91 |
| Platinum | 0.15 | 3.21 | 10,453.47 |
| **Sub total** | **0.63** | **31.62** | **16,816.38** |
| PMs | | | |
| Gold | 2.53 | 119.69 | 124,989.28 |
| Silver | 11.40 | 6.81 | 6,078.50 |
| **Sub total** | **13.93** | **126.5** | **13,1067.8** |
| **Total** | **456.42** | **178.88** | **156,359.04** |

The highest weight by material was tin (291.27t) and this is recovered from the solder on the PCBs. The antimony (42.55t) is in the glass used in the screens and in the PCBs. Nickel (25.29t) is used on the PCBs under the gold edge connectors. The highest amounts of REMs are yttrium (24.35t), lanthanum (9.54t) and cerium (4.31t) all of which are used in the flat screens and lighting. Neodymium (5.91t) is used in the hard drives for the laptops and PCs.

Although the weight of gold and palladium is low compared to many other materials, their high value (£119m and £28m respectively) makes them both the most valuable metals for recovery.

Using the Material Focus report on WEEE flows, the total weight, value and carbon footprint of the different technology metals has been calculated. The materials have been grouped by other technology metals (OTMs), REMs, PGMs and PMs. Each calculation is presented in a table and two Sankey diagrams over the following tables. The first Sankey shows the elements in each group and the second shows the totals for each group.

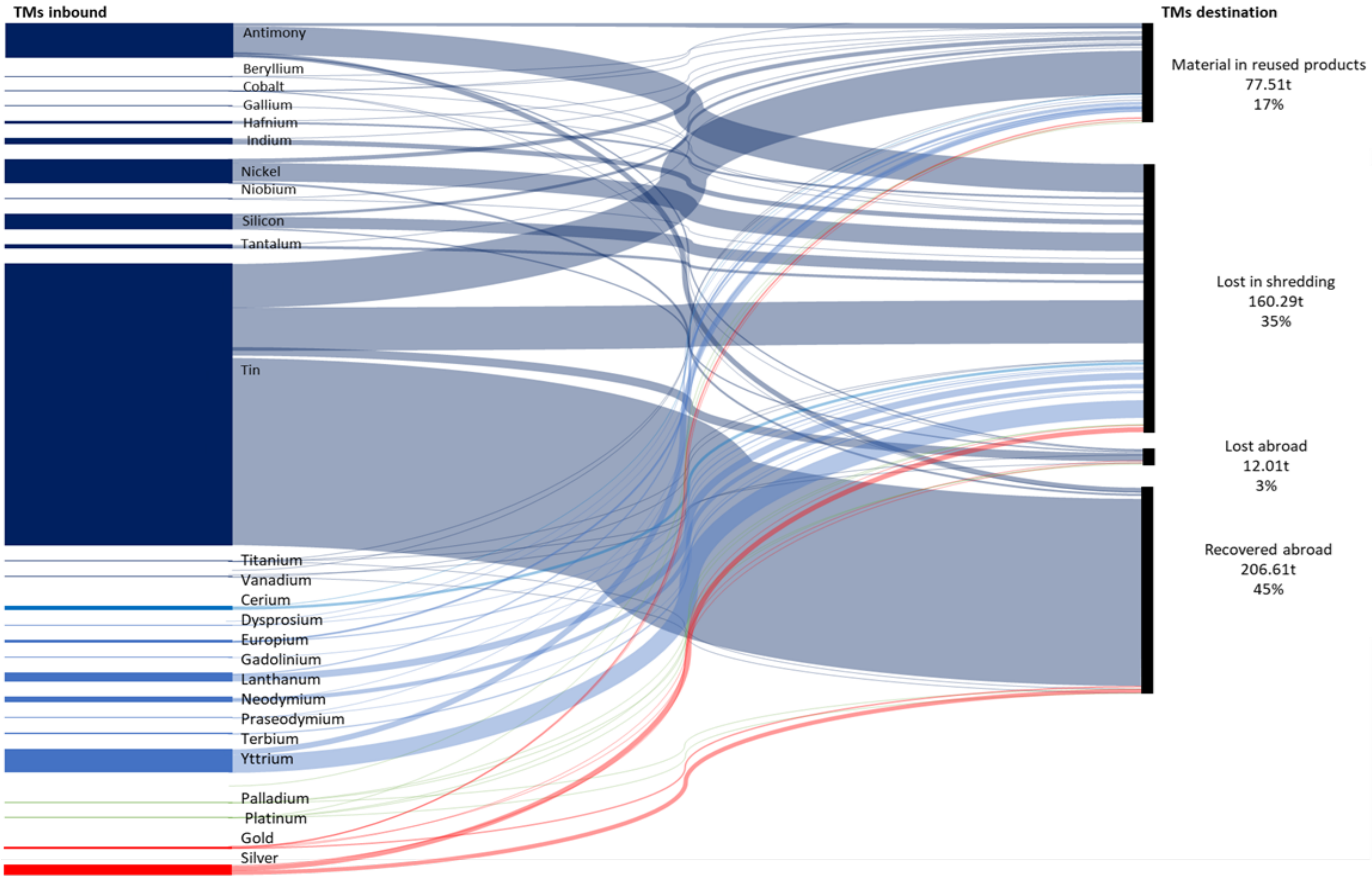
## 7.2 Technology Metals Flows by Mass

The fate of technology metals by weight contained within the selected WEEE (PCs, laptops, tablets, TVs, monitors, mobile phones and lighting products) is shown in the table below (Table 13) and as a Sankey diagram (Figure 16). Reuse of products originally sent for recycling retains 17% of the contained technology metals (~78 t) within the UK economy. It is estimated that 160 t (35%) of the TMs are dissipated and lost during shredding of WEEE in automated pre-processing within the UK. Less than half of the TMs contained in this WEEE (48%, ~218.6 t) are retained in the output fractions of pre-processing which are exported for recovery with ~206.6 t recovered and ~12 t lost during this recovery process.

*Table 13: Tonnes of materials – reused, recovered and lost.*

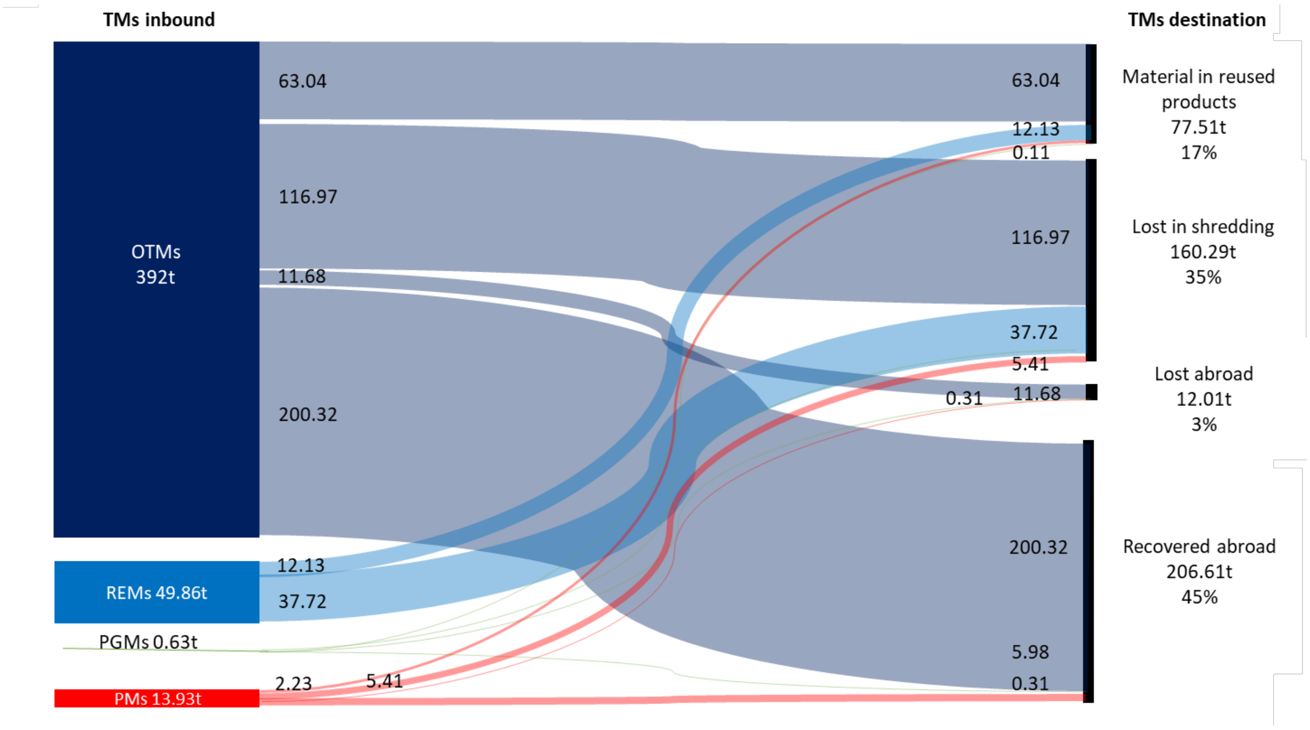
| Element | Elements in reused products | Lost in shredding | Lost abroad | Recovered abroad |
|---|---|---|---|---|
| OTMs | | | | |
| Antimony | 7.62 | 29.43 | 1.30 | 5.21 |
| Beryllium | <0.01 | 0.01 | 0.00 | 0.00 |
| Cobalt | 0.02 | 0.01 | 0.01 | 0.03 |
| Gallium | 0.06 | 0.29 | 0.00 | 0.00 |
| Hafnium | 0.69 | 2.15 | 0.00 | 0.00 |
| Indium | 1.04 | 5.30 | 0.00 | 0.00 |
| Nickel | 4.14 | 18.67 | 0.00 | 2.49 |
| Niobium | 0.16 | 0.51 | 0.00 | 0.00 |
| Silicon | 2.64 | 11.94 | 1.59 | 0.00 |
| Tantalum | 1.08 | 3.05 | 0.00 | 0.00 |
| Tin | 45.38 | 44.65 | 8.74 | 192.49 |
| Titanium | 0.11 | 0.52 | 0.02 | 0.05 |
| Vanadium | 0.10 | 0.44 | 0.02 | 0.04 |
| Sub total | 63.04 | 116.97 | 11.68 | 200.31 |
| REMs | | | | |
| Cerium | 1.05 | 3.26 | 0.00 | 0.00 |
| Dysprosium | 0.06 | 0.20 | 0.00 | 0.00 |
| Europium | 0.56 | 1.74 | 0.00 | 0.00 |
| Gadolinium | 0.12 | 0.36 | 0.00 | 0.00 |
| Lanthanum | 2.31 | 7.17 | 0.00 | 0.00 |
| Neodymium | 1.44 | 4.47 | 0.00 | 0.00 |
| Praseodymium | 0.21 | 0.64 | 0.00 | 0.00 |
| Terbium | 0.47 | 1.47 | 0.00 | 0.00 |
| Yttrium | 5.93 | 18.42 | 0.00 | 0.00 |
| Sub total | 12.15 | 37.73 | 0 | 0 |
| PGMs | | | | |
| Palladium | 0.08 | 0.14 | 0.01 | 0.24 |
| Platinum | 0.03 | 0.04 | 0.00 | 0.07 |
| Sub total | 0.11 | 0.18 | 0.01 | 0.31 |
| PMs | | | | |
| Gold | 0.45 | 0.46 | 0.08 | 1.54 |
| Silver | 1.78 | 4.96 | 0.23 | 4.44 |
| Sub total | 2.23 | 5.42 | 0.31 | 5.98 |
| **Total** | **77.51** | **160.29** | **12.01** | **206.61** |

*Figure 16: Mass flows of technology metals in assessed WEEE, by element.*



The fate of materials by group and weight (t) contained within PCs, laptops, tablets, TVs, monitors, mobile phones and lighting products, grouped as other technology metals (OTMs), rare earth metals (REMs) platinum group metals (PGMs) and precious metals (PMs) is plotted on the following Sankey diagram.

*Figure 17: Mass flows of technology metals in assessed WEEE grouped as precious metals (PMs), platinum group metals (PGMs), rare earth metals (REMs) and other technology metals (OTMs).*



The analysis of the products shows that the total weight of materials recovered and lost is 456.42t. Of this, 77.51t of the materials are bound within whole products that are recovered from the WEEE and diverted for reuse, 206t are recovered abroad, 12t lost in the processing of the WEEE abroad and 160.9t lost dues to shredding during UK processing. The majority of these losses may be preventable with use of alternative technologies to treat the WEEE specifically for the recovery of TMs (covered later).

Of the 160t of materials that are lost in shredding, 37.7t are REMs, 5t are PMs and 0.19t are PGMs. Most of these losses can be attributed to Tin (44t), Antimony (29.4t), Nickel (19t), Yttrium (18.4t) and Lanthanum (7t). Antimony was mainly found in the CRT TVs and desktop PCs, whilst nickel is found on the PCBs and Yttrium and Lanthanum are found in the fluorescent tubes.

Across all end destinations, Tin is by far the highest weight of material present (291t), as this is mainly in the PCB solder. There is also a significant amount of Antimony (43.5t), which is found in the screens and PCBs. Nickel (25t) is also found on the PCBs. The weight of CRTs and LCDs screens will reduce over time, which will impact upon the amount of Antimony that is recoverable in the future.

## 7.3 Technology Metals Flows by Material Value

The economic (material) value (Table 14) associated with the mass flow of TMs (Figure 18) shows the dominance of precious metals (PMs) which, despite having a low relative mass still account for ~71% of the economic value (£126.5m). Platinum Group Metals (PGMs) account for ~£32m (18%), Other Technology Metals (OTMs) account for ~£11m (6%) and Rare Earth Metals (REMs) ~£9m (5%).

The value of TMs currently retained in products that are reused instead of being recycled is approximately 18% of the value inherent in the WEEE collected in the UK. Meanwhile, £47 million worth of TMs are lost during UK pre-processing activities (e.g., mechanical shredding operations), and ~£100m worth of contained technology metals are exported for recovery, or 56% of the total inherent value of TMs in this WEEE. Of the exported WEEE 53% of the overall value is recovered primarily as PMs and PGMs, with the remaining 3% being lost during recovery processes.

The table and Sankey diagrams below (Table 14, Figure 18) show the value (£millions) of the materials reused in products recovered and lost.

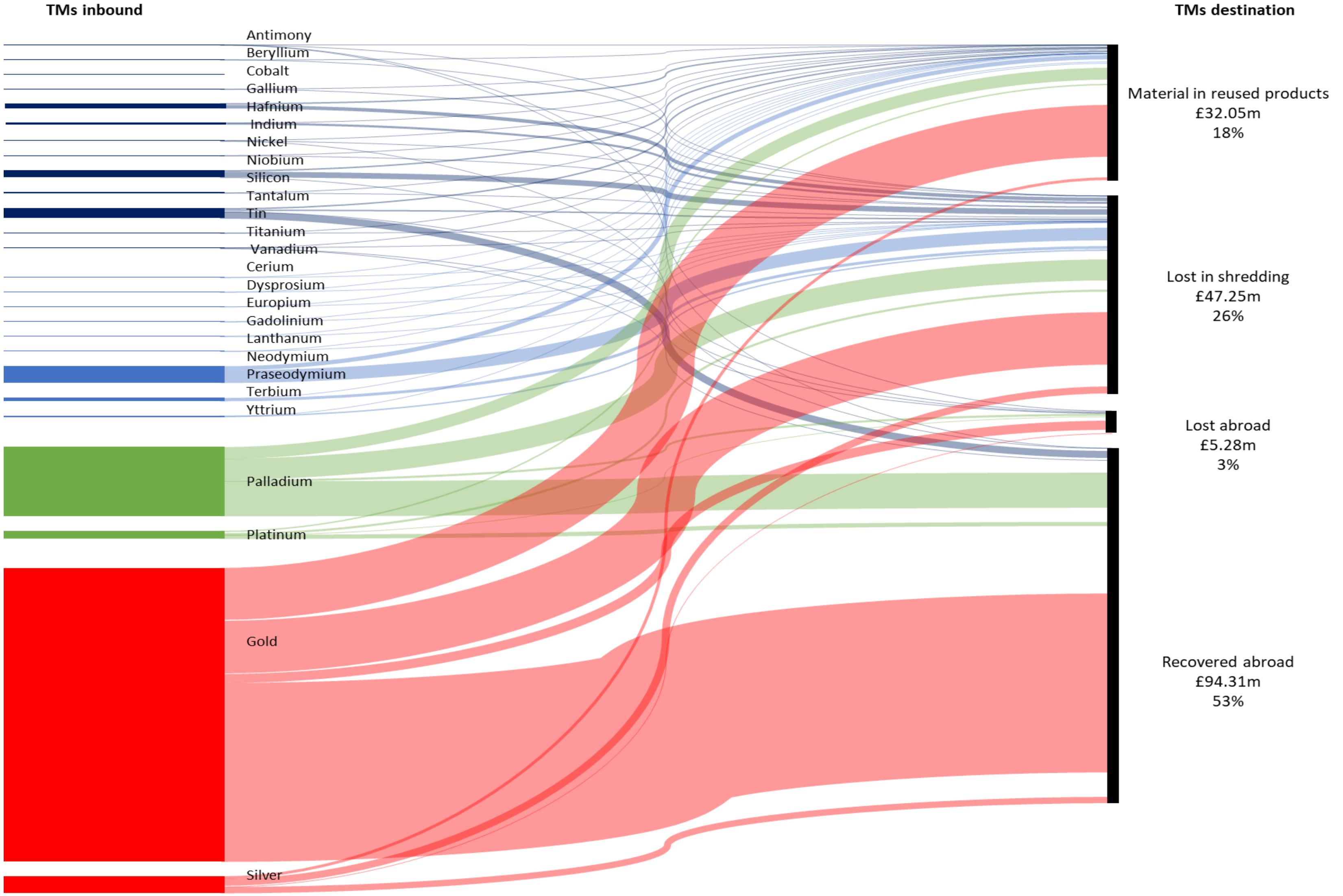
*Table 14: Materials (in £m) recovered and lost via different end destinations.*

| Element | Elements reused in products | Lost in shredding | Lost abroad | Recovered abroad |
|---|---|---|---|---|
| OTMs | | | | |
| Antimony | 0.04 | 0.14 | 0.01 | 0.02 |
| Beryllium | <0.01 | 0.01 | 0.00 | 0.00 |
| Cobalt | <0.01 | <0.01 | 0.00 | 0.00 |
| Gallium | 0.01 | 0.03 | 0.00 | 0.00 |
| Hafnium | 0.45 | 1.41 | 0.00 | 0.00 |
| Indium | 0.17 | 0.87 | 0.00 | 0.00 |
| Nickel | 0.05 | 0.21 | 0.00 | 0.03 |
| Niobium | 0.01 | 0.03 | 0.00 | 0.00 |
| Silicon | 0.50 | 2.25 | 0.30 | 0.00 |
| Tantalum | 0.13 | 0.36 | <0.01 | 0.00 |
| Tin | 0.64 | 0.63 | 0.12 | 2.73 |
| Titanium | <0.01 | 0.01 | <0.01 | 0.00 |
| Vanadium | 0.03 | 0.14 | 0.01 | 0.01 |
| Sub total | 2.03 | 6.09 | 0.44 | 2.79 |
| REMs | | | | |
| Cerium | <0.01 | 0.01 | 0.00 | 0.00 |
| Dysprosium | 0.02 | 0.05 | 0.00 | 0.00 |
| Europium | 0.01 | 0.04 | 0.00 | 0.00 |
| Gadolinium | <0.01 | 0.01 | 0.00 | 0.00 |
| Lanthanum | 0.01 | 0.02 | 0.00 | 0.00 |
| Neodymium | 0.08 | 0.24 | 0.00 | 0.00 |
| Praseodymium | 1.66 | 5.16 | 0.00 | 0.00 |
| Terbium | 0.35 | 1.09 | 0.00 | 0.00 |
| Yttrium | 0.16 | 0.49 | 0.00 | 0.00 |
| Sub total | 2.29 | 7.11 | 0 | 0 |
| PGMs | | | | |
| Palladium | 4.88 | 8.53 | 0.76 | 14.24 |
| Platinum | 0.55 | 0.96 | 0.09 | 1.61 |
| Sub total | 5.43 | 9.49 | 0.85 | 15.85 |

| Element | Elements reused in products | Lost in shredding | Lost abroad | Recovered abroad |
|---|---|---|---|---|
| PMs | | | | |
| Gold | 21.24 | 21.58 | 3.85 | 73.01 |
| Silver | 1.06 | 2.96 | 0.14 | 2.65 |
| Sub total | 22.3 | 24.54 | 3.99 | 75.66 |
| **Total** | **32.054** | **47.25** | **5.28** | **94.31** |

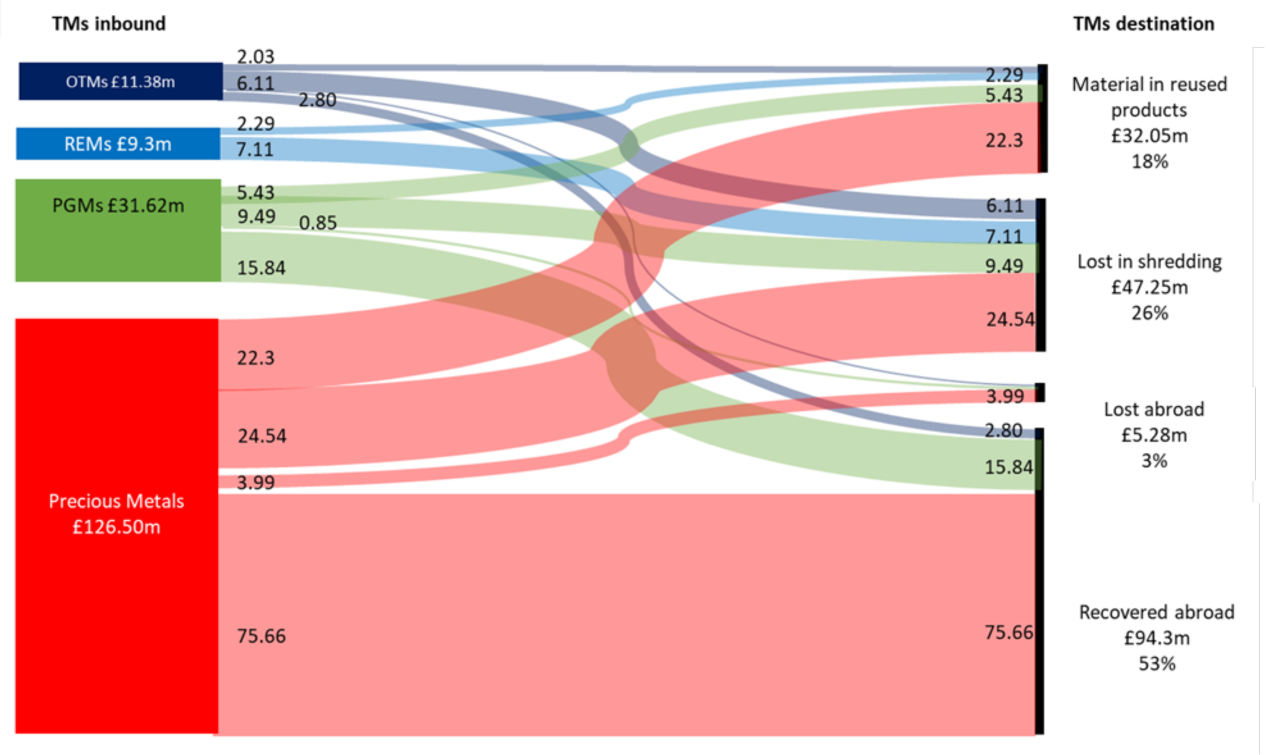
The fate of materials by value (£m) contained within PCs, laptops, tablets, TVs, monitors, mobile phones and lighting products, grouped as other technology metals (OTMs), rare earth metals (REMs) platinum group metals (PGMs) and precious metals (PMs) is plotted on the following Sankey diagram. Thickness of connecting lines and size of boxes is proportional to the value of technology metals indicated.

*Figure 18: Value flows (£) of technology metals in assessed WEEE, by element.*



The fate of materials by group and value (£m) contained within PCs, laptops, tablets, TVs, monitors, mobile phones and lighting products, grouped as other technology metals (OTMs), rare earth metals (REMs) platinum group metals (PGMs) and precious metals (PMs) is plotted on the following Sankey diagram.

*Figure 19: Value flows of technology metals in assessed WEEE, grouped as precious metals (PMs), platinum group metals (PGMs), rare earth metals (REMs) and other technology metals (OTMs).*



The results show the dominance of precious metals (PMs) which, despite having a low relative mass, still account for ~71% of the economic value (£126.5m). Platinum Group Metals (PGMs) account for ~£32m (18%), Other Technology Metals (OTMs) account for ~£11m (6%) and Rare Earth Metals (REMs) ~£9m (5%).

## 7.4 Technology Metals Flows by Environmental Impact

The environmental impact (Table 15) associated with the mass flow of TMs (Figure 20) is estimated using each TM's global warming potential (GWP) expressed as carbon dioxide equivalent ($CO_2e$) from primary production – i.e., the $CO_2$ emitted whilst extracting the same amount of these technology metals from ore. The dominant environmental impact is assigned to PMs, comprising ~ 84% of the total carbon footprint of the contained technology metals. PGMs account for 11%, OTMs 4% and REMs 1% of the environmental impact. Figure 21 also demonstrates how the $CO_2e$ of the TMs is split by their destinations i.e., that the equivalent of 25,180.32 tonnes of $CO_2e$ from PMs is lost in shredding.

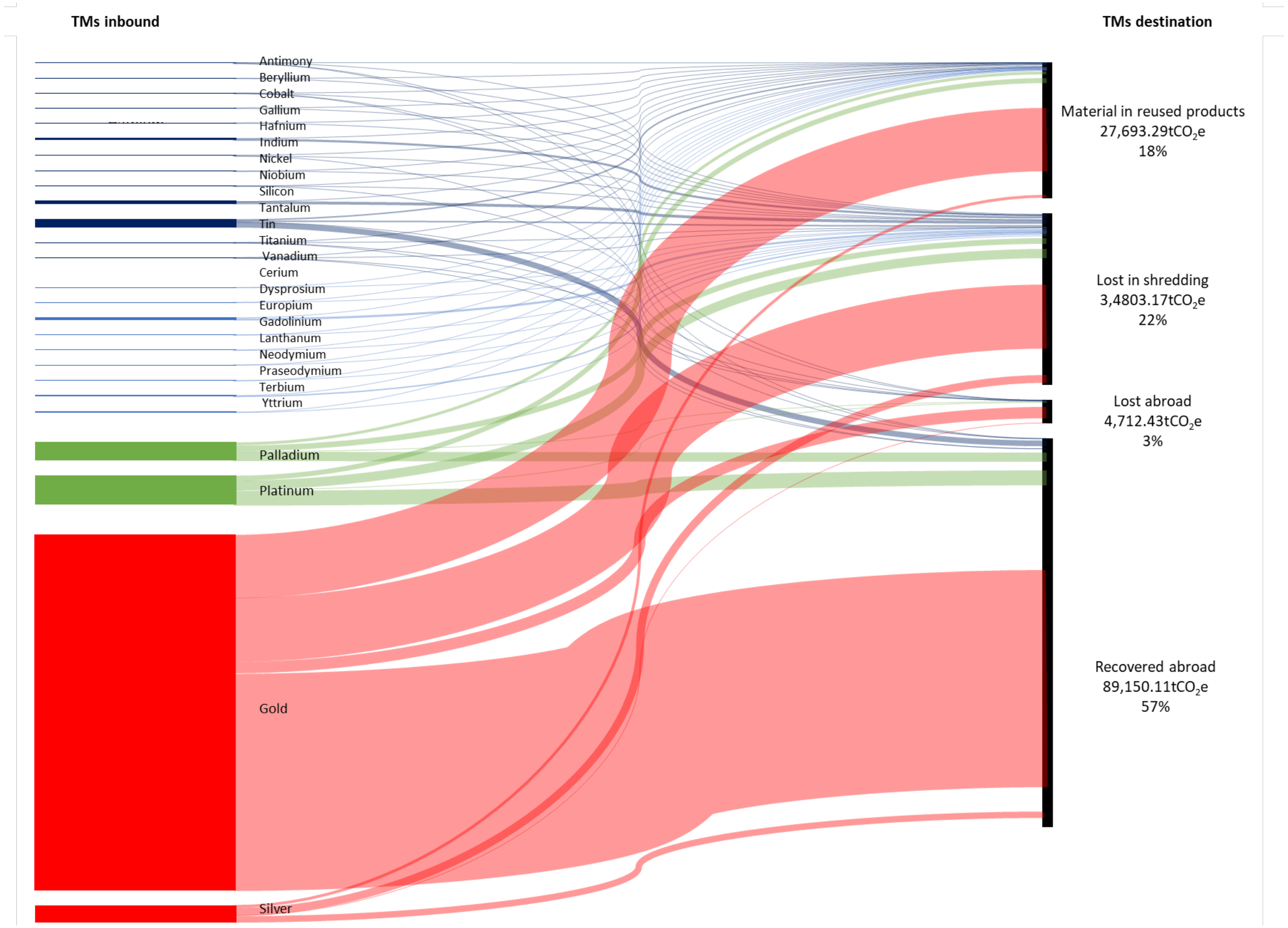
The table and Sankey diagrams below show the carbon footprint, in $tCO_2e$, of the materials reused, recovered and lost.

*Table 15: Materials recovered and lost in $tCO_2e$.*

| Element | Elements reused in products | Lost in shredding | Lost abroad | Recovered abroad |
|---|---|---|---|---|
| Technology metals | | | | |
| Antimony | 66.80 | 258.07 | 11.42 | 45.68 |
| Beryllium | 0.58 | 1.79 | 0.00 | 0.00 |
| Cobalt | 0.18 | 0.12 | 0.13 | 0.36 |
| Gallium | 8.92 | 44.59 | 0.00 | 0.00 |
| Hafnium | 25.62 | 79.64 | 0.00 | 0.00 |
| Indium | 136.78 | 699.52 | 0.00 | 0.00 |
| Nickel | 56.26 | 253.90 | 0.00 | 33.83 |
| Niobium | 6.03 | 18.74 | 0.00 | 0.00 |
| Silicon | 7.93 | 35.81 | 4.77 | 0.00 |
| Tantalum | 313.62 | 883.89 | 0.00 | 0.00 |
| Tin | 476.53 | 468.82 | 91.75 | 2,021.19 |
| Titanium | 3.64 | 16.79 | 0.68 | 1.64 |
| Vanadium | 21.92 | 101.10 | 4.11 | 9.85 |
| Sub total | 1,124.81 | 2,862.78 | 112.86 | 2,112.55 |
| REMs | | | | |
| Cerium | 7.98 | 24.80 | 0.00 | 0.00 |
| Dysprosium | 3.85 | 11.96 | 0.00 | 0.00 |
| Europium | 220.78 | 686.37 | 0.00 | 0.00 |
| Gadolinium | 5.38 | 16.73 | 0.00 | 0.00 |
| Lanthanum | 38.03 | 118.24 | 0.00 | 0.00 |
| Neodymium | 39.70 | 123.42 | 0.00 | 0.00 |
| Praseodymium | 5.29 | 16.45 | 0.00 | 0.00 |
| Terbium | 140.01 | 435.27 | 0.00 | 0.00 |
| Yttrium | 89.47 | 278.16 | 0.00 | 0.00 |
| Sub total | 550.49 | 1,711.4 | 0 | 0 |
| PGMs | | | | |
| Palladium | 1,093.62 | 1,910.29 | 170.43 | 3,188.55 |
| Platinum | 1,796.69 | 3,138.37 | 280.00 | 5,238.40 |
| Sub total | 2,890.31 | 5,048.66 | 450.43 | 8,426.95 |
| PMs | | | | |
| Gold | 22,180.57 | 22,538.32 | 4,024.70 | 76,245.69 |
| Silver | 947.13 | 2,642.01 | 124.44 | 2,364.92 |
| Sub total | 23,127.7 | 25,180.33 | 4,149.14 | 78,610.61 |
| Total | 27,693.29 | 34,803.17 | 4,712.43 | 89,150.11 |

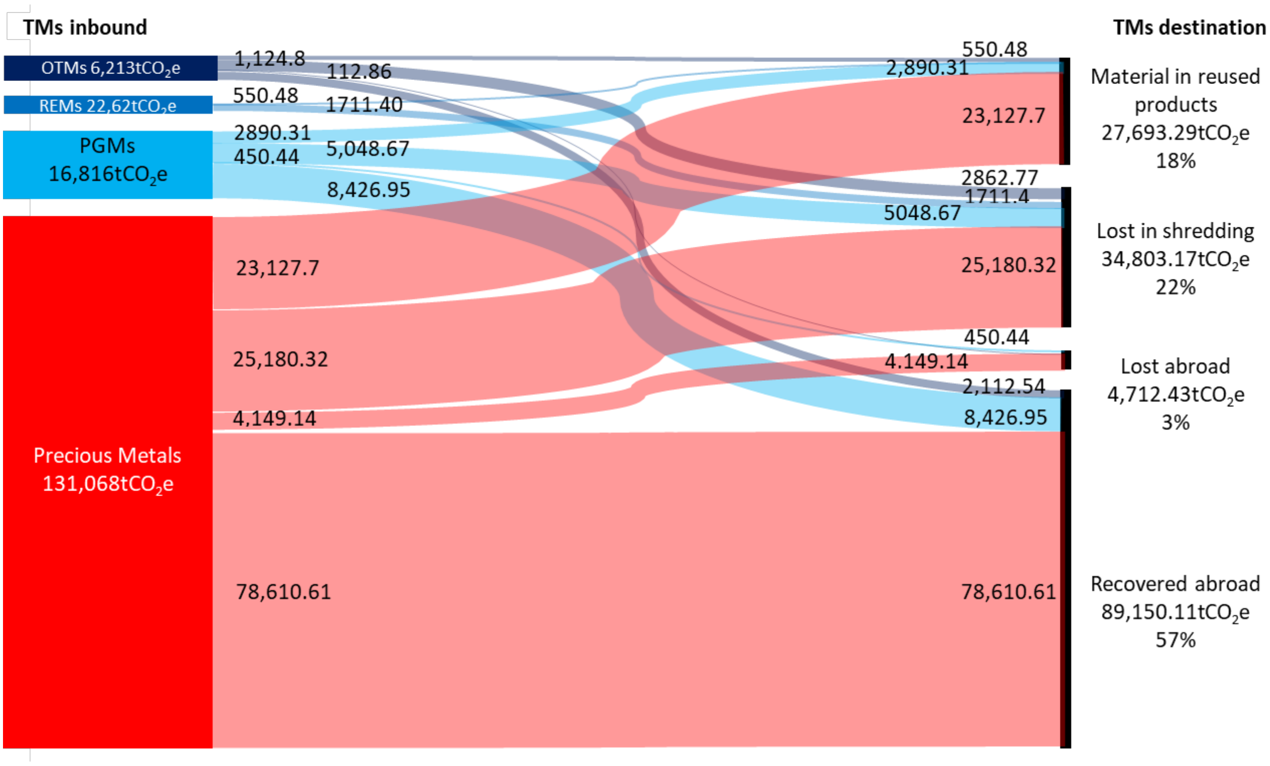
The fate of materials by environmental impact ($tCO_2e$) contained within PCs, laptops, tablets, TVs, monitors, mobile phones and lighting products, grouped as other technology metals (OTMs), rare earth metals (REMs) platinum group metals (PGMs) and precious metals (PMs) is plotted on the following Sankey diagram. Thickness of connecting lines and size of the boxes is proportional to the to the embodied carbon footprint of the technology metal indicated.

*Figure 20: Flows of embodied carbon due to primary production of technology metals in assessed WEEE, by element.*



The fate of materials by group and environmental impact (tCO$_2$e) contained within PCs, laptops, tablets, TVs, monitors, mobile phones and lighting products, grouped as other technology metals (OTMs), rare earth metals (REMs) platinum group metals (PGMs) and precious metals (PMs) is plotted on the following Sankey diagram.

*Figure 21: Flows of embodied carbon due to primary production of technology metals in assessed WEEE grouped as precious metals (PMs), platinum group metals (PGMs), rare earth metals (REMs) and other technology metals (OTMs).*



The total carbon footprint for lost and recovered materials was 156,359 tCO$_2$e. 34,803 tCO$_2$e is lost due to shredding, whilst an estimated 27,693tCO$_2$ of the TMs are retained in products recovered from the WEEE and reused.

It can be seen that PMs present in this WEEE account for 84% of the total carbon footprint of the contained technology metals with PGMs, REMs and OTMs accounting for 11%, 1% and 4% respectively.

The materials accounting for the highest carbon footprints are gold (80%), platinum (7%), palladium (4%) and silver (4%). All of these elements are mainly found in the PCBs. Of the REMs, europium (accounting for 1% of the carbon footprint) and terbium (0.4%) are mostly found in lighting. Neodymium (0.1%) is present in HDD magnets, and although some is retained through HDD reuse, all that enters recycling is lost in shredding processes.

# 8 The nature of TM losses in pre-processing of WEEE

The vast majority of TMs in UK WEEE are lost in pre-processing, before the outputs of this preliminary stage of recycling leave the UK's shores for recovery. When items are of sufficient value to justify manual disassembly, dismantling of WEEE and separation of materials and components such as housings, printed circuit boards (PCBs), liquid crystal displays (LCDs), cables, hard disk drives (HDDs) and phosphor coated glass should be carried out. These items can then be sent on to appropriate specialised recovery processes which prevents the dissipative losses of TMs in comminution and so TMs in components find their way to recovery facilities.

Pre-processing is currently conducted via manual disassembly, in which WEEE items are disassembled by operators into components for further downstream processing, or via automated systems. Due to high labour intensity, manual disassembly is costly in the UK, and so only WEEE which has sufficiently high recoverable economic value (determined primarily by PM content), and that can be disassembled in a sufficiently short period of time (determined by product design and capability of the recycling line worker) can be economically processed this way. The advantage of manual pre-processing is that all TMs present in the WEEE are retained in output fractions for recovery. However, due to the high cost, cut-off-grades (COGs i.e., the minimum grade of WEEE that can be viably processed) for manual pre-processing are high and those items which fall below COG are diverted to automated pre-processing.

These processes require initial comminution of WEEE in which devices are shredded and outputs of this process are subjected to automated materials separation based on physio-mechanical properties to recover bulk material fractions i.e., ferrous metals, copper, aluminium, plastics, glass etc. During shredding, TMs present are dispersed as fine particles throughout bulk material fractions which adhere to surfaces of larger particles. TMs are then permanently lost through unintended co-separation when bulk material fractions are separated and directed onto recovery processes which do not target these TMs for recovery. In the long term, this problem may be tackled by product eco-design to facilitate rapid product disassembly at end of life (Eol). However, in the short term, measures are required to stem these ongoing losses from WEEE that exists today. Alternative cost-effective means of comminution and product disassembly means are urgently required in order to stem these ongoing losses of TMs from our economy.

## 8.1 The nature of TM losses during materials recovery

Losses in traditional recovery processes, although occurring overseas today, must be considered and overcome with new approaches to recovery if the UK is to implement domestic infrastructure to increase secondary supply of technology metals from WEEE. Generally, some amount of further pre-processing may be necessary before further treatment to extract target materials followed by recovery and refining. Issues of pre-processing via comminution have been discussed above and are equally applicable to the processes employed in recovery plants. The discussion in this section focusses on technical limitations of extraction and recovery.

Recovery of technology metals from pre-processing output streams is conducted predominantly from printed circuit boards (PCBs, vital components of almost all WEEE) in large scale metal refineries (e.g., smelters such as Umicore in the EU, Mitsubishi metals in Asia, Glencore in North America) using a combination of specialist pyrometallurgical extraction and hydrometallurgical processes for refinement.

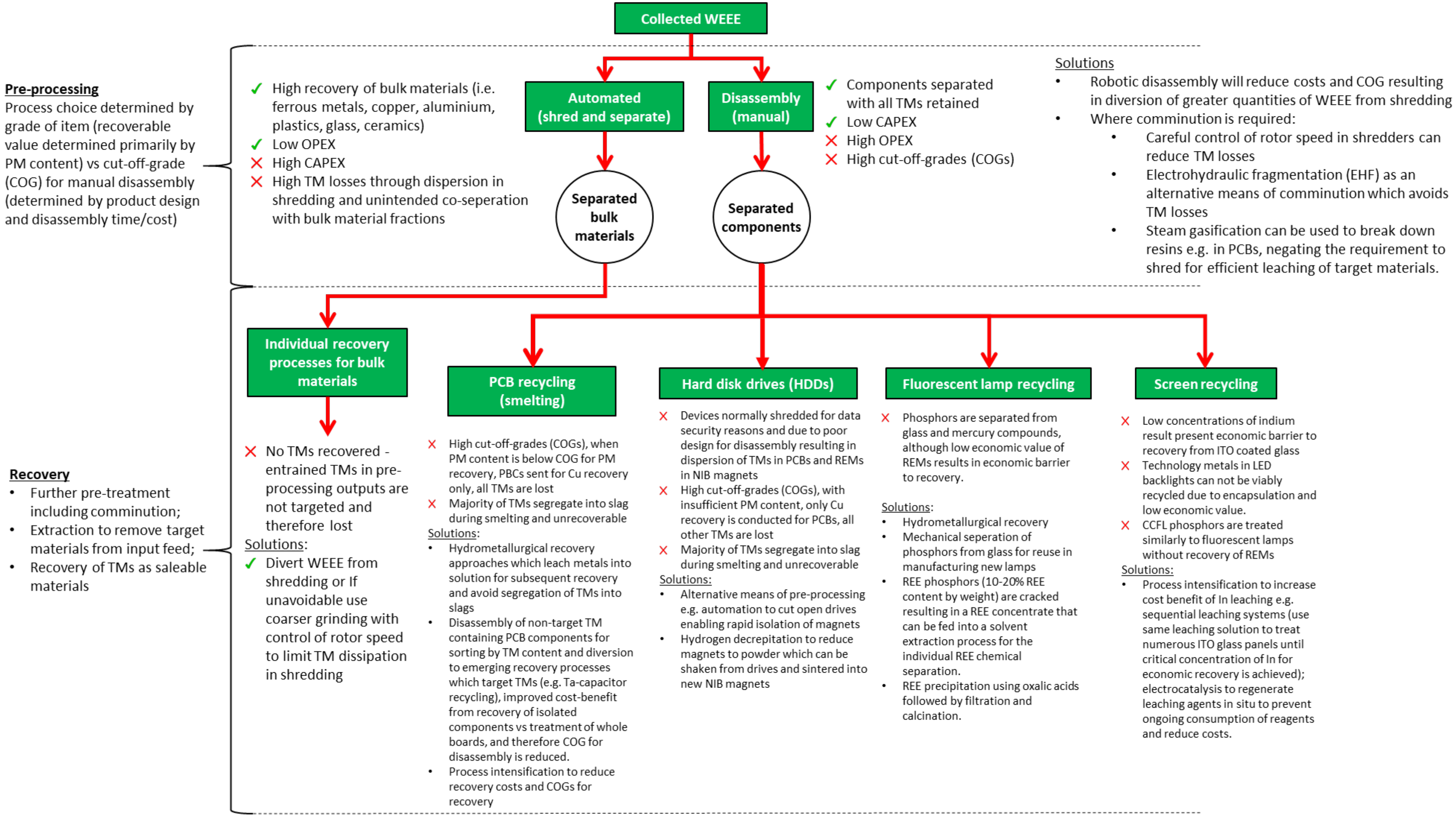
Smelting is the primary approach to PCB treatment globally due to the high recovery efficiencies afforded for high value target metals i.e., PMs, PGMs and copper, and the suitably low cost of recovery at large scale afforded by integrated smelting plants. Smelting of items such as PCBs enables collection of PM-rich copper bullion for further refinement. As copper in boards melts, it leaches other metals from the feed, carrying them to the bottom of the furnace where precious metal and PGM-rich copper bullion is produced for further refinement into individual elements. The vast majority of TMs segregate into the slag phase during smelting, from which thermodynamic and economic barriers to their recovery result in permanent loss. As such, there is no opportunity to recover these materials from outputs of this extractive smelting process owing to the way these elements distribute between molten slag and metal phases during the treatment. Methods to prevent these losses either through adoption of improved pre-processing and segregation of non-TM bearing and TM bearing components, will be essential to ensure WEEE in the UK becomes a viable domestic secondary TM source for the benefit of industry, the environment and our society.

Another issue of smelting is that it is only viable at large scale, with few plants around the world consuming the vast majority of waste PCBs generated globally. Thus, smelting as a domestic solution for a single nation, even one with such a high per capita WEEE generation as the UK, may not be viable. In addition, the high energy consumption and emissions produced by these processes make them a less attractive option in light of the global environmental crisis.

Due to the low economic value of the trace amounts of TMs present in many devices e.g., indium used in flat screens, alternative means to pyrometallurgy for recovery, such as hydrometallurgical processes which use chemical dissolution to extract materials for subsequent recovery, also face economic barriers to implementation. Although potentially viable at smaller scale, and suitable for decentralised local recovery of TMs from WEEE while consuming less energy and generating fewer emissions, efficient processing is necessary to reduce costs to the extent that recovery may be viable.

The following chart (Figure 22) shows the fate of technology metals in WEEE throughout the recycling process chain with recommended strategies to avoid losses and enhance recovery rates.

*Figure 22: The fate of technology metals in WEEE throughout the recycling process chain with recommended strategies to avoid losses and enhance recovery rates.*



**Pre-processing**

Process choice determined by grade of item (recoverable value determined primarily by PM content) vs cut-off-grade (COG) for manual disassembly (determined by product design and disassembly time/cost)

✓ High recovery of bulk materials (i.e. ferrous metals, copper, aluminium, plastics, glass, ceramics)
✓ Low OPEX
✗ High CAPEX
✗ High TM losses through dispersion in shredding and unintended co-seperation with bulk material fractions

✓ Components separated with all TMs retained
✓ Low CAPEX
✗ High OPEX
✗ High cut-off-grades (COGs)

Solutions
- Robotic disassembly will reduce costs and COG resulting in diversion of greater quantities of WEEE from shredding
- Where comminution is required:
  - Careful control of rotor speed in shredders can reduce TM losses
  - Electrohydraulic fragmentation (EHF) as an alternative means of comminution which avoids TM losses
  - Steam gasification can be used to break down resins e.g. in PCBs, negating the requirement to shred for efficient leaching of target materials.

**Recovery**
- Further pre-treatment including comminution;
- Extraction to remove target materials from input feed;
- Recovery of TMs as saleable materials

Individual recovery processes for bulk materials

✗ No TMs recovered - entrained TMs in pre-processing outputs are not targeted and therefore lost

Solutions:

✓ Divert WEEE from shredding or If unavoidable use coarser grinding with control of rotor speed to limit TM dissipation in shredding

PCB recycling (smelting)

✗ High cut-off-grades (COGs), when PM content is below COG for PM recovery, PBCs sent for Cu recovery only, all TMs are lost
✗ Majority of TMs segregate into slag during smelting and unrecoverable

Solutions:
- Hydrometallurgical recovery approaches which leach metals into solution for subsequent recovery and avoid segregation of TMs into slags
- Disassembly of non-target TM containing PCB components for sorting by TM content and diversion to emerging recovery processes which target TMs (e.g. Ta-capacitor recycling), improved cost-benefit from recovery of isolated components vs treatment of whole boards, and therefore COG for disassembly is reduced.
- Process intensification to reduce recovery costs and COGs for recovery

Hard disk drives (HDDs)

✗ Devices normally shredded for data security reasons and due to poor design for disassembly resulting in dispersion of TMs in PCBs and REMs in NIB magnets
✗ High cut-off-grades (COGs), with insufficient PM content, only Cu recovery is conducted for PCBs, all other TMs are lost
✗ Majority of TMs segregate into slag during smelting and unrecoverable

Solutions:
- Alternative means of pre-processing e.g. automation to cut open drives enabling rapid isolation of magnets
- Hydrogen decrepitation to reduce magnets to powder which can be shaken from drives and sintered into new NIB magnets

Fluorescent lamp recycling

✗ Phosphors are separated from glass and mercury compounds, although low economic value of REMs results in economic barrier to recovery.

Solutions:
- Hydrometallurgical recovery
- Mechanical seperation of phosphors from glass for reuse in manufacturing new lamps
- REE phosphors (10-20% REE content by weight) are cracked resulting in a REE concentrate that can be fed into a solvent extraction process for the individual REE chemical separation.
- REE precipitation using oxalic acids followed by filtration and calcination.

Screen recycling

✗ Low concentrations of indium result present economic barrier to recovery from ITO coated glass
✗ Technology metals in LED backlights can not be viably recycled due to encapsulation and low economic value.
✗ CCFL phosphors are treated similarly to fluorescent lamps without recovery of REMs

Solutions:
- Process intensification to increase cost benefit of In leaching e.g. sequential leaching systems (use same leaching solution to treat numerous ITO glass panels until critical concentration of In for economic recovery is achieved); electrocatalysis to regenerate leaching agents in situ to prevent ongoing consumption of reagents and reduce costs.

# 9 Summary of Technology Metal (TM) recovery methods

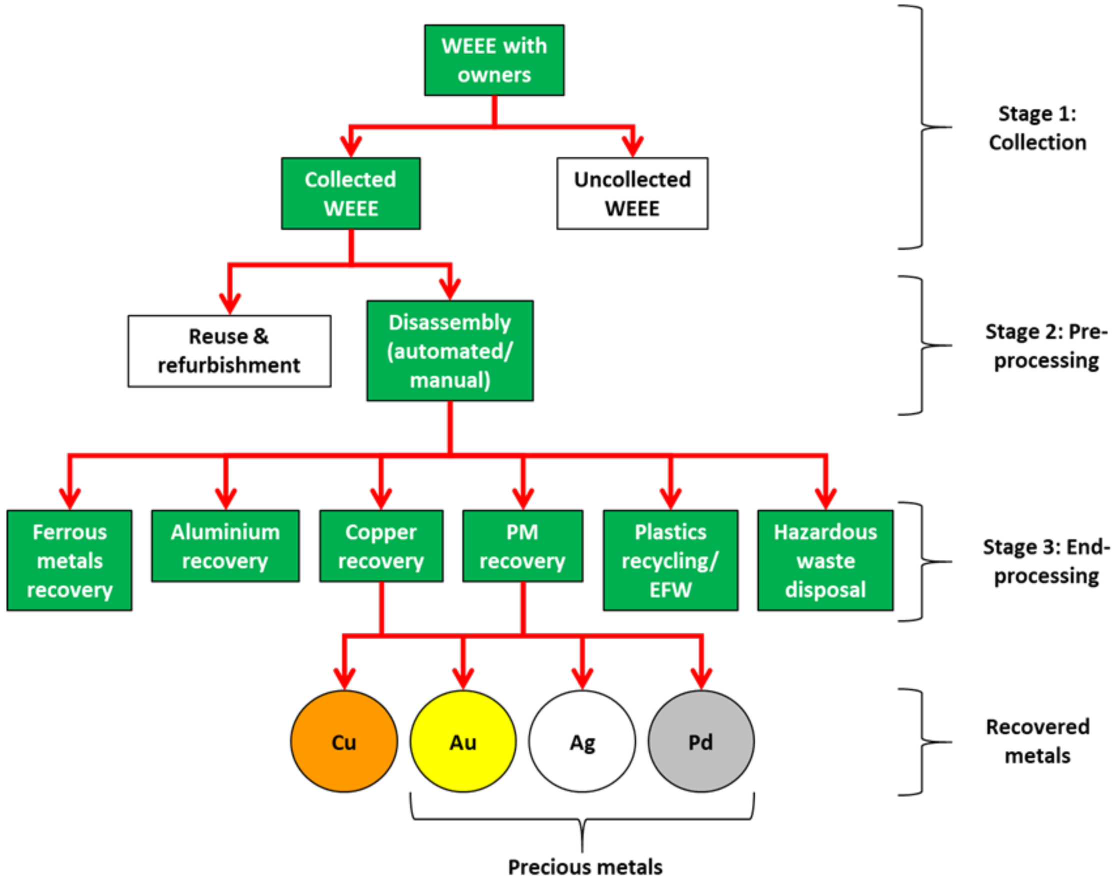
It is clear that action to mitigate materials criticality issues for the UK is urgently needed, and that Government has long since acknowledged that WEEE has high potential to serve as a domestic secondary resource of technology metals to feed UK manufacturing sectors. This section summarises the different technologies for TM recovery, which are subsequently covered in detail in sections 11-16.

Achieving high TM recovery rates is dependent on throughput, yield and thermodynamic limitations to recovery efficiency. Pre-processing losses and extraction efficiency are critical factors in achieving high TM recovery rates. The technologies presented in this report are those with highest potential to reduce technology metal losses within the recycling process chain. In order to identify both the potential economic, strategic and environmental opportunity presented by UK WEEE as a domestic source of technology metals, this report has identified promising proprietary (TRL≥4) and emerging technologies (below TRL<4) that could be deployed within the WEEE recycling process chain.

Technologies described in this report are those which hold potential to overcome barriers to TM recovery referring to >250 research, industry and Government publications. The fate of technology metals in WEEE throughout the recycling process chain with recommended strategies to avoid losses and enhance recovery rates is shown in Figure 4.

The basic WEEE recycling processes are shown in the figure below (Figure 23). This shows the recovery of PMs, Palladium and copper as the main recovered metals. There is also WEEE which is uncollected as it is either disposed of in the domestic refuse, placed in a commercial skip, hoarded, stolen or illegally exported.

*Figure 23: Simplified WEEE recycling process chain with focus on precious metal (PM) and copper recovery (Charles, 2018).*



To exploit this value, once collected, WEEE is committed to the recycling process chain which is divided into 2 distinct stages; pre-processing, and recovery (or end-processing). Pre-processing is conducted in the UK with either manual disassembly of devices into constituent components and materials, or with automated systems which shred whole devices and separate bulk materials according to physical properties i.e., magnetic separation to remove ferrous fraction, eddy current or corona separation to remove copper and aluminium, whilst density separation is used to remove plastics.

The output fractions of pre-processing are then sent onward to appropriate processes, which recover the valuable constituents and dispose of hazardous components in a safe and environmentally compliant manner. To-date, the UK has been entirely reliant upon foreign recovery infrastructure, with efforts to mitigate materials criticality driven by the EC at an EU level. With the prospect of becoming an 'economic competitor' to the EU, the UK may be more vulnerable than ever to resource criticality issues, having to export TM rich wastes (potential secondary raw materials for the UK) at greater cost for recovery, relinquishing TMs from the UK's economy. This may raise cut-off-grades[22] for viable recovery via manual pre-processing conducted in the UK, forcing greater quantities of WEEE to be processed via the highly automated recycling routes which disperse the vast majority of contained TMs during comminution through unintended co-separation with bulk fractions, resulting in their permanent loss. However, the ability to recover TMs more efficiently, and generate added-value TM feedstocks for use by UK industries would improve the overall cost-benefit of WEEE recycling and

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[22] When assessing primary resources, a cut-off grade (COG) is established, below which it is not economically feasible to mine. It is possible to establish similar recycling COGs based upon the recoverable material value from an item and costs incurred in the recycling process chain. COGs are higher in the UK for manual pre-processing (which disassembles products and separates their constituent components/materials with all TMs intact, making them available to downstream recovery processes), than for automated pre-processing, which dissipate TMs during comminution of WEEE via shredding for subsequent physio-mechanical separation of bulk materials.

TM recovery. This would place the UK in a strong position to increase value derived from WEEE and secure greater quantities of TM-bearing WEEE and WEEE-derived materials from overseas. This would increase the UK's access to TMs, despite fierce competition from established re-processors.

## 9.1 Pre-processing

Alternative approaches to mechanical shredding for use in automated pre-processing, particularly electrohydraulic fragmentation (EHF), represent a high technology readiness level (TRL) alternative which avoids dissipation of technology metals, enabling cost-effective pre-processing which delivers greater quantities of technology metals in output fractions for subsequent recovery.

The removal of solder and components from PCBs using hydrometallurgy or thermal desoldering can minimise the need for shredding and significantly reduce TM losses. Other approaches to remove polymeric components from WEEE components such as supercritical water treatment, or pyrolysis may also enable access to contained technology metals for recovery without incurring the dissipative losses observed in shredding. If shredding is unavoidable, using coarser grinding with feedstock-appropriate control of rotor speed will limit technology metal dissipation in the shredding process.

## 9.2 TM Recovery and extraction technologies

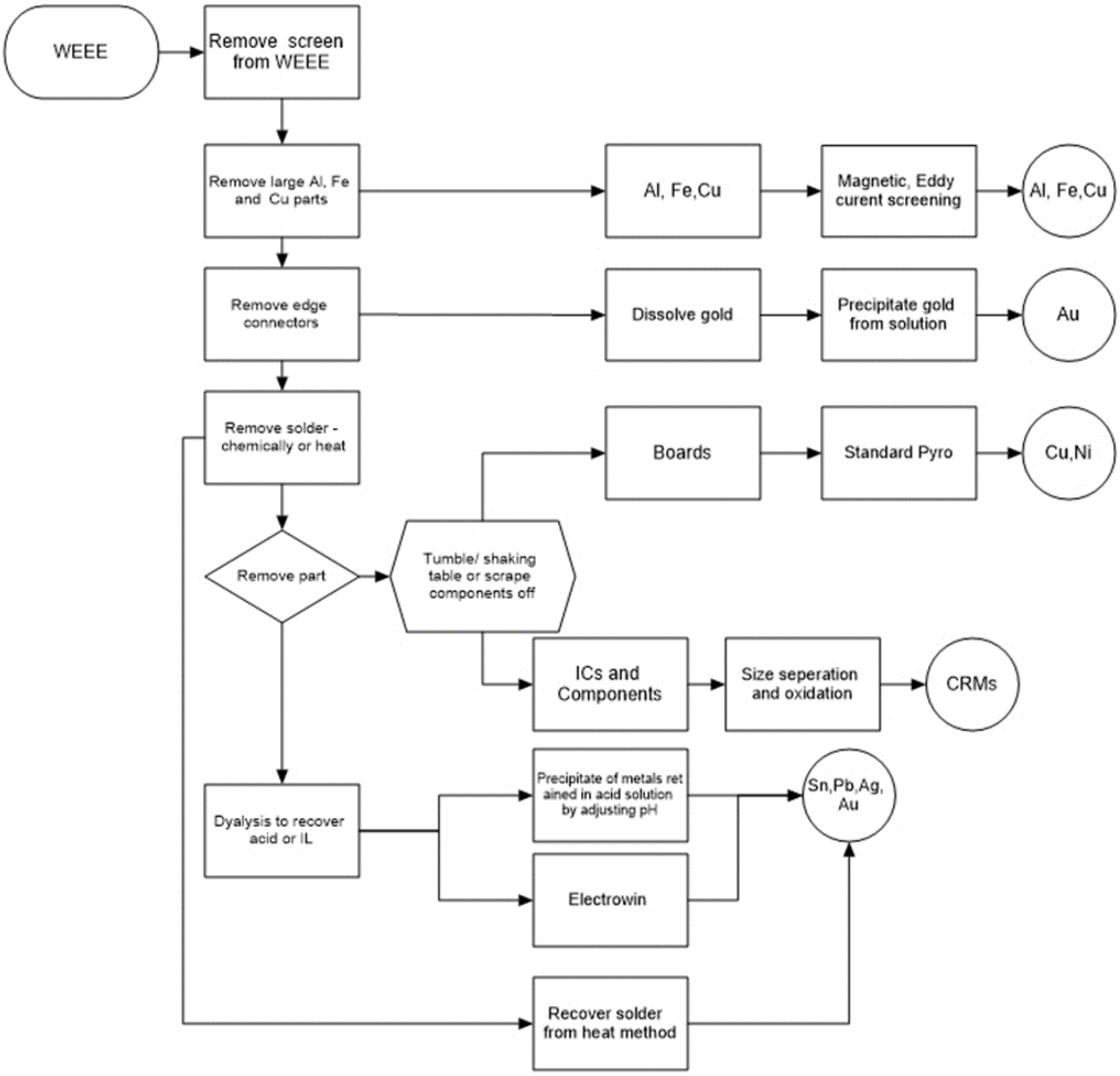
Leaching of technology metals from pre-processing outputs using hydrometallurgical methods followed by electrowinning is considered the most promising method for high recovery rates of materials. Electrolytic refinement has a high TRL and is already widely adopted in recycling. Proprietary systems for hydrometallurgical recovery of precious metals from PCBs are already available, and in operation around the world. Examples include the systems of EMAK, Mint Innovation, and Enviroleach which have focused recovery on PMs, PGMs, tin and copper but with further development could also recover a wide range of TMs. Adoption of such technology for processing PCBs in the UK would enable domestic recovery, allowing the pre-processing outputs and their contained technology metals to be retained within the UK. The small scale of these plants, compared to large pyrometallurgical facilities, and relatively low cost make them suitable for regional recovery hubs in the UK, potentially sighted close to industry that can make use of recovered materials.

### *9.2.1 Hydrometallurgical recovery system – TM recovery from PCBs*

Figure 24 shows a flow chart of a potential hydrometallurgical recovery system for treating PCBs, which makes use of acids or Ionic Liquid technologies available today. By way of example, this would involve the following process steps:

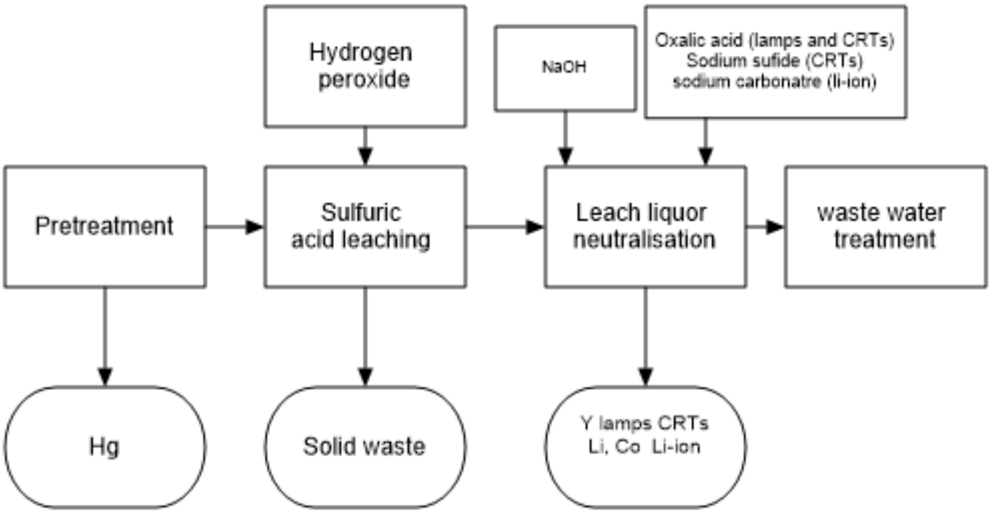
1. Remove any attached screen, followed by removal of large aluminium, ferrous or copper parts, such as heat sinks, brackets and transformers, for separate treatment;
2. Removal of edge connectors for treatment via digestion of gold, using acids or ILs, followed by recovery from solution by precipitation;
3. The solder can then be removed from the PCB, releasing the components through chemical digestion with acids or ILs, or by thermal treatment to melt the solder. These can then be shaken or scraped from the PCB for sorting and recovery;
4. Following digestion (dissolving of the metals) silver and tin in solution can be concentred in the acid using dialysis and precipitated, enabling the rest of the acid to be recycled in the system;
5. Depopulated PCBs are sent for copper and nickel recovery; and
6. The ICs and other components can then be treated for technology metal recovery, potentially in component specific processes which target all contained technology metals.

*Figure 24: Flow chart for Acid /Ionic Liquid (IL) PCB treatment for technology metal recovery.*



Once again, by combining existing technologies, recovery of TMs from lighting, LCDs and CRTs can be carried out with a similar process to that shown in Figure 25. This process uses a pre-treatment to remove any mercury (Hg) and then uses hydrometallurgical processes to leach the technology metals from the crushed lamps, LCDs and CRTs and the technology metals are precipitated from the acid solution. Dialysis can also be used in this process to concentrate the technology metals in solution for treatment and the rest of the acid recycled for reuse. The process is also suitable for recovery of lithium from batteries. This demonstrates the flexibility of hydrometallurgical plants.

Figure 25: TM recovery from lamps, LCDs, CRTs and Li-ion batteries.



Based upon the review of the different technology metal recovery and recycling opportunities and technologies to enable this recovery, there is a wide range of different options for recovering technology metals from WEEE which are discussed throughout the following sections and summarised in Table 16.

## 9.3 Alternative approaches to TM Recovery

The main purpose of this technology roadmap is to identify emerging technologies with potential to increase recovery of TMs from WEEE, enabling those seeking to develop next generation recycling strategies which are compatible with the circular economy. It is therefore a tool to simplify navigation and selection of the most appropriate technologies for given applications. This roadmap details the following:

- Material flow analysis (MFA) to quantify mass (tonnes), value (£) and environmental impact ($CO_2e$) of TM quantity and fate within WEEE;
- Recent research into processes for TM recovery from WEEE;
- Current state of the art TM recovery technologies, pilot plants and proprietary solutions;
- Technology readiness level (TRL) of the different developing technologies; and
- Practical considerations for industrial adoption, including potential cost factors and health and safety (H&S) considerations required for their integration into current recycling practices.

Technologies described in this roadmap are those which hold potential to overcome barriers to TM recovery, referring to >250 research publications. The wide range of technologies suitable for high levels of TM recovery will enable businesses to choose which would be most appropriate for the types of WEEE they wish to treat.

Each main process is given a technology readiness level (TRL) rating of 1 to 9 based upon the following criteria:

- TRL 1 – Basic principles observed
- TRL 2 – Technology concept formulated
- TRL 3 – Experimental proof of concept
- TRL 4 – Technology validated in lab
- TRL 5 – Technology validated in relevant environment (industrially relevant environment in the case of key enabling technologies)
- TRL 6 – Technology demonstrated in relevant environment (industrially relevant environment in the case of key enabling technologies)
- TRL 7 – System prototype demonstration in operational environment
- TRL 8 – System complete and qualified (proven to work as specified)
- TRL 9 – Actual system proven in operational environment

An overview of WEEE recycling technologies and their application for TM recovery is given below (Table 16), details of each of these is given in sections 11-16, and specific component treatments in the Appendices.

*Table 16: Overview of WEEE recycling technologies and their application for TM recovery.*

| Method and TRL | Description | Applications in WEEE recycling | Pros | Cons | Examples |
|---|---|---|---|---|---|
| **Pre-processing/pre-treatment** | | | | | |
| **Mechanical-physical separation**<br>Technology Readiness Level (TRL) 9. | Crushing and size reduction. Material separation according to physical properties.<br>Batch or continuous process. | Pre-processing and pre-treatment in recovery:<br>Size reduction and enrichment of target materials in output fractions for further processing. | Methods are well-established industrially; materials-specific plants are available for purchase.<br>Corona electrostatic methods are now capable of producing two streams from PCB waste comprising a metallic and a non-metallic portion with little cross-contamination.<br>Dry method at room temperature and as such is almost zero polluting depending on the quality of the dust extraction system.<br>Dust recovery for processing may be necessary, this is common practice at refineries that pre-treat PCBs via comminution prior to smelting to ensure TM losses are minimised. | TM-bearing PCB components may lead to complicated material composition in crushed PCB powder, which increases difficulty of material separation.<br>TMs are dissipated during shredding and lost through unintended co-separation.<br>Hazardous materials may be dispersed through bulk of shredded materials leading to contamination, reduced value and limits to potential safe applications of recovered materials. | Separation of metals and non-metals of PCBs by electrostatic or gravity separation.<br>Density separation of plastics, current collector metals and black mass of batteries. |
| **Steam gasification**<br>Technology Readiness Level (TRL) 4. | Pre-treatment: Method used to breakdown resins on boards and ECs to facilitate further hydrometallurgy processing.<br>Batch process. | Elimination of encapsulation polymers to expose metals for leaching. | Lab-scale method only but potential to scale up using existing equipment.<br>Industrial gasification systems for other applications are available.<br>Removal of polymeric materials avoids comminution and associated TM losses. | Fumes generated from resin breakdown requiring abatement.<br>Not commercially available. | |
| **Electrohydraulic fragmentation (EHF)** | EHF is a materials selective fragmentation technology. Electrical discharges between electrodes propagate through a surrounding | Contactless alternative to comminution, separation of materials at weak points such as | Enables isolation of components such as PCBs from mobile phones while avoiding TM losses incurred through traditional comminution. | Processed samples required drying.<br>Potentially a slow process due to drying. | Has been demonstrated successfully on battery cells, mobile phones, |

| Method and TRL | Description | Applications in WEEE recycling | Pros | Cons | Examples |
|---|---|---|---|---|---|
| Technology Readiness Level (TRL) 9. | carrier medium (water) and hit WEEE samples, causing breakage at weak interfaces allowing materials to be separated.<br>Batch process. | joints and materials interfaces. | Current research underway by Fraunhofer IWKS to enable selective disassembly of PCB components from boards.<br>Process has been commercialised for several applications by ImpulsTec GmBh as 'shockwave recycling'.<br>Fraunhofer IWKS offer support to optimize process for specific materials using their pilot plant, and support upscaling for commercialisation. | | PCBs, PV modules and Ta-capacitors. |
| **Underwater Explosion technique**<br>Technology Readiness Level (TRL) 5. | A small explosive is attached to an item and detonated with an electrical pulse once submerged in water, the shockwave created travels at different rates in dissimilar materials, and at material interfaces, causing them to break apart.<br>Batch process. | Contactless alternative to comminution for material and component separation. | Where excessive comminution needs to be avoided to prevent dispersion of hazardous materials or dissipative losses of TMs, technique offers a method to increase degree of material liberation and increase extraction efficiencies. | Materials need to be dried.<br>Consumes explosive materials and electricity.<br>Only developed to lab scale currently.<br>Potentially a slow process due to drying. | Demonstrated for rapid disassembly of mobile phones and other composite materials e.g., DVDs, tungsten carbide scrap and concrete slabs. |
| **Extraction** | | | | | |
| **Pyrometallurgy**<br>Technology Readiness Level (TRL) 9. | Extraction and purification of metals via the application of heat followed by electrorefining which uses electrolysis to separate the metals.<br>Batch process. | Smelting for recovery of non-ferrous and precious metals.<br>Pyrolysis (thermal decomposition of materials at elevated temperatures in | High recovery efficiencies of target metals.<br>Methods are well established and capable of high throughput.<br>Conversion of organics to syngas, or combustion and use as reductant in smelting offsets energy and raw materials demand. | To avoid serious pollution in pyrometallurgy, high-cost abatement plants for off-gases required.<br>High energy consumption.<br>Partitioning of TMs into slag phase during smelting is a barrier to TM recovery e.g., | Well-established smelting processes for PM, PGM & Cu recovery from PCBs; cathode metal recovery from Lithium-Ion Batteries (LIBs.)<br>Reduction of organic content prior to metals |

| Method and TRL | Description | Applications in WEEE recycling | Pros | Cons | Examples |
|---|---|---|---|---|---|
| | | an inert atmosphere), gasification (thermal breakdown of organic compounds such as the resin on PCBs) and calcination (a thermal treatment process in the absence or limited supply of air or oxygen to bring about a thermal decomposition) as pre-treatment for recovery to concentrate metals for recovery. | | Li from batteries (LIBs); Co, Ga, Ge, Y, Ta and W from PCBs.<br>Further hydrometallurgical refinement is necessary to separate the metals. | recovery from PCBs, treatment of WEEE plastics. |
| **Hydrometallurgy**<br>Technology Readiness Level (TRL) 9. | The use of chemicals to create aqueous solutions from which metals are recovered.<br>Batch process. | Extraction and recovery of all technology metals, dependent upon the chemicals used such as acids and Ionic Liquids (ILs). | More efficient process with potential to lower energy use and emissions to air compared to other recovery processes, such as physical separation and pyro-metallurgical processes.<br>Favoured over traditional pyrometallurgy due to an ability to recover metals from a variety of challenging sources, including low grade ores, mixed metal ores and complex material secondary sources such as WEEE. | Conventional industrial processing of WEEE requires aggressive acid or alkali digestion often with hazardous chemicals.<br>Use of corrosive inorganic acids requires costly acid proof plant.<br>Continuous consumption of chemicals can add cost. | Dissolution and refinement of metals from alloy output of PCB smelting.<br>Off the shelf hydrometallurgical PCB recycling systems are available e.g., EMAK and Enviroleach. |

| Method and TRL | Description | Applications in WEEE recycling | Pros | Cons | Examples |
|---|---|---|---|---|---|
|  |  |  | Allows the use of low process temperatures and recycling of reagents.<br>When attempting to leach trace amounts of metals such as PMs from PCBs, selectivity of traditional leaching systems is poor and large volumes of reagents are consumed in dissolution of other metals present e.g., Cu and Sn.<br>Selective leaching is possible through treatment in consecutive leaching systems, with careful selection of leaching agents, and selective recovery from mixed metal solutions.<br>Suitable for smaller scale local recovery, far lower capex than pyrometallurgical plants, making it easier and cheaper to install in existing ATFs and AATFs.<br>Far easier to expand capacity as required, compared with traditional pyrometallurgical plants. | Large volumes of waste effluent may require costly treatment. |  |
| **Biological leaching**<br>Technology Readiness Level (TRL) 9. | The extraction of metals through their solubility in inorganic and organic acids produced by bacteria.<br>Batch process. | Successful recovery of TMs from WEEE dust, lighting and NdFeB magnets. | Microorganisms generate weaker organic acids in-situ, avoiding the requirement to purchase strong inorganic acids for metal leaching, saving environmental impacts and costs of treating waste inorganic acids.<br>Potential exists to develop strains of bacteria to target specific metals and therefore perform selective leaching, thus minimising further treatment technologies and further reducing pollution.<br>The removal of the metallic components via bioleaching will then leave the non-metallic fraction for processing with | These methods tend to be slower than acid or ILs for TM leaching, unless they are agitated or stirred.<br>Not suitable for use in strong acid solutions which may kill microorganisms. | Pilot scale testing of Ta recovery from capacitors.<br>Recovery of metals from PCBs has been proven e.g., gold by Mint Innovation. |

| Method and TRL | Description | Applications in WEEE recycling | Pros | Cons | Examples |
|---|---|---|---|---|---|
| | | | relatively low contamination (vs traditional hydrometallurgy).<br>Low temperature extraction is possible, processes replicate natural biogeochemical processes and therefore lower environmental impacts. | | |
| **Electrochemical dissolution**<br>Technology Readiness Level (TRL) 4. | Differs from traditional hydrometallurgical leaching, as oxidising agents for dissolution of metals are regenerated at anode of an electrochemical cell, or samples for digestion can be connected to a cathode of an electrochemical cell for electrochemical reduction and dissolving the metals.<br>Batch process. | Digestion of metals. | Reduces use of reagents vs traditional hydrometallurgical leaching approaches.<br>Low temperature processes.<br>Can dissolve metals selectively by via adjustment of potential across cell.<br>Oxidising agents can be regenerated in the cell, reducing consumption of chemical leachants.<br>Use of Fe species as oxidising agents avoids use of toxic chemicals. | Electrochemical cells consume energy.<br>Some oxidising agents can be hazardous (e.g., $Cl_2$) | Electrochemical generation of $Cl_2$ for leaching of PMs and PGMs.<br>Electrochemical regeneration of $Fe^{3+}$ to selectively leach base metals from PCBs for lower cost subsequent extraction of noble metals. |
| **Recovery** | | | | | |
| **Electrowinning**<br>Technology Readiness Level (TRL) 9. | Recovery of metals from solution by electrochemical reduction of metallic anions to metals at electrodes. | Recovery of metals from solution. | Can be designed for specific TM recovery.<br>Selective recovery through control of reduction kinetics at anode via cell potential.<br>Extensively adopted throughout industry, off the shelf systems are available. | Requires additional energy in most systems but can be designed to be self-generating. | Widespread application in refining of copper from smelting. |
| **Solvent extraction** | Dissolution of materials and selective liquid- | Extraction of materials through dissolution for | Supercritical fluids may eventually offer an attractive separation process by selectively dissolving out fractions of PCB waste using | The solvent extraction process makes use of solvents which are | |

| Method and TRL | Description | Applications in WEEE recycling | Pros | Cons | Examples |
|---|---|---|---|---|---|
| Technology Readiness Level (TRL) 4. | liquid extraction from mixed metal solutions<br>Batch process. | refinement e.g., separation of the Ta, Ni and Fe from the component resins. | different solvents at room temperatures and effective solvent recovery systems would reduce pollution to almost zero levels, depending on the vapour recovery process. | potentially flammable, volatile and toxic. | |
| **Ionic liquid (IL) / deep eutectic solvents (DES) dissolution**<br>Technology Readiness Level (TRL) 4. | Ionic liquids (ILs) are salts which are liquid at room temperature and present interesting alternatives to traditional solvents as reaction media and extraction systems.<br>(DES) are a new generation of solvents that can offset the major drawbacks of common ILs.<br>Batch process. | Leaching and solvent extraction. | Can tune solvent system to selectively recover metals from complex metal mixtures with similar redox potentials in aqueous systems.<br>The advantages of ILs as a reaction medium compared to existing conventional solvents lies in their tuneability and potential environmental benefits.<br>Minimising waste generation and acid consumption whilst maintaining selective leaching.<br>Potential to circulate and recycle ILs in processes to offset high initial costs.<br>DES avoid toxicity, cost and environmental impacts of traditional ILs.<br>Potential exists to selectively dissolve and precipitate metals and organics (e.g., epoxy of PCBs) through careful temperature control of digestion processes – negates mechanical pre-treatment for PCBs.<br>TMs have been selectively recovered from superalloys with DES. | Significant barriers remain to the commercial use of ILs for treating WEEE. More focus is required to improve upstream separation of WEEE to generate specific TM-bearing components for recovery through selective, more efficient, ILs for metal recovery.<br>Some more commonly used ILs are toxic, not biodegradable, and have complex synthesis, requiring purification, adding to their environmental impact.<br>ILs have relatively high costs vs traditional organic solvents.<br>DES can be biologically derived (amino acid type compounds).<br>Technology Readiness Level (TRL) remains low. | |
| **Ion exchange** | Packed columns of ion exchange resins are used to selectively extract and retain ionic | Recovery of TMs from mixed metal solutions. | Enables selective recovery of low concentrations of TMs in mixed metal solutions, resulting from leaching and digestion of WEEE. | Requires specific pre-treatment of the column. More suitable for low metal content solutions. | |

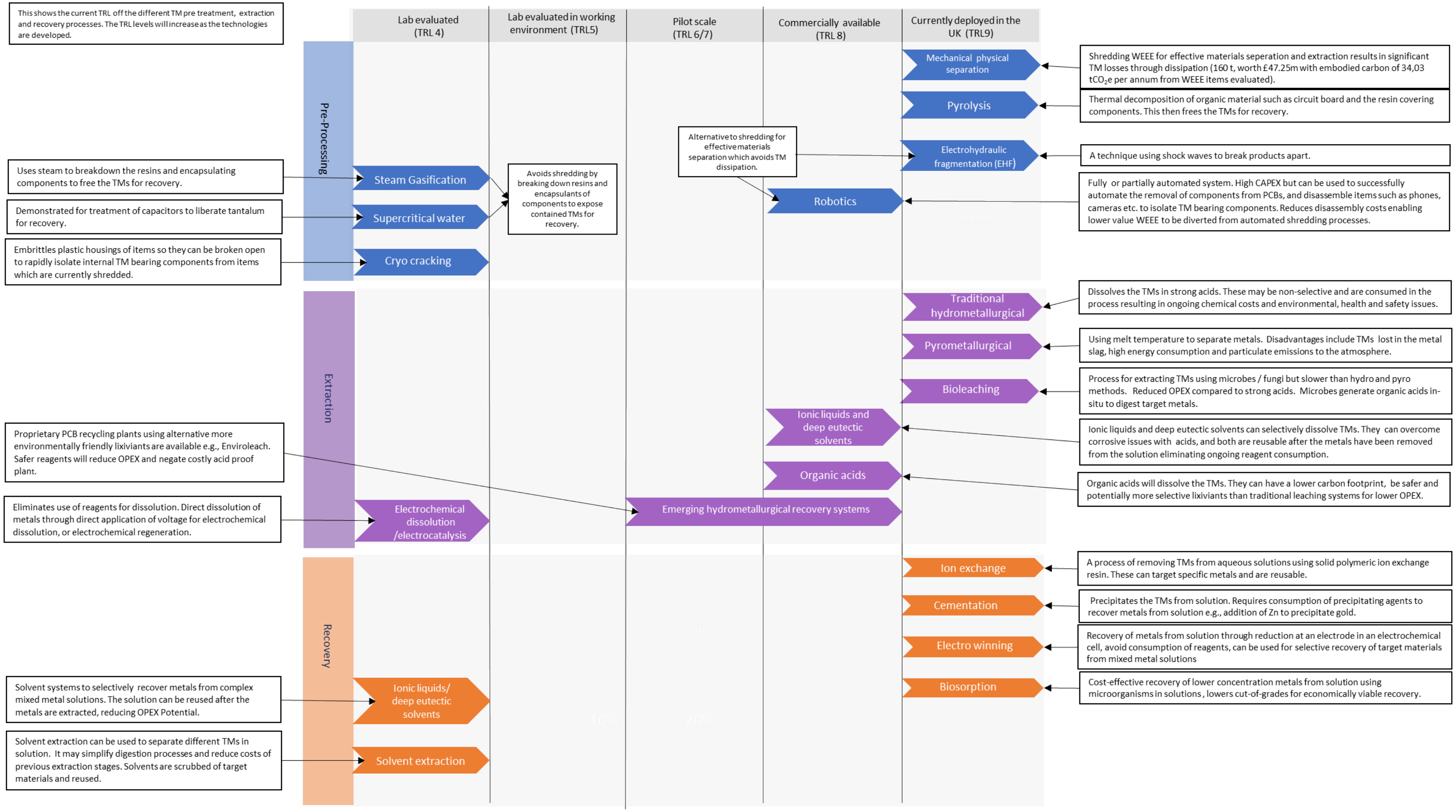
| Method and TRL | Description | Applications in WEEE recycling | Pros | Cons | Examples |
|---|---|---|---|---|---|
| Technology Readiness Level (TRL) 9. | species, such as TMs, from solutions passed through the column.<br>Continuous process. | | | | |

# 10 Technology Roadmap

A technology roadmap indicating the most promising technologies reviewed in this report, with their TRL levels, is shown in Figure 26. Brief comments on each of the different processes is given to highlight how they are used to overcome existing barriers to TM recovery. Due to the variation in the TRL levels and the unknown probability of future development, it is assumed that the lower the TRL level, the longer it would take to become a commercially available process.

All these processes could be incorporated into existing WEEE recycling plants, along with ancillary equipment such as extraction to deal with any dust or fumes generated from the processes. In particular, the hydrometallurgical extraction of technology metals from WEEE could be carried out via leaching in columns, with lixiviants flowing through them, or in tanks where the acid is stirred or circulated. The use of diffusion dialysis or counter-current leaching is strongly recommended in order to reduce the operational expenditure (OPEX), by reducing the consumption of the acids. Electrocatalysis to regenerate oxidising agents in solution, or electrochemical dissolution would also eliminate the need for ongoing consumption of reagents for digestion and extraction of technology metals from WEEE materials.

*Figure 26: Technology Metal (TM) recovery technology roadmap indicating the technology readiness level of most promising technologies identifies, and the stages to WEEE recycling to which they are applicable.*



This shows the current TRL off the different TM pre treatment, extraction and recovery processes. The TRL levels will increase as the technologies are developed.

Uses steam to breakdown the resins and encapsulating components to free the TMs for recovery.

Demonstrated for treatment of capacitors to liberate tantalum for recovery.

Embrittles plastic housings of items so they can be broken open to rapidly isolate internal TM bearing components from items which are currently shredded.

Avoids shredding by breaking down resins and encapsulants of components to expose contained TMs for recovery.

Alternative to shredding for effective materials separation which avoids TM dissipation.

Shredding WEEE for effective materials seperation and extraction results in significant TM losses through dissipation (160 t, worth £47.25m with embodied carbon of 34,03 $tCO_2e$ per annum from WEEE items evaluated).

Thermal decomposition of organic material such as circuit board and the resin covering components. This then frees the TMs for recovery.

A technique using shock waves to break products apart.

Fully or partially automated system. High CAPEX but can be used to successfully automate the removal of components from PCBs, and disassemble items such as phones, cameras etc. to isolate TM bearing components. Reduces disassembly costs enabling lower value WEEE to be diverted from automated shredding processes.

Dissolves the TMs in strong acids. These may be non-selective and are consumed in the process resulting in ongoing chemical costs and environmental, health and safety issues.

Using melt temperature to separate metals. Disadvantages include TMs lost in the metal slag, high energy consumption and particulate emissions to the atmosphere.

Process for extracting TMs using microbes / fungi but slower than hydro and pyro methods. Reduced OPEX compared to strong acids. Microbes generate organic acids in-situ to digest target metals.

Ionic liquids and deep eutectic solvents can selectively dissolve TMs. They can overcome corrosive issues with acids, and both are reusable after the metals have been removed from the solution eliminating ongoing reagent consumption.

Organic acids will dissolve the TMs. They can have a lower carbon footprint, be safer and potentially more selective lixiviants than traditional leaching systems for lower OPEX.

Proprietary PCB recycling plants using alternative more environmentally friendly lixiviants are available e.g., Enviroleach. Safer reagents will reduce OPEX and negate costly acid proof plant.

Eliminates use of reagents for dissolution. Direct dissolution of metals through direct application of voltage for electrochemical dissolution, or electrochemical regeneration.

A process of removing TMs from aqueous solutions using solid polymeric ion exchange resin. These can target specific metals and are reusable.

Precipitates the TMs from solution. Requires consumption of precipitating agents to recover metals from solution e.g., addition of Zn to precipitate gold.

Recovery of metals from solution through reduction at an electrode in an electrochemical cell, avoid consumption of reagents, can be used for selective recovery of target materials from mixed metal solutions

Cost-effective recovery of lower concentration metals from solution using microorganisms in solutions , lowers cut-of-grades for economically viable recovery.

Solvent systems to selectively recover metals from complex mixed metal solutions. The solution can be reused after the metals are extracted, reducing OPEX Potential.

Solvent extraction can be used to separate different TMs in solution. It may simplify digestion processes and reduce costs of previous extraction stages. Solvents are scrubbed of target materials and reused.

The increase in percentage of TMs being recovered in the roadmap is an average for all TMs. The actual level of recovery will be dependent upon numerous factors, including the pre-processing efficiency and losses of TMs in dust, efficiency of the leaching processes, the original concentration of the TMs in the WEEE, concentration of the acids and their selectivity, ILs and DES used and their selectivity, material throughput, length of time the parts are exposed to the acids, IL and DESs (dwell time) etc.

Installation of alternative processes to shredding for the breakdown of technology-rich parts and using hydrometallurgical processes, including the use ionic liquids and deep eutectic solvents, will significantly increase the potential to recover technology metals. Other options would be to use electrohydraulic fragmentations (EHF), steam gasification or supercritical water on circuit boards instead of shredding, otherwise most of the technology metals will be lost in the shredding dust.

Hydrometallurgical extraction (traditional, organic acids, ILs or DES) of technology metals from WEEE (with electrowinning or cementation to recover the technology metals) should, given the numerous factors previously mentioned, recover at least 75% of the contained technology metals.

The use of Biosorption, ILs and DES should also be reviewed as alternative technology metal recovery methods, bearing in mind that the ILs and DES are currently only at TRL 4. Biosorption will be useful for extracting low concentrations of technology metals in solution.

Based upon a conservative recovery and recycling rate of 75% of this mass of TMs, a total of 204.5t of PMs, PGMs and tin, worth £100.40m, could potentially be recovered and recycled in the UK using currently commercially available TM recovery, extraction and recycling plants. Due to the energy intensive processes associated with raw material extraction for primary material, the amount of $CO_2$ that mining for these same TMs would emit is estimated to be ~96,499 tonnes of $CO_2$e, though the $CO_2$ emitted by these advanced recovery processes themselves is not known.

In order to recycle the remaining 120.81t of TMs, with a value of approximately £12.95m, that are lost during shredding, lost abroad and recovered abroad, additional processing of the WEEE would be required

In general, setup and running costs for existing TM recovery facilities were not available. However, a capital expenditure of ~$2m to $2.5m (£1.4m to £1.8m) is thought to be indicative for a fully operational EMAK recovery and recycling plant (recovering PMs, PGMs, tin and copper) with a capacity to treat 1 tonne of PCBs a day (~250t per annum). Further investment will be required in the developing technologies identified by this research to recover the full range of TMs. At least 56% of the of £133.86m value of TMs in the WEEE covered in this research resides in the PCBs and an estimated 18 such plants would be needed to treat the estimated 4,500 tonnes of precious metal-bearing PCBs extracted from the computers, laptops, tablets, TVs and smart phones sent for recycling in the UK each year, with an estimated value in excess of £75m. This would require an approximate investment of ~£25m - £32m in TM recovery plant. Based upon approximate operational costs (as provided by EMAK) for chemicals, electricity and labour of £1,077 per tonne in a 1t a day EMAK plant, the cost to treat the 4,500t would be an estimated additional cost of £4.85m per year. Note that these costs do not include the cost of purchasing the WEEE to feed the plants, permitting costs and various other relevant overheads.

Although this would require significant investment by the WEEE recyclers or others, operational plants, such as that manufactured by EMAK (Table 22), are believed to be commercially viable where market conditions are favourable (e.g., gold bullion price).

As noted, additional specialised plant would be required to treat the other types of TM-bearing WEEE, such as lighting, display equipment and computer hard drives, in particular to focus on the recovery of the TMs beyond the PMs, PGMs and tin.

# 11 Taxonomy of emerging recycling processes for technology metal recovery from WEEE

We have seen that the potential to recover technology metals from WEEE and exploit significant economic and environmental opportunities for the UK, whilst minimising risks to businesses (such as those producing EEE, wind turbines and electric cars) sensitive to global resource security issues for technology metals is significant. However, to exploit these opportunities and generate domestic secondary sources of technology metals from WEEE, next generation recycling systems must be implemented which minimise losses of these materials during recycling as currently occur in both pre-processing domestically and foreign recovery processes; and minimise costs of recovery. New processes which target technology metals which today are unrecoverable due to techno-economic constraints must also be identified and adopted. This section gives an overview of emerging alternative technologies for application in pre-processing, extraction and recovery that meet these requirements. Subsequent chapters consider technologies which may be specifically applied in recycling of WEEE products or components identified as having a high potential for technology metal recovery.

Discussions of technologies here are limited to those the authors believe to be the most promising for application in technology metals recovery from WEEE for the UK. These have been extracted from a review of >250 research papers and technical reports on current and emerging technologies for WEEE recycling, as well as interviews with researchers working in the field of WEEE recycling and recycling equipment manufacturers, conducted to determine available options for application in each stage of the recycling process chain. However, the composition of the waste stream changes over time. Technological innovation and obsolescence, thrifting in manufacturing and changes to product design, as well as consumer behaviours, all mean that the nature of the occurrence of technology metals in the future, and the appropriate means of their recovery may alter. For this reason, a detailed taxonomy of all technologies reviewed can be found in the following sections and in Appendices B and C, including further details on technologies and processes according to the stages of recycling to which they apply; type of technology; types of WEEE/components they treat; and target technology metals.

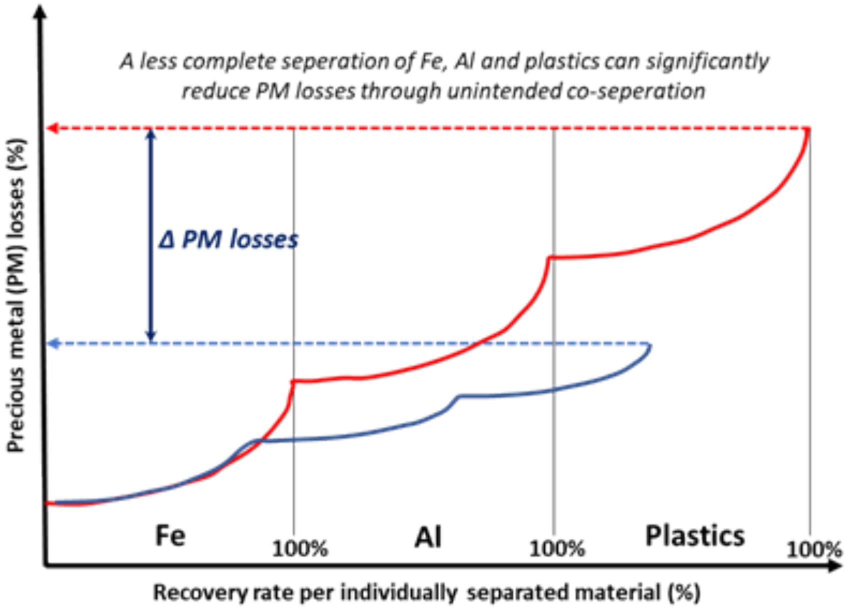
Higher value can be recovered from items with high concentrations of precious metals in their PCBs with manual processing than through automated processes, justifying higher process costs. Manual processing also gives the opportunity to isolate components in which small amounts of TMs are present and items suitable for reuse.

Automated recycling involves shredding items (following depollution) with subsequent automated separation of material fractions i.e., magnetic, eddy-current, electrostatic and density separation, to give ferrous, aluminium, stainless-steel, glass, copper, brass and plastic fractions.

As target material content is concentrated in a material flow by separation of non-target materials, the overall recovery yield diminishes. This is because shredding disperses particles of trace amounts of precious (and critical) materials present throughout the mass of the shred, where they adhere to the surfaces of larger particles. At each stage in separation, these metals are lost through unintended co-separation with bulk fractions. Recovery processes for these bulk fractions do not target technology metals and so these are permanently lost.

From a resource-efficiency point of view, such processes should be avoided where possible in favour of semi-automated or manual pre-processing. Grinding also results in dispersal of hazardous materials throughout larger mass fractions, resulting in higher potential toxicity of output fractions than WEEE items prior to shredding. The 'Grade-Recovery Function' which results in losses of target precious metals and other trace elements as larger bulk fractions are removed from shredded WEEE in automated pre-processing is shown in Figure 27.

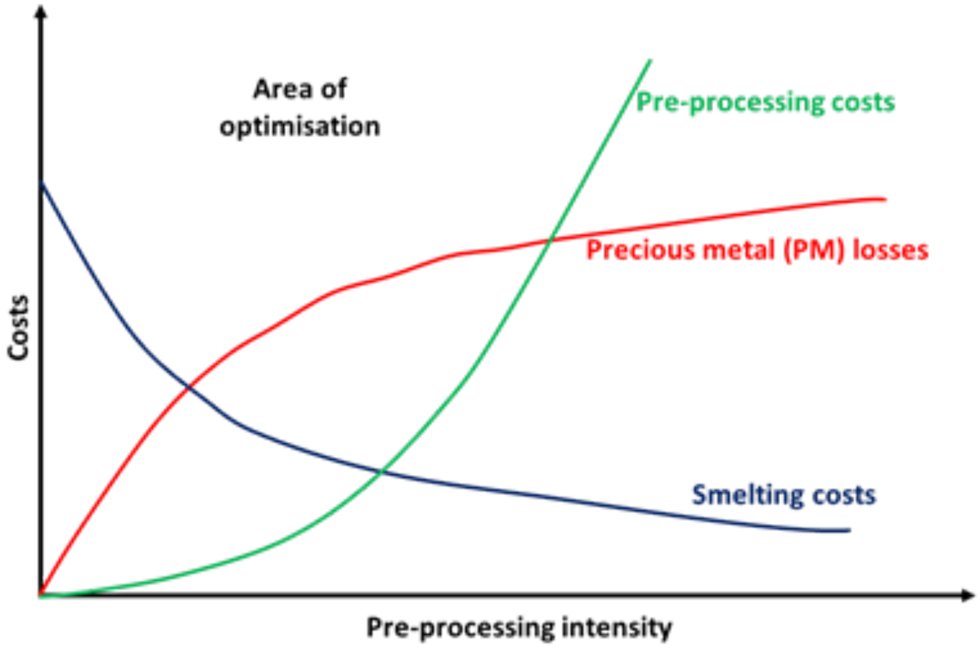
*Figure 27: The 'Grade-Recovery Function' which results in losses of target precious metals and other trace elements as larger bulk fractions are removed from shredded WEEE in automated pre-processing, (Hagelüken, 2006).*



The choice of process is largely determined by the recoverable material value of items. Low value WEEE with low precious metal content (e.g., toasters, hair dryers etc.), is recycled with automated processes, and items with high inherent value (e.g., PCs) are processed manually.

If the grade of a PCB is sufficiently high (i.e., above the cut-off grade) it will generally undergo recovery in pyrometallurgical smelting processes to recover Cu bullion, rich in PMs and PGMs, which are subsequently refined electrolytically. There are economic and thermodynamic limitations to viable recovery of other TMs in this process. The way in which metals partition between the liquid copper and slag phases during smelting is important in determining recovery potential (Reck and Graedel, 2012). Metals which do not dissolve in liquid copper partition into slag, from which their recovery is uneconomical. These include Co, Ga, Ge, Y, Ta and W. (Wan *et al.*, 2018; Sukhomlinov *et al.*, 2019; Klemettinen L, Avarmaa K, 2017). The qualitative cost function in WEEE recycling interface optimisation is shown in Figure 28.

*Figure 28: Qualitative cost function in WEEE recycling interface optimisation (Hagelüken, 2006).*



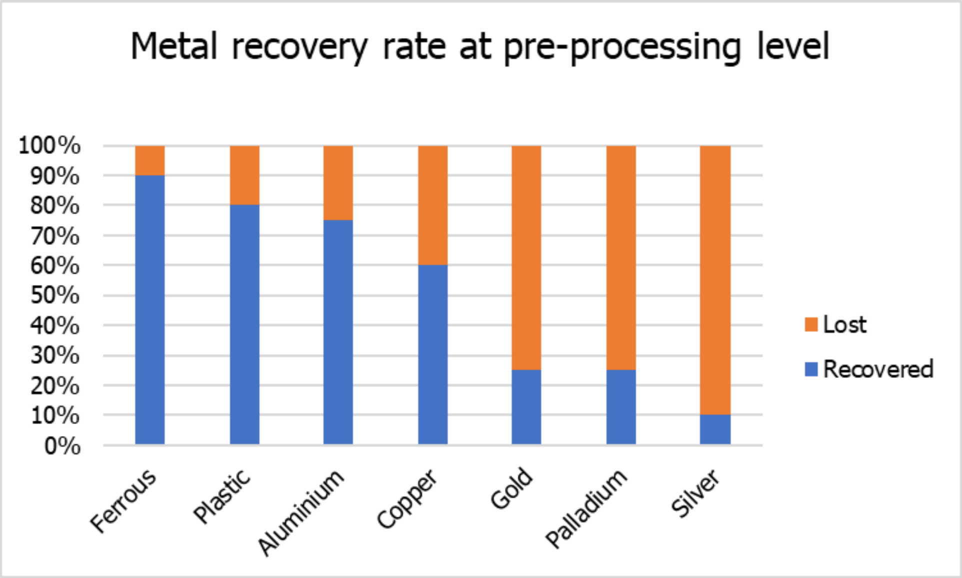
Despite high recovery efficiencies for target metals in these processes, many of the other materials present are not recovered, either because they are of low value and therefore recovery is uneconomical, or there are thermodynamic limitations to recovery e.g., tungsten which is present in ICs as an interconnect material is lost in slag during smelting.

Processors of WEEE often do not know the TM content of their incoming WEEE streams as there are no standardised methods for carrying out assays of these streams for recovery, further inhibiting their willingness to process unknown input streams. The only way of determining the recovery rates is to carry out a detailed material flow analysis (MFA), to determine the metals that were lost in the processing.

A report on the CRM losses across the different WEEE streams (Parker and Arendorf, 2012a) throughout the recycling process chain was due as inappropriate pre-processing as a major cause of CRM dissipative losses.

There are also losses of all of the material in WEEE, but in particular the metals at trace levels (Rotter, 2011) (Chancerel *et al.*, 2009). The graph below (Figure 29) shows the potential material losses at the pre -processing stage.

Figure 29: Metal recovery rate at pre-processing level.



TMs, due to their small concentrations in WEEE, are irrelevant to meeting the current collection and recycling targets, as the mass targets will be met by bulk materials recovery e.g., steel, aluminium, copper and glass. Although TMs may be economically, environmentally, and strategically important to the economies of the world, they contribute little towards mass-based recovery efficiencies when used only in trace amounts within EEE. The amounts vary depending upon the type of EEE (Batinic *et al.*, 2018), for example PMs account for a maximum of 0.01% in mobile phones and Eu, La and Ce account for less than 0.01% each in fluorescent tubes.

## 11.1 Pre-processing technologies

WEEE is an extremely heterogenous combination of materials. The pre-processing involves material separation and size reduction, prior to processing for TM recovery. This will enable effective TM recovery at minimal cost, as discussed in Chapter 4. Losses of target materials when reducing size through shredding must be stemmed, and costs of pre-processing must also be minimised for the overall cost-benefit of technology metal recovery. The following sections discuss approaches to achieving this.

### *11.1.1 Cryocracking*

Cryocracking has been demonstrated as a method to augment manual disassembly processes, for rapid disassembly of mobile phones, MP3 players and digital cameras with plastic housings which are prohibitively time consuming to manually disassemble and access internal technology metal-rich PCBs, screens and other components (Charles, Douglas, Dowling and Davies, 2020). Cryogenic cooling to embrittle plastics is common in numerous applications. It has been found that by submersion of the WEEE items in liquid nitrogen for 20 seconds to embrittle plastic housings before striking with a hammer, housings can be shattered to enable collection of internal components. Due to difficulty with manual disassembly, these items are usually shredded or charged whole to PCB smelting processes, incurring TM losses previously discussed.

It was proposed that cooling with liquid nitrogen to -178 °C may be unnecessary; investigation of embrittlement temperatures of commonly used WEEE plastics (e.g., PC-ABS, HIPS) may enable less costly alternative cryocracking methods, e.g., using solid $CO_2$. Industrial scale-up may be possible by freezing multiple items simultaneously and feeding them into an automated process to shatter casings which does not damage components; a jaw crusher may be a good solution.

### *11.1.2 Mechanical and thermomechanical processes*

Although mechanical processes, such crushing and shredding, as employed currently, often result in dissipation in technology metals, there are advantages of their use in numerous applications. The low OPEX of such processes compared to manual approaches reduces costs of recycling and if losses of technology metals can be stemmed when applied appropriately, they hold the potential to increase technology metal recovery.

Mechanical coarse grinding of WEEE with hammer mills to break open housings and access PCBs whilst attempting to minimise damage and dissipation of technology metals is used, but material flow analysis (MFA) studies have shown bottom-line recovery efficiencies of Au from ICT equipment using this method to be as low as 70% (Hagelüken and Meskers, 2013). These losses, although undesirable, are considerably less than is achieved when items are committed to regular comminution processes for automated pre-processing. Tsunazawa *et al.* evaluated comminution of smartphones, microwave ovens and rice cookers, with a drum-type agitation mill to liberate PCBs whilst minimising damage to them (Tsunazawa *et al.*, 2018). Process simulation using discrete element method (DEM) with a particle-based rigid-body model allowed equations to be developed which determine optimum process parameters, such as feed rate and rotational speed, to minimise PCB breakage while achieving good separation from appliances. This kind of modelling in combination with mechanical treatment has good potential to reduce technology metal dissipation during WEEE recycling, especially if it can be conducted for plant already in place, removing CAPEX barriers to adoption that investment in new plant for this purpose would create.

Thermomechanical methods of treating PCBs in order to upgrade them for subsequent recovery are also emerging commercially (Table 17). Such methods use mechanical abrasion with heating to soften solder securing components to PCBs in order to liberate technology metal-bearing components from the large mass, low value portions of the PCB i.e., the board itself and other larger mechanically fastened components such as brackets and heat sinks. Where PCBs fall below COGs for recovery, concentrating the value in this way will reduce the refining costs, whilst delivering all contained target metals for recovery without losses.

*Table 17: Thermomechanical technologies for depopulation of PCBs.*

| CRM recovery | Application | Technology Readiness | Reference |
|---|---|---|---|
| Pre-treatment of CRM bearing components on PCBs. | Removal of solder using heat, centrifugal separation and vacuum pyrolysis. | Lab trial. TRL 4. | (Zhou and Qiu, 2010) |
| Pre-treatment of CRM bearing components on PCBs | Removal of solder using heat, centrifugal separation and vacuum pyrolysis. | Lab trials. TRL 4. | (de Castro *et al.*, 2019) |
| Pre-treatment of CRM bearing components on PCBs | PCB disassembly apparatus includes infrared (IR) heating with wire brushes for mechanical abrasion. | Lab trials/pilot TRL 5. | (Park *et al.*, 2015) |
| Pre-treatment | Removal of components from PCBs using different heat and mechanical methods. | Equipment readily available. TRL 4 for trialling | (Charles et al., 2019) |
| Pre-treatment of CRM bearing components on PCBs | This system contains two parts: PCBs automatic disassembly and off-gas purification. | Pilot scale. TRL 5. | (Wang, Guo and Xu, 2016) |

Referring to Table 17, the first two processes heat the PCBs in a chamber up to the melt temperature of the solder and the boards are then spun to remove the solder, which frees the components.  The third process also heats the boards but then applies a brush to abrade the board releasing the components. The fourth process investigated a variety of different processes including using a solder bath and a heat gun to remove the solder from the PCBs.  The last process used a heated rotating drum set at an angle to drain off the heated solder and any off gassing was treated before being released.

### *11.1.3 Robotics*

Robotics present automation options for rapid throughput and cost-effective disassembly, which avoid TM dissipation. Although high CAPEX is required, low OPEX and rapid-cost effective disassembly possibilities make such technologies a promising option for the UK where manual disassembly is prohibitively costly for many devices with high technology metal content. By way of example, Apple have introduced 'Daisy' – successor of 'Liam the robot' for disassembly of devices received through their product take-back program, utilising in-house knowledge of manufacturing processes to 'inverse manufacture' devices into components (Apple Inc., 2019). The challenge of utilising robots on a general recycling line is non-trivial due to a lack in uniformity of device types and models for processing. Kopacek and Kopacek have developed robotised, semi-automatised, flexible disassembly cells for mobile phones which are directed by an image analysis system to identify the phone type and link to a database of disassembly information based on previous disassembly studies of the most common mobile phone models (Kopacek and Kopacek, 2006).

Current technical problems in identification, classification, disassembly and manipulation of WEEE could be tackled by a combination of robotised and manual operations, where a human teaches a robot where to cut and separate parts, and the machine performs the operations, an approach which has been demonstrated for disassembly of TVs, monitors, and PCBs (Alvarez-de-los-Mozos and Renteria, 2017). Advances in AI and machine learning, combined with image recognition technology, have enabled automated robotic processing of nuclear wastes, suffering the same issue of non-uniformity as WEEE, and transfer of this technology for WEEE disassembly is being explored by the CrEAM network in the UK (Loye, 2018). Another effective approach to automation of pre-processing of non-uniform devices is incorporation of RFID tags in EEE to enable rapid online product identification and quick access to stored information, which directs disassembly and further processing (Kellner, 2009a; Loye, 2018; Ryen, Babbitt and Babbitt, 2018).

### *11.1.4 Underwater explosion technique*

An underwater explosion technique has been used by Matsuo *et al*. for rapid disassembly of mobile phones and separation of composite materials (MATSUO *et al*., 2011). A small explosive is attached to an item and detonated with an electrical pulse once submerged in water, the shockwave created travels at different rates in dissimilar materials at material interfaces causing them to break apart. This represents a contactless alternative to comminution for disassembly and separation of dissimilar materials in components which may otherwise face the prospect of grinding and technology metals losses. Where excessive comminution needs to be avoided, this technique offers a method to increase the degree of material liberation and increase extraction efficiencies.

Disadvantages of the process are that materials may need to be dried prior to further processing, it consumes explosive materials and electricity, and to date, it has only been applied to WEEE at lab scale. Due to the flexible nature of the process, it may be applicable both in pre-processing stages of recycling for materials separation, and for upgrading or separation processes to maximise technology metal recovery efficiencies. The application of explosives to individual items may however be manually intensive and prohibitively costly to apply at scale.

### *11.1.5 Electrohydraulic fragmentation (EHF)*

EHF is a materials-selective fragmentation technology (Nevala *et al*., 2019), where shock waves generated by pulsed voltage spark discharges between electrodes propagate through a surrounding carrier medium (water) and hit WEEE samples, causing breakage at weak interfaces and allowing materials to be separated. As with the underwater explosion technique, this too is a contactless alternative to comminution, causing separation of materials at interfaces and weak points such as joints and materials interfaces. The process has been developed by the Fraunhower IWKS in Germany where a pilot plant is operational, which has been successfully used for isolation of components such as PCBs (from mobile phones and otherwise), battery cells, PV modules, and liberation of tantalum from its plastic encapsulation in capacitors, which would otherwise prevent effective extraction of Ta in recovery (WRAP, 2018; Nevala *et al*., 2019; Charles, Douglas, Dowling, Liversage, *et al*., 2020; Mossali *et al*., 2020). This process avoids technology metal losses incurred through traditional comminution.

Current research is underway by Fraunhofer IWKS to enable selective disassembly of PCB components from boards, presenting not only a means to depopulate PCBs, but also to do this in a selective manner. EHF has been commercialised for several applications by ImpulsTec GmBh as 'shockwave recycling'. Fraunhofer IWKS also offer support to optimize the process for specific materials using their pilot plant, and support upscaling for commercialisation. Materials may require drying following this process, depending on the nature of the process to which they will be directed on to. Unlike underwater explosion technique, this process does not consume explosive materials, and relies only on supply of electricity to function. Additional cost-benefits are likely to result from the fact that WEEE can be charged to the system without careful application of explosives, for bulk batch treatment.

Due to the nature of this process relying on differences in physical properties of materials, it is potentially suitable for treatment of WEEE in pre-processing as an alternative to automated pre-processing, yielding disassembled components with limited technology metal losses. Additionally, it has already been successfully applied as a pre-treatment for materials recovery. At a high technology readiness level (TRL) of 8 and with support of the inventors to optimise the process for any feedstock using the pilot plant and scale up for commercialisation, this is one of the most promising technologies for adoption to stem pre-processing losses of technology metals.

### *11.1.6 Supercritical Water (scH$_2$O)*

Beyond the critical point (>647.096 K and >22.064 MPa) water is supercritical (scH$_2$O), existing as small but liquid-like hydrogen-bonded clusters, dispersed within a gas-like phase and it is an excellent solvent for non-polar molecules (a nonpolar molecule has no separation of charge, so no positive or negative poles are formed, in other words, the electrical charges of nonpolar molecules are evenly distributed across the molecule), due to its low relative permittivity (dielectric constant) and poor hydrogen-bonding. scH$_2$O has been used to breakdown the organic resins covering capacitors into smaller particles or powders.

Research on time, pressure and addition of $H_2O_2$ and its impact on breaking down the resins was reported by Niu *et al.* (Niu, Chen and Xu, 2017). This showed successful separation of the Ta, Ni and Fe from the resins after 1.5 hours. This method holds great potential for liberation of metals from plastics without incurring technology metal losses to the extent seen in current pre-processing operations. Extensive particle size reduction for effective extraction of metals from components can be avoided through application of this process. The batch process does require elevated temperature and pressure, which add to OPEX, and a specialised plant for the process is required. However, reaction rigs for supercritical fluids are commercially available and widely used, for example, for the freeze drying of coffee using supercritical $CO_2$.

### *11.1.7 Steam Gasification*

Gasification is a process that enables complete elimination of organic fractions (such as the resins in circuit boards and those covering components) to the gaseous phase at low oxygen potential of the system. Typical gasifying reagents are steam, oxygen (air, oxygen enriched air) or carbon dioxide. Steam gasification has been shown to break down organic resins, leaving WEEE with open inner structures which could facilitate access to internal technology metals and technology metal-bearing components for recovery (Gurgul, Szczepaniak and Zabłocka-Malicka, 2018) using acids or ILs. This again presents an option for rapid product disassembly, which avoids shredding, and also presents an option for pre-treatment of components in which technology metals are encapsulated within plastics, preventing efficient extraction for recovery.

The gaseous product of the process may, after cleaning, be used for energy recovery or chemical synthesis, presenting an opportunity to valorise the plastic fractions which may be problematic to recycle due to high flame-retardant content, or those plastics present in components such as those on PCBs which are consumed in pyrometallurgical recovery, and which necessitate grinding of PCBs before recovery to ensure efficient extraction. Although there are currently limited examples of the application of this technology in WEEE recycling, there is great potential for its application in pre-processing of PCBs and components as it offers fast product disassembly and avoids the need of shredding.

### *11.1.8 Vacuum Pyrolysis*

Where it is necessary to separate plastics from metals prior to recovery, pyrolysis presents interesting options for pre-treatment as it allows plastic fractions to be removed whilst leaving metallic fractions and glass behind which can be treated for recovery. Numerous processes for treatment of PCBs have been developed, which enable technology metal recovery from the outputs, without the need to grind boards to sufficiently small particle size. Vacuum pyrolysis heats materials under vacuum in the absence of oxygen, which causes organic parts to volatilise.

The products of this process are oils and gases that can be valorised as fuels, leaving behind metallic parts for separation and further treatment. Pyrolysis has been advocated as the most promising option for delamination of PV panels to harvest internal components for reuse, by decomposing the thermal polymers sealing modules, that would otherwise require shredding for treatment, without the ability to degrade these organic components (Farrell *et al.*, 2020).

Pyrolysis could improve the economics of recycling by valorising plastic fractions that are currently lost in recycling, it also holds the potential to concentrate target materials prior to shipping for recovery, thereby improving logistics costs and reducing refining charges. Numerous pyrolysis systems are commercially available for treatment of plastics and tyres, with support from manufacturers in commissioning of the systems. Therefore, this is a process that could be deployed and adopted quickly for pre-processing of suitable WEEE. Downsides include the energy needed and the requirement to treat gases produced by the pyrolysis.

## 11.2 Extraction technologies

Given the limitations of pyrometallurgical extraction treatment (only exposes the TMs that would require further extraction treatment), hydrometallurgical approaches to extract technology metals from WEEE-derived materials are a more promising option. Traditionally hydrometallurgy uses various leaching systems (lixiviants) such as strong acids, caustic solutions, or microorganisms to dissolve specific metals (Table 18). Here we discuss the relative pros and cons of different leaching systems for application to technology metal recovery, highlighting some of the most promising emerging systems.

Hydrometallurgy introduces opportunities for sequential leaching of metals following exposure of WEEE materials to different leaching systems which are selective for metals. This helps to reduce costs associated with consumption of more costly strong oxidizing materials for leaching of noble metals by removing the relatively high level of base metals with an alternative leaching agent and can simplify recovery by avoiding generation of such complex metal solutions as would be achieved with single digestion for all metals present (Sun *et al.*, 2017).

### *11.2.1 Pros and cons of leaching solutions*

The main issues in using strong acids or alkalis are around health and safety and the disposal of the spent liquids as well as the additional cost of acid proof plant. Although traditional systems such as aqua regia, see table below, are capable of efficient and rapid dissolution of gold and numerous other technology metals present in WEEE, their use and ongoing consumption in leaching processes is costly, and the hazards associated with them are undesirable, adding to overall recycling costs and raising COGs for recovery. For this reason, research into leaching of technology metals from WEEE has shifted to the use of organic acids, ionic liquids, and bioleaching alternatives. Numerous proprietary hydrometallurgical treatment systems for PCBs are now commercially available (discussed in Chapter 13.3), and as will be shown, these are now beginning to emerge, using these alternative leaching solutions.

The pros and cons of different leaching systems employed for technology metal recovery from WEEE are shown in Table 18 below.

*Table 18: Pros and cons of different leaching systems employed for technology metal recovery from WEEE.*

| Reagents | Pros | Cons |
|---|---|---|
| Sulfuric acid ($H_2SO_4$) | • Strong proton donor (strong acid).<br>• Lower cost compared to other inorganic/mineral acids.<br>• Highly selective, useful for sequential leaching to strip out base metals before alternative more costly lixiviants are used to extract precious metals from PCBs. | • Requires heating for efficient extraction.<br>• Corrosive<br>• Need follow up with thiourea as a lixiviant (liquid medium used in hydrometallurgy to selectively extract the desired metal) to recover the metals present in PCBs |
| Aqua Regia (AR)<br>($3HCl+1NHO_3$ mixture) | • Fast kinetics (fast acting).<br>• Effective at dissolving metals, including gold. | • High reagent cost<br>• Highly corrosive.<br>• Low selectivity (will dissolve a wide range of metals)<br>• NOx emissions generated |
| $NHO_3$ 3 moles per litre with HCl 0.05 moles per litre | • Fast kinetics (fast acting), Effective at dissolving metals. | • High reagent cost.<br>• Highly corrosive.<br>• Low selectivity (will dissolve a wide range of metals).<br>• NOx emissions generated<br>• Ammonium sulfamate addition needed to stop the reaction heating up too much and producing NOx and dissolving the copper. Ferric Nitrate added to stop corrosion. |
| Fluoroboric acid ($HBF_4$) | • Rapid selective dissolution of solder.<br>• Addition of $H_2O_2$ aids leaching. | • Not trialled on TMs. |
| Hydrofluoric acid | • Effective at dissolving Ta which is insoluble in alternative powerful oxidising acid systems e.g., aqua regia. | • Not trialled on other TMs.<br>• Extremely toxic requiring specialist health and safety measure. |
| Thiourea (+ $Fe^{3+}$)<br>$SC(NH_2)_2$ | • Low toxicity, noncorrosive, high dissolving power. | • High cost, difficulty in downstream metal recovery. |
| Thiosulphate<br>$(NH_4)_2S_2O_3$<br>$Na_2(S_2O_3).5H_2O$. | • High selectivity, nontoxic, noncorrosive, fast leaching rate. | • Oxygen carrying catalyst is required<br>• pH sensitive<br>• low stability<br>• high cost<br>• high reagent consumption |
| Ionic liquids | • Thermally stable.<br>• Less hazardous than acids. | • High initial cost (although can be recycled in processes).<br>• Requires high dosage rate. |
| Supercritical water | • Rapid breakdown of resin covering EC to liberate internal technology metals. | • High temperature and pressure requiring specialized equipment. |

### *11.2.2 Sulfuric acid and thiourea*

Sequential sulfuric acid followed by thiourea leaching of PCBs and lighting, following mechanical pre-treatment, has enabled recovery of over 90% of technology metals present. These included Y, Au, Ag, Li, Co and In (Amato *et al.*, 2019). A similar strategy was used by Marra *et al.* to recover TMs from WEEE dust arising from shredding (Marra, Cesaro and Belgiorno, 2019). Using sulfuric acid, most rare earth elements in the dust were extracted with high efficiency, and 50% of gold was extracted in a second leaching step using 0.25 M thiourea solution. Although the extraction of gold with the parameters used is low, this shows that technology metals entrained in dust from shredding processes can be viably recovered with hydrometallurgical approaches, helping to stem the losses of technology metals occurring in automated pre-processing. Zhang *et al.* reported alternative methods for recovering gold using this same strategy i.e., initial leach with sulfuric acid, to minimise costs of thiourea in subsequent leaching, on other methods using sulphuric acid and thiourea leaching with recovery of >90% for gold present in the feedstock (Zhang *et al.*, 2012), showing the potential of low toxicity thiourea for application to extraction of gold with high efficiency.

Lister *et al*. have proposed a plant for recovery of REMs, base metals and precious metals from WEEE in which sulfuric acid is used to leach REMs from ground feedstock following magnetic separation of ferrous materials, prior to subsequent leaching steps to remove other metals sequentially (Lister *et al.*, 2016).

### *11.2.3 Chloride acid media*

Aqua regia is a traditional leaching system which uses a combination of nitric acid (HNO3) and hydrochloric acid (HCl) in a 3:1 molar ratio to leach metals. It is one of the most traditional leaching systems for gold and PGMs, owing to the power of the system to oxidise the highly noble metals, and solvate them as chloride complex anions (e.g., chloroaurate – [AuCl4]— or chloroplatinate [PtCl6]2-). Aqua regia is the leaching system used for EMAKS's proprietary off-the-shelf PCB recycling system (discussed in Chapter 13.3.1). Hursthouse et al tried a variety of acid leachates and found that hydrogen peroxide with aqua regia digestion at 95°C was determined to be the optimum digestion method for WEEE waste (Hursthouse *et al.*, 2018). Aqua regia produces toxic NOx gas during its reactions which require an abatement plant in commercial processes adding to process costs.

The toxic effluent resulting from processes that utilise aqua regia also need to be managed appropriately before they can be discharged. The reagents are also consumed in digestion and are not regenerated upon recovery of metals from solution. It is for this reason that in PGM refining, they utilise an HCl system through which chlorine gas is bubbled to digest the metals producing chloride complexes in solution, but recovery results in regeneration of these species which can be cycled in the process. NOx gas generation is also avoided. The advantage of these systems is their rapid and efficient dissolution of these noble metals, so that high throughput of WEEE materials can be accommodated.

### *11.2.4 Fluoroboric acid*

Fluoroboric acid ($HBF_4$) and hydrogen peroxide ($H_2O_2$, 30%) was successfully used to selectively dissolve solder on PCBs to liberate components for further processing without dissolution of copper (Park *et al.*, 2015; Zhang *et al.*, 2015). 2.5 mol/L $HBF_4$ achieved total dissolution of solder in 35 mins, providing a promising approach to disassembly of components from PCBs to concentrate value and improve cost benefit or technology metal recovery. $HBF_4$ should have a lower environmental impact than alternative leaching systems, and its application to pre-processing of PCBs could be a promising approach to increase technology metal recovery from PCBs with appropriate treatment of isolated components.

### *11.2.5 Citric acid and lactic acid*

Organic acids have lower environmental impact than traditional leaching systems. The necessity for benign recycling methods is important to limit environmental impacts of recycling processes and to maximise the overall environmental benefits of technology metal recycling from WEEE. Organic acids alleviate the need for high temperature and oxidative acid leaching conditions. Citric acid in combination with $H_2O_2$ has been used at lab scale to recover lithium and cobalt from Li-ion batteries (LIB) cathodes, and gallium and indium from waste Copper Indium Gallium DiSelenide (CIGS) PV panels (Amato and Beolchini, 2019). Environmental assessments of both processes confirm the relatively low environmental impacts of the citric acid system, with a carbon footprint of up to 90% lower than other acids. Its applicability in these cases to recovery of these metals lead us to conclude that it may find application in treatment of other WEEE components containing these metals e.g., LCD glass for indium recovery. Lactic acid too as an alternative to friendly organic acid, has been used in combination with $H_2O_2$ for recovery of technology metals from LIBs (Meshram *et al.*, 2020a).

Citric and lactic acid have also been used in combination with deep eutectic solvents (DES a type of ionic liquid) composed of choline chloride, lactic acid, citric acid and water. This leaching system has the property that it will dissolve metal oxides and metals of interest, including cobalt, nickel and lanthanides. This approach has been used for treatment of hard disk drives magnets at lab scale (Riaño *et al.*, 2017).

### *11.2.6 Ionic liquids (ILs)*

A promising non-biological method is the use of ionic solvents (ionic liquids -ILs) to process metals in WEEE. By avoiding water-based systems resulting in the absence of hydroxide ions ($OH^-$), less hazardous reagents can be used to solubilise metal salts and that side electrode reactions involving water and its ionic constituents ($H^+$ and $OH^-$) can be avoided in electrochemical dissolution and/or electrowinning. This can improve on the current efficiency, reducing costs and environmental impacts in recovery. The IL solvent can be tailored to suit the specific electrochemical properties of metals in particular wastes, through careful choice of cationic and anionic components of the IL.

Solvent choice affects the speciation of any dissolved metals, such as gold, neodymium, tin, silver, tellurium and antimony, and provides control over properties such as solubility and redox potential. ILs have been studied in depth for liquid-liquid extraction of metals, but this requires prior digestion using traditional strong mineral acids, although it does replace the use of hazardous organic solvents and complexing agents for metal ion extraction.

Tailoring functional groups on the anion or cation components of ILs allows 'task specific' ILs to be produces, which are selective for a particular metal in the liquid. The synthetic complexity of such ILs means that they are costly and must be used in small volumes, but their selectivity enables considerable process intensification.

ILs have been used for desoldering of PCBs, and extraction of TMs from lightning, NiMH batteries, rare earth magnets and to separate TMs in weak acid solutions. Applications of ILs in the recycling of WEEE have been reviewed by others (Schaeffer *et al.*, 2018b). Extraction of TMs using ionic liquids has been demonstrated extensively, including treatment of NIB HDD magnets (Riaño *et al.*, 2017), solder dissolution (Zeng *et al.*, 2013), dissolution of precious metals (Abbott *et al.*, 2015), (Gawen R T Jenkin *et al.*, 2016; Gawen R T Jenkin *et al.*, 2016; Gawen R.T. Jenkin *et al.*, 2016), tellurium and antimony (Gawen R.T. Jenkin *et al.*, 2016; Jenkin *et al.*, 2015).

### *11.2.7 Deep eutectic solvents (DESs)*

An alternative approach to ILs is the use of deep eutectic solvents (DESs), a mixture of quaternary ammonium salts (usually chlorides) and hydrogen bond donors, such as amides or polyols. The comparatively high chloride concentration of these DESs (~5 Molar) means that oxide chemistry of metals can be lessened or even avoided completely. It has been shown that many metal oxides show a greater solubility in these media, compared to ionic liquids with weaker anions.

The use of DES technologies can be disruptive to current process thinking and in principle can deliver benefits, including increased efficiency, lower costs and better process control and more efficient recovery of WEEE metals (Ryder *et al.*, 2020).

### *11.2.8 Electrochemical dissolution*

Electrochemical dissolution has been demonstrated for several metallic compounds, including superalloys which represent complex mixtures of TMs. To achieve this, materials to be recycled are placed in a DES solution and the application of a potential results in electrochemical dissolution of metals into solution. This may be unsuitable for WEEE treatment when waste comes in small pieces or powdered form (e.g., PCB fractions post comminution to enable efficient leaching) and hence cannot be made into suitable anodes. Chemical oxidation is therefore the only viable way to solubilise materials (e.g., traditional oxidative acid digestion) but consumes chemical reagents and generates high volumes of waste. The use of an electrocatalyst, i.e., a chemical oxidising agent that can be regenerated in-situ by an electrochemical reaction, could avoid ongoing reagent consumption and provide a sustainable dissolution process for complex heterogenous polymetallic wastes as WEEE (see previous comments on electrochemical dissolution in table 16). Indeed, this approach is common industrially, with toxic chlorine/chloride redox couple, and replacement with less toxic $Fe^{2+}/Fe^{3+}$ redox has been demonstrated to solubilise base metals in waste PCBs to reduce consumption of more expensive toxic oxidising acids necessary to solubilise more noble metals, such as Au.

The use of DESs in electro-dissolution of technology metals of semi-conductor compounds using the iodine/iodide redox couple, a strong oxidising agent for solubilising metals, in the DES Ethaline (1:2 molar ratio of choline chloride and ethylene glycol) has been demonstrated successfully (Abbott *et al.*, 2015) at lab scale. Ethaline is both air and moisture stable and can be used under atmospheric conditions. It has an electrochemical window of approximately 2 V on a Pt working electrode, sufficiently large that all redox processes related to the systems studied should be observed.

Electrocatalysis has great potential as a green method for solubilising and recovering metals as it has very high atom efficiency, by using, in effect, only electrons (which can potentially be generated from solar energy using PV or photoelectrode technology). Electrorefining uses the same principle but directly dissolves the metal from the anode. The advantage of electrocatalysis is that it no direct contact is required between the anode and the material to be oxidised. The method can hence be applied more widely to powdered and non-conducting materials, such as ores and e-waste components.

Examples of a range of IL and DES applications and their merits in recycling are given below (Table 19: Ionic liquid TM recovery methods).

*Table 19: Ionic liquid TM extraction methods.*

| TMs recovery | Application | Technology Readiness | Reference | Description |
|---|---|---|---|---|
| PGMs, PMs, Ts, Nd. | Reviews previous literature and trials adopting ionic liquids in extraction, synthesis and processing of TMs. | Review of methods. TRL 4. | (Park *et al.*, 2014) | Identifies potential of ILs to replace strong acids for extraction of TMs. Also, could be used for electrolysis. Potential to extract 99% of PGMs from HCl solutions. |
| **Batteries** | | | | |
| Co, Cu, Ln. | Hydrometallurgical separations of Li-ion and NiMH battery waste based on use of a deep eutectic solvent (DES). | Lab trials. TRL 4. | (Colabats, 2016) | Research project. The CoLaBATS technology has been designed to provide a multi- stage process capable of extracting key high value individual metals (Ni, Co, Cu, Ln, Zn), plus the extraction of other metal mixtures suitable for further refining. The process chosen involves a DES composed of choline chloride, lactic acid, citric acid and water. This material has the property that it will dissolve metal oxides and metals of interest, including cobalt, nickel and lanthanides.<br>The key performance benefits of the process are that the deep eutectic solvents could be recycled and reused multiple times. |
| Co, Cu, Ln, Ni | Battery recycling using ILs. | Review of potential use of IL solutions only all lab based. TRL 4. | (Schaeffer *et al.*, 2018a) | Review of previous research demonstrating processes clearly showing the potential of ILs to recover metals from complex aqueous solutions using careful selection of the ILs and understanding their influence on the metal extracted complex (chloride vs. nitrate, etc).<br>Demonstrated IL potential to replace strong acids for TM recovery from WEEE. |
| **Lighting** | | | | |
| Eu, Gd, Y, Tb, La | Waste fluorescent phosphor. | Review of potential use of IL solutions only all lab based. TRL 4. | (Schaeffer *et al.*, 2018a) | Review of previous research demonstrating processes clearly showing the potential of ILs to recover metals from lighting.<br>Demonstrated IL potential to replace strong acids for TM recovery from WEEE. |

| TMs recovery | Application | Technology Readiness | Reference | Description |
|---|---|---|---|---|
| Eu, Y | Lighting, using ionic liquids.<br>Reviews different acid leaching processes. | Wide range of different methods the majority could be used immediately. TRL 4. | (Sethurajan *et al.*, 2019) | Showed the ability to selectively dissolve REE oxides ($Y_2O^3$ and $Eu_2O3$) |
| **PCBs** | | | | |
| Pre- treatment of TM-bearing components on PCBs | Dismantling of PCBs using water-soluble ionic liquid. | Lab trials. TRL 4. | (Zeng *et al.*, 2013) | The WSIL such as [BMIm]BF4 was employed as the heating medium due to the wide temperature range of its liquid state, its suitable viscosity and the low expense. |
| **Magnets** | | | | |
| Co, Dy, Gd, Nd, Pr | NdFeB magnets. | Lab trials. TRL 4. | (Dupont and Binnemans, 2015) | In this paper a new recycling process for (microwave) roasted NdFeB magnets is proposed, based on the carboxyl-functionalized ionic liquid: betainium bis(trifluoromethylsulfonyl)imide, [Hbet][Tf2N]. Using the thermomorphic properties of the [Hbet] [Tf 2N]−H2O system, a combined leaching/extraction step was designed.<br>IL is regenerated during the stripping step and contamination of the water phase was avoided by salting-out the ionic liquid with Na2SO4. This innovative recycling process features a combined leaching/extraction in mild conditions using a reusable acidic ionic liquid and an energy-efficient microwave roasting of the magnets. The oxalate precipitation stripping method is very efficient, meaning that stoichiometric amounts of oxalic acid are sufficient to obtain 100% stripping. The stripping is fast (10 min) even at room temperature |
| Co, Dy, Gd, Nd, Pr | NdFeB magnets. | Lab trials. TRL 4. | (Riaño *et al.*, 2017) | A deep-eutectic solvent based on choline chloride and lactic acid (molar ratio 1:2) was used for the leaching of rare earths and other metals from NdFeB magnets. A process for the separation of Fe, B and Co from Nd and Dy in the deep-eutectic solvent was developed by using the ionic liquid tricaprylmethylammonium thiocyanate (Aliquat 336 SCN, [A336] [SCN]) diluted in toluene (0.9 M). |
| **Other** | | | | |

| **TMs recovery** | **Application** | **Technology Readiness** | **Reference** | **Description** |
|---|---|---|---|---|
| Light REE | Recovery of light REEs from mildly acidic, high nitrate salt, aqueous solutions. | Complex solutions need to be prepared. TRL4. | (Hunter *et al.*, 2018) | Solvent extraction of REMs by [(n-octyl)3NMe] [NO3] (IL). These experiments show that REE recovery using IL is a rapid process and that IL is highly durable.<br>Maximum recoveries of La (100%), Nd (98%), and Dy (92%) were obtained at 1.00 M IL with an expected gradual decrease in percentage recovery observed as the concentration of IL (M) decreased [14,15]. |
| In, Y and Ta | Recovery of TMs from PCBs and flat screens | Lab and small-scale industrial trial. TRL 3 to 5. | Potential EC tender bid by RINA.<br>https://www.rina.org/en | Sustainable recycling of End-of-Life products for CRM production by combining Microwaves and Deep Eutectic Solvents.<br>Use of microwaves to increase speed of digestion and higher extraction rate. |

### *11.2.9 Bioleaching*

Bioleaching is a way of recovering TMs from WEEE using bacteria and fungi, mainly from three different groups: chemolithotrophic prokaryotes, heterotrophic bacteria and fungi. Research on the biotechnical recovery of technology metals has been mostly based upon recovery from solution. Biotechnology is an established route for extraction of Au, Ag, As, Co, Cu, Mn, Mo, Ni, U, V, and W, Zn from primary ores (Morin *et al.*, 2006). They will not work in strong acid solutions so are not suitable for most of the initial extraction, due to the low pH, but are suitable for dilute waste streams such as effluent and have been used in extraction of technology metals from WEEE dust created during automated pre-processing. This represents a potential route to recapture some of the technology metals dissipated during comminution, although biological methods tend to be slower than hydrometallurgy or ILs unless they are agitated or stirred. Reports show only 20% of REEs were recovered after 24 hours (Morin et al., 2006).

In the UK, N2S are working in partnership with Coventry University to develop a novel biorefining system for metals from WEEE which is hoped will avoid the impacts of large logistics links to refineries, avoid losses of target materials which currently occur in recycling chains, and mitigate potential environmental impacts of recovery processes (N2S Ltd, 2020). Rather than using traditional lixiviants for precious metals, they are developing systems in which micro-organisms will extract precious metals from waste PCBs, enabling subsequent recovery from solution.

One issue is that Cu is a dominant part of the material composition of PCBs and other types of WEEE. It competes with the other metals i.e., Au and Ag, for bio recovery of precious metals, due to preferential complexation and leaching, meaning that it will leach Cu first before the other metals (Morin *et al.*, 2006). Therefore, it is often required that base metals are leached out of materials prior to more selective extraction with microbes.

The table below (Table 20) gives further information on the different technologies used for the recovery of TMs from WEEE using biotechnical methods. Bioleaching avoids the use of strong mineral acids by utilising the natural activity of microorganisms to leach metals. WEEE materials are exposed to microorganisms in solution that produce organics acids which digest and extract metals into solution for subsequent recovery. As microorganisms generate organic acids in situ, the requirement to purchase strong inorganic acids is avoided, saving environmental impacts and disposal costs. This lends itself well to recovery of traces of technology metals from WEEE-derived materials. Bioleaching also tends to be slower than digestion with traditional acid leaching systems or ILs, although again, this can be countered to some extent with appropriate agitation or stirring of the system.

Potential exists to develop strains to target specific metals and therefore perform selective leaching thus minimizing further treatment technologies and further reducing pollution, and recovery of metals from PCBs has been proven. The removal of the metallic components via bioleaching will then leave the non-metallic fraction for processing with relatively low contamination (vs traditional hydrometallurgy). Room temperature extraction is possible and as the process replicates natural biogeochemical processes, with no additional chemicals, it will have a lower environmental impact, compared to other extraction methods. Due to the slow leaching of TMs, it may only be suitable for extraction of these materials from extraction dust etc.

Examples of a range of bioleaching methods, as well as their merits and drawbacks for recovering TMs, are given below (Table 20).

*Table 20: Biotechnical technology metal recovery methods.*

| TMs recovery | Application | Technology Readiness | Reference | Description | Pros | Cons |
|---|---|---|---|---|---|---|
| **Applications for WEEE recycling** | | | | | | |
| Au, Co | Extraction from PCBs | Lab trials but suitable for use using readily available equipment. TRL4. | (Hursthouse *et al.*, 2018) | Bioleaching using 5 different mixed cultures of cyanogenic bacteria were evaluated. | Significant recovery of Au (up to 70%), Co (up to 96%) and Cu (up to 99%). | Limited information on extraction time. |
| Dy, Nd, Pr | NdFeB magnet leaching. | Lab trials. TRL3. | (Brewer *et al.*, 2019) | Biosorption-based flow-through process for selective TM recovery from electronic wastes. | High purity of extracted Nd. | Optimal REE.<br>extraction performance is achieved at pH 6 and limited information on extraction time. |
| Co, Dy, Eu, La, Nd, PMs, Se, Yb | Extraction from PCBs | Lab trials using different microbial biotechnology. TRL3. | (Yarlagadda, Mohan and Lens, 2016). | Extraction from different solutions, battery waste and WEEE. | Limited information on efficiencies and % recovered. | Limited information on efficiencies and % recovered |
| Ag, Au, Co | Extraction from PCBs | Lab trials using different microbial biotechnology. TRL3. | (WRAP, 2015, 2018). | High leaching rate but slow process. | Bioleaching performed with three types of microbes (acidophiles, fungus and cyanogenic) to determine leaching selectivity from ground PCBs. Acidophilic treatment with A. ferrooxidans resulted in significant recovery of Au (up to 70%), Co (up to 96%) and Cu (up to 99%) with minor additional recovery in the subsequent cyanogenic stage. | |
| **Other applications for recovery of technology metals** | | | | | | |
| REE listing not broken down | Research on extracting REE from monazite using biological methods. | Lab trials. Potential to trial on WEEE. TRL4. | (Hassanien, Desouky and Hussien, 2014). | Potential to be used on WEEE. | The highest REEs dissolution was 60.6 and 52.6 % from monazite. Low cost compared to acid leaching. | 8- 9 days of leaching required to reach this level of extraction. |
| Co, Ga, Ge, Li, PGMs, Sb | Extraction from WEEE dust. | A wide variety of lab trials reviewed. TRL4. | (Işıldar *et al.*, 2019). | Autotrophic, heterotrophic and fugal leaching of REE and PMs from WEEE dust. | Good extraction yields from low-grade source. | Very slow processes; 8 days to leach 50-60% of the TMs. |
| Ce, La, Dy, Eu, Ne, Pr, Te, Y | Recovery of metals from spent catalyst. | Lab trials. TRL3. | (Jin *et al.*, 2019) | Bioleaching using biological methods could be used for La and Ce bearing WEEE. | Using waste starch for bioleaching. | 20-30% REE recovery rate. |

| TMs recovery | Application | Technology Readiness | Reference | Description | Pros | Cons |
|---|---|---|---|---|---|---|
| PMs | Recovery of PM acid solutions. | Lab trials. TRL3. | (Donia, Atia and Elwakeel, 2007) | Recovery of gold and silver from acidic solution using chitosan resin. | The resin has good durability as well as good efficiency for repeated use. Uptake values of 3.6 and 2.1 mmol/g were reported for Au (III) and Ag(I), respectively. | |
| Au, Pd, Pt | Recovery from solutions. | Lab trials. TRL3. | (Fujiwara *et al.*, 2007). | Recovery of Platinum (IV), Palladium (II) and Gold (III) from Aqueous Solutions onto L-Lysine Modified Crosslinked Chitosan Resin. | The maximum adsorption capacity was found at pH 1.0 for Pt (IV), at pH 2.0 for Au (III) and Pd(II) The maximum adsorption capacity was found to be 129.26 mg/g for Pt (IV), 109.47 mg/g for Pd(II) and 70.34 mg/g for Au(III). | |

## 11.3 Section summary

Numerous approaches to achieving more cost-effective disassembly of WEEE, comminution and extraction of TMs have been discussed, which would enhance recovery potential of technology metals from WEEE by avoiding dissipative losses incurred in current WEEE recycling processes.

Electrohydraulic fragmentation (EHF) holds high potential to replace existing mechanical comminution approaches, and this will reduce TM losses.

Supercritical water, steam gasification and vacuum pyrolysis processes all hold great potential for separation of metals and plastics in components which would negate the requirement for comminution via shredding in order to access technology metals for efficient extraction and recovery. Each of these processes will require specialised plant.

Organic acids and ionic liquids are promising future options for extraction of metals into solution for hydrometallurgical recovery, which will not only avoid losses of the REEs incurred in current pyrometallurgical recycling processes, but also offer a safer, lower environmental impact and potentially lower cost option than strong inorganic acids that are traditionally used in hydrometallurgical treatment. Sequential selective leaching of metals will also enable optimisation of reagent costs, and simplification of recovery from solutions as compared to the complex mixed metal solutions produced when WEEE is treated with strong oxidising acids. Organic acids and ILs should be suitable to be used as direct replacements for current acids in existing acid leaching plant.

# 12 Recovery Technologies

Once extracted into solution, target technology metals must be recovered again. The processes for achieving this are well-established technologies, and the main ones are listed below.

## 12.1 Cementation

Cementation is a type of precipitation, a heterogeneous process in which ions are reduced to zero valence at a solid metallic interface when a sacrificial metal is added to the solution as a reducing agent. A general equation for the process is given via the following equation where TM is a technology metal ion in solution and SM is a sacrificial metal used to cement the target metal which, when reduced back to its metallic species, precipitates out of solution and sinks to the bottom of the reaction vessel.

$$\mathrm{TM}^{n+}{}_{(aq)} + \mathrm{SM}_{(s)} \rightarrow \mathrm{TM}_{(s)} + \mathrm{SM}^{n+}{}_{(aq)}$$

Zn is often used for cementation of PMs. This process is well developed and widely used for metals recovery. La, Ce, Pr, Nd, Sm, Eu, and Yb have all be evaluated for cementation using different methods[23].

## 12.2 Ion exchange

Ion exchange usually describes a process of purification of aqueous solutions using solid polymeric ion exchange resin. More precisely, the term encompasses a large variety of processes where ions are exchanged between two electrolytes. Pregnant leachate solutions containing target metals are passed through ion exchange columns, which adsorb target metals. These can then be flushed out of the system and collected. It is possible to pass complex mixed ion solutions through several columns containing ion exchange resins designed to target specific metals for recovery. The methods outlined in Table 21 give examples of where ion exchange has been used to recover metals from leachate

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[23] La, Ce, Pr, Nd, Sm, Eu, and Yb have all be evaluated.

solutions of WEEE materials. Ion exchange columns can be designed to size to suit plant throughput. Although CAPEX may be high for larger columns, the ability to efficiently recover solubilised metals selectively from complex mixed metal solutions lends itself to technology metal recovery from complex metal leachate solutions.

*Table 21: Ion exchange TM recovery methods.*

| Target technology metals | Application | Technology Readiness | Reference |
|---|---|---|---|
| Co, PGMs | Metal scavenging from acid solutions containing TMs. | Silica based scavengers developed by Phosphonic. TRL 9. | (*Metal Recovery & Recycling \| phosphonics*, no date). |
| Dy, Er, Ho, Tm Yb, | Recovery of REE in solution via ion exchange using Ethylenediaminetetraacetic acid (EDTA). | Equipment and EDTA commercially available. TRL 9. | (Anderson, 2016). |
| Ho La, Sm, and Th | Recovery from sulphuric acid used to dissolve WEEE metals. The adsorption properties of chelating ion exchange resins were investigated. | Potential to use in current ion exchange columns. TRL4. | (Page, Soldenhoff and Ogden, 2017). |

## 12.3 Electrowinning

Electrowinning is the recovery of metals from solution through reduction at an electrode in an electrochemical cell by passing a current from an inert anode through a leachate solution containing the target metals so that the metal is reduced and collected as it plates onto the cathode. This is a well-developed technology in current use and systems are commercially available. It is commonly used in hydrometallurgical PCB recycling to recover copper from solution, leaving traces of other technology metals including PMs and PGMs present in anode slime for further refinement. By tuning the potential applied to the cells, it is possible to selectively recover target materials by virtue of their different redox potentials. Although the process requires specialist plant, this requires relatively low CAPEX compared to pyro-based plants. Ongoing energy consumption in the process adds to OPEX.

## 12.4 Solvent extraction

Solvent extraction is a liquid-liquid extraction process in which a non-miscible solvent is added to a pregnant leachate solution containing dissolved target metal ions. The solvent selected will have high affinity for a particular target metal in the leachate and so will selectively extract it. When the two non-miscible liquids are allowed to settle, the target material can be simply recovered from the solvent, overcoming complexities and inefficiencies encountered when attempting to extract single metals from complex mixed metal solutions. Solvent extraction is commonly used in labs and industry, for example removing phenol from the effluent of catalytic cracking in petroleum refinery, and processes for selective extraction of metals have been established in the minerals processing industry. A prominent example of a selective solvent used in technology metal recovery is butyl diglyme, which is used to extract gold selectively from aqueous mixed metal solutions. Once separated again, gold can be recovered by precipitation from solution.

## 12.5 Biosorption

As previously discussed in the section on bioleaching, a range of microorganisms are able to bind and adsorb metals from solution. For examples of applications in WEEE treatment see Table 20. One advantage to the use of microorganisms in this application in addition to those previously discussed is that they can effectively recover metals at very low concentration. This lends itself to recovery of low concentrations of technology metals present in WEEE. One prominent commercialised example is the proprietary PCB recycling process invented by Mint Innovation Ltd, which will be discussed in Chapter

13. Biosorption has also been shown to generate precious metal nanoparticles, which may be of higher value than metals recovered as bullion or cathodes in electrolytic systems, which sell at the going market value for the metals. There is a high demand for end-use applications of nano gold in healthcare, electronics, cosmetics and photometers[24]. This may be an advantage as added value products could potentially be generated from technology metals in solution, which would improve overall cost benefit of recovery.

## 12.6 Systems for process intensification

It has been discussed that minimising recovery costs is of paramount importance to increase recovery of technology metals from WEEE. The following technologies are those which enable more efficient use of reagents in hydrometallurgical recovery and therefore reduced OPEX.

### *12.6.1 Diffusion dialysis*

To make the hydrometallurgical recovery of technology metals from WEEE more cost effective, the reuse of the acids and alkalis used in extraction needs to be pursued. Diffusion dialysis is a membrane separation process. It has been successfully used for many years for the separation and recovery of acids from dissolved metal-bearing solutions (Boddu and Rajagopalan, 2011). This method exploits the acid permeability of anion-exchange membranes. These membranes are very permeable to anions while effectively preventing the transport of cations, due to repulsion by the fixed positive charges on the polymer matrix. The cation repulsion effectively prevents salts from diffusing through these membranes. This can lead to normal acid recovery of 80-90% with removal of 70-90% of the dissolved metals. Once treated, the acid can then be reused, and the concentrated dissolved metals extracted from solution using cementation, ion exchange or electrowinning.

The diffusion dialysis process has been used in acid pickling of stainless steel and recovery of solder etch since the 1990s. (*Acid/Alkali Recovery & Metals: Membranes | Ionomr Innovations Inc*), (*Base and Acid Recycling with Electrodialysis | Saltworks Technologies*), (*Acid Purification - Diffusion Dialysis Membrane Technology - AcidRecovery.com*).

The advantages of this method, based upon these reports, are:

- Eliminated disposal costs and reduced inventory purchases of acid;
- Eliminated production downtime associated with dumping and recharging acid baths;
- Minimized direct operator contact with dangerous chemicals; and
- This method can be scaled up as required and it is estimated that the return on investment would be within 4 years.

### *12.6.2 Counter-current leaching*

Counter-current leaching is a technique that uses an acid to leach TMs and then the acid is processed and restored after leaching. For example, sulfuric acid is used to leach indium from LCD glass. The pregnant leachate is then captured, restored with a small amount of sulfuric acid, and used again to treat a new batch of LCD glass (Rocchetti *et al).* Until now, economic recovery of indium has been problematic due to its low concentration and therefore value compared to costs of processing. In this way, process intensification is achieved, resulting in a recovery rate of over 98% with a purity of >95%, reducing chemical costs and improving the overall cost-benefit of the system. Similar strategies may be suitable for economic recovery of traces of technology metals, such as cobalt, lithium and REEs from other WEEE derived materials.

## 12.7 Section summary

Solvent extraction, ion exchange, and electrolytic recovery hold the highest potential for selective

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[24] https://www.inkwoodresearch.com/reports/global-gold-nanoparticles-market-forecast/

recovery of metals from mixed metal solutions, and therefore lend themselves to hydrometallurgical recovery of technology metals from WEEE.

In the interests of maximising recovery economics and thereby technology metal recovery potential, process intensification options are recommended, to reduce reagent costs and volumes of waste for treatment, and these include: electrocatalysis, counter-current leaching, and diffusion dialysis.

Diffusion dialysis is a cost-effective way of recovering acids and alkali for reuse and should reduce the OPEX of hydrometallurgical processes.

Bioleaching and biosorption hold great potential for the future as low cost lower environmental impact alternatives for extraction and recovery, but due to their slow extraction of TMs, may only be suitable for extraction from shredding dust etc.

# 13 Systems for technology metal recovery from PCBs

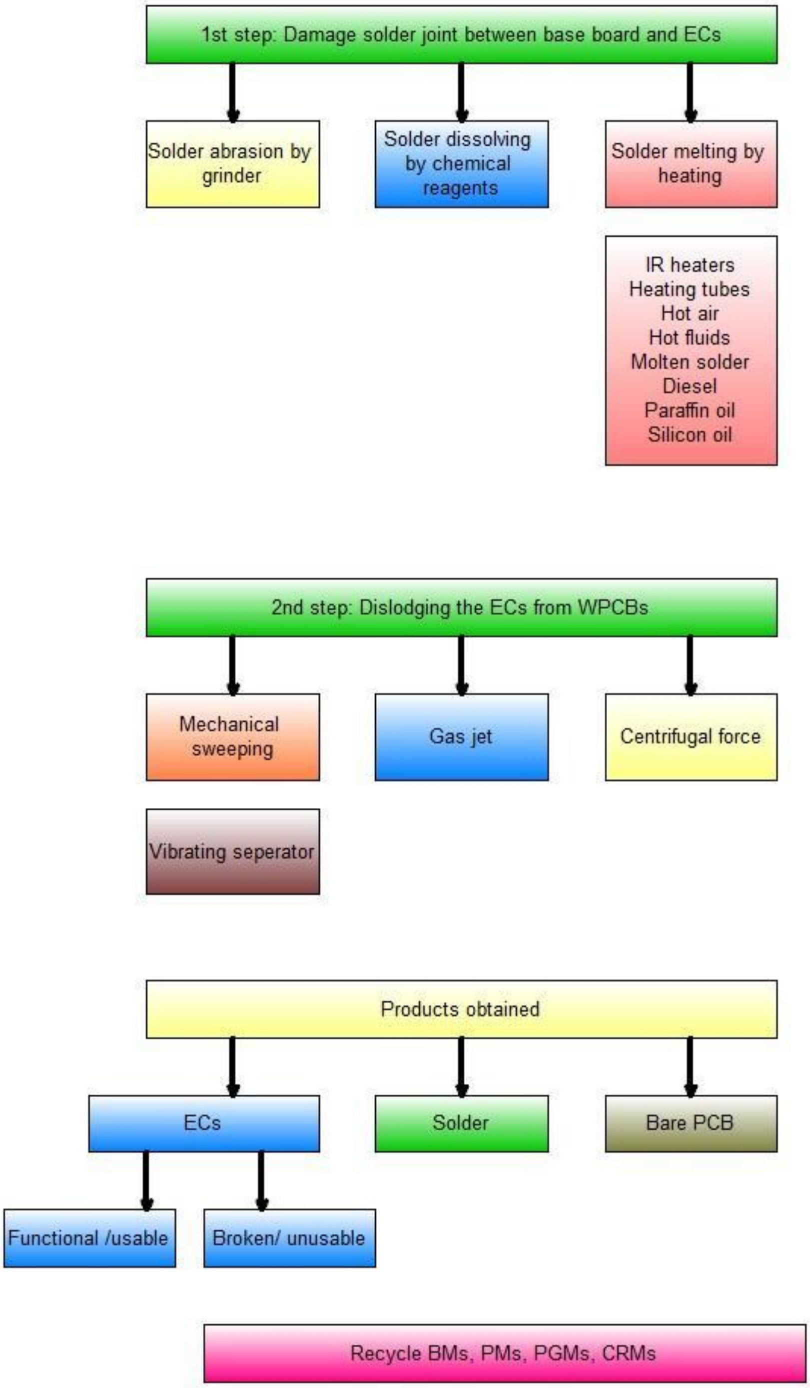
It has been shown that there is the potential for hydrometallurgical recovery approaches to increase recovery of technology metals and avoid the losses which are inherent in smelting processes. These processes can be adopted at smaller scale and have more inherent flexibility, resulting in far lower CAPEX, flexibility, simpler process expansion, and the potential to employ process intensification strategies and even selectively recover a wide range of technology metals using emerging leaching systems. In addition, they are more suitable for small-scale decentralised recovery, reducing logistics costs associated with transporting materials for recycling. The following discussion outlines proprietary commercially available systems for printed circuit board (PCB) recycling which are available now, as well as approaches to upgrading recycling technology to recover metals from PCBs through disassembly of their components.  If these are adopted, they could enable the UK to exploit the potential of TMs as a domestic source of TMs and help alleviate some of the risks identified throughout this report.

## 13.1 Dismantling/Disassembly of TM-bearing WEEE parts

Disassembly processes are employed to segregate components and/or materials that are reusable, identifiable or hazardous in such a manner as to maximise economic return and to minimise environmental pollution, enabling subsequent processes to be performed more efficiently and effectively (Kaya, 2020a) (Figures 30 and 31). This enables the maximum possible inherent value of WEEE to be exploited, providing the best possible chance for viable TM recovery.

Isolating Surface Mounted Devices (SMDs) is carried out manually, which is time consuming and places limits on processes in terms of minimum costs involved (Kellner, 2009a). Disassembly is the major cost element and is a time-consuming operation within the recycling process (Kaya, 2019). Low processing capacities hinder the application of manual disassembly in large scale industrial processes, necessitating development of automated disassembly to limit costs and increase capacity to process the globally available quantities of PCBs (Goosey and Kellner, 2002).

*Figure 30: PCB dismantling (Kaya 2020a).*

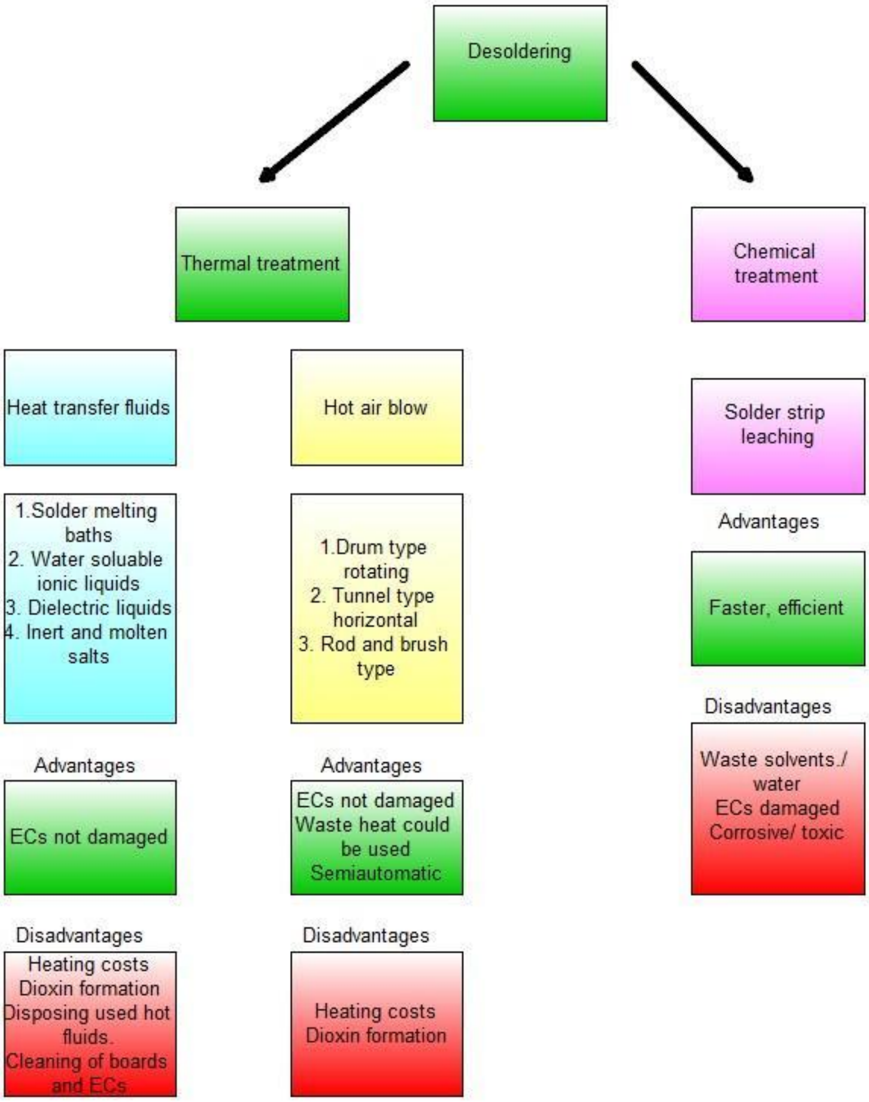


To depopulate PCBs of their components, a 2-step process is utilised in which: i) Solder joints attaching components to boards are broken in some way, and ii) detached components are dislodged from the board. Desoldering has been conducted in a number of ways (Figure 30), including abrasion of solder joints on the backside of boards, chemical dissolution of the solder, or thermal methods to melt solder. Usually, chemical and heating technologies are used for dismantling components from PCBs. Damage from chemical agents to components has prevented commercialisation to date.

Thermal treatment e.g., infrared (IR) heating, electric heating tubes, liquid-medium heating and solder-bath heating are the most commonly used methods used to recover components. All these methods generally operate at about 225–265°C, which is 40–50°C higher than solder melting point.

The critical temperature to generate toxic fumes from PCBs is 270–280°C (Kumari, Jha and Singh, 2016) so dioxin formation, a highly toxic compound, may be an issue during heating. Appendix A has a detailed list of PCB disassembly processes reported in literature, outcomes of studies, and comments of potential for application at industrial scale.

*Figure 31: Thermal and chemical desoldering treatment of PCBs for disassembly (Kaya, 2020a).*



## 13.2 Hydrometallurgical removal of solder from PCBs

PCBs generally have a chemical coating (solder mask), which is commonly made of epoxy. The solder mask covers the metals mounted on the PCBs and does not allow leaching agents to penetrate through for efficient extraction of metals. For this reason, comminution to reduce particle sizes is normally adopted, which incurs technology metals losses. However, alternative solutions to this problem have begun to emerge, e.g a 10 Molar (M) sodium hydroxide (NaOH) solution has been shown to remove this chemical coating, thereby granting access to the contained metals for leaching and recovery without losses of target materials (U Jadhav and Hocheng, 2015).

It has also been seen that non-target technology metals (such as REEs, and tantalum) within PCB components, charged to recovery processes that do not target them, are permanently lost. In addition, mass-based refining charges are incurred even for the plastic and epoxy portions of PCBs, although these cannot be valorised, thereby increasing recovery costs for technology metals. One solution is to desolder and disassemble components from PCBs, thereby concentrating the value and technology metals contained without incurring losses to improve overall recovery potential. This also provides the opportunity to segregate these components according to technology metal content, to direct them to bespoke processes for recovery of all technology metals contained as these emerge such, as those for recycling of tantalum capacitors (Niu and Chen, 2017), (Mineta and Okabe, no date), (Chen and Ho, 2019). The following are some examples of available technologies for desoldering and disassembling components from PCB's.

### *13.2.1 Acid and ionic liquid (IL) removal of solder*

In 2013 (Zeng *et al.*, 2013) reported on an "environmental-friendly" dismantling process through heating with water-soluble ionic liquid to separate electronic components and tin solder from two main types of PCBs—cathode ray tubes and computer mainframes. The work systematically investigates the influencing factors, heating mechanism, and optimal parameters for opening solder connections on PCBs during the dismantling process and addresses its environmental performance and economic assessment.

The method was to submerge PCBs in Ionic liquid 1-Butyl-3-methylimidazolium tetrafluoroborate [BMIm]BF4, heat with oil bath and stir in the absence of oxygen/air to avoid generation of hazardous compounds. [BMIm]BF4 was employed as the heating medium due to the wide temperature range of its liquid state, its suitable viscosity and the low expense. The results obtained demonstrate that the optimal temperature, retention time, and turbulence resulting from impeller rotation during the dismantling process, were 250ºC, 12 min, and 45 rpm, respectively. Nearly 90% of the electronic components were separated from the PCBs under the optimal experimental conditions. This novel process offers the possibility of large industrial-scale operations for separating electronic components and recovering tin solder, and for a more efficient and environmentally sound process for PCBs recycling.

Although the IL tends to be expensive (250g of [BMIm]BF4 costed £350 at the time of writing[25]) it could be recovered and reused. The use of ILs could be reduced by using a heated spraying system rather than immersion of the boards. Combining IL cycling with the spray technique represents a method of process intensification which would considerably reduce reagent costs, by at least 60% (Zeng et al., 2013). This is a typical example of what could be achieved using ILs. They have also been used for the extraction of technology metals from lighting, NiMH batteries, rare earth magnets and to separate CRMs from weak acid solutions. Zeng *et al.*, have conducted an economic assessment of their proposed industrial IL process (without the previously mentioned alterations), compared to manual dismantling with pneumatic screwdrivers and a semiautomated mechanical process (see Figure 32) (Zeng et al., 2013).

Results show that for treatment of <1kt of PCBs, manual approaches are superior to the other options compared, hence the popularity of manual approaches in many developing countries where labour costs are generally lower than more developed countries. Above this 1kt threshold though, high total labour costs for manual processes result in economic advantages for alternative approaches. The IL approach is also more cost competitive than the semi-automated mechanical dismantling of components from the boards for PCB quantities >3kt.

The graph below shows a comparison of the costs of manual, mechanical and heated ILs dismantling of waste PCB (WPCB).

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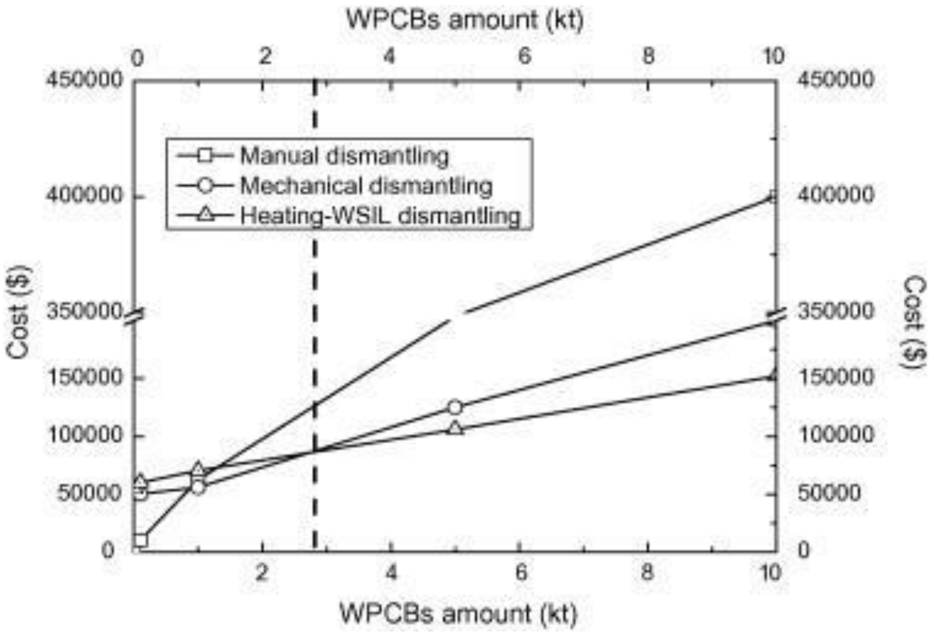
[25] https://www.sigmaaldrich.com/catalog/product/aldrich/91508?lang=en&region=GB

*Figure 32: Cost comparison of PCB dismantling technologies (Zeng et al., 2013).*



Selective PCB disassembly allows valuable components to be 'cherry picked' from PCBs. Simultaneous disassembly removes all SMDs en-masse, and these require further sorting into material flows for reuse and TM recovery. A number of processes are employed industrially, most commonly in the US and China, with much research underway to develop lower environmental impact and more cost-effective solutions. In general, processes rely on chemical leaching to dissolve solder or thermal heating to melt solder.

Commercialisation of chemical desoldering has been hampered by use of strong inorganic acids such as hydrochloric acid (HCl), which damages components, thus preventing reuse, and requires subsequent rinsing of treated materials, wastewater treatment and acid-proof plant, all of which add considerable cost. One proprietary chemical process, licensed by Itrimex, was used to disassemble PCBs by Axion during the 'CRM Closed Loop Recovery' trials (Itrimex, 2013; WRAP, 2015). Although effective at solder removal, prior removal of large metallic components was necessary to prevent vigorous effervescence and reagent consumption. In particular, through-hole device (THD) and mechanically fastened components were not as effectively removed as surface mounted components (SMDs) (WRAP, 2018).

To overcome issues associated with the use of strong chemicals, Zhang *et al.* (2017, 2015) developed fast, economical and lower environmental impact selective leaching systems which achieve ~100% solder dissolution at room temperature in 35 minutes using 2.5 Molar Fluoroboric acid ($HBF_4$) with 0.4 mol/L hydrogen peroxide ($H_2O_2$); and 45 mins using 3.5 M methanesulfonic acid (MSA) with 0.5 mol/L $H_2O_2$. Ionic liquids also hold great potential to avoid consumption of reagents and component damage.

Zhu *et al.* (2012a, 2012b) achieved complete solder dissolution by treating PCBs with 1-Ethyl-3-methylimidazolium tetrafluoroborate ($[EMIM^+]$ $[BF_4^-]$) at 240 °C for 10 minutes while stirring at 150 rpm. Interestingly, ramping temperature to 260 °C for 10 minutes following SMD removal achieved total dissolution of flame-retardant epoxy enabling Cu-foil and fibreglass recovery from PCBs, which are promising preliminary results towards valorisation of depopulated PCBs.

### *13.2.2 Thermal and electrohydraulic fragmentation desoldering*

Thermal desoldering is more commonplace, using IR heating, electric heating tubes, liquid heating media (including dielectric liquids and molten salts) and solder bath heating (Kaya, 2020). Careful control of temperature uniformity across the boards, and peak temperature, are important to avoid component damage and evolution of toxic gases. Traditional Sn/Pb solder melts at ~183 °C. Pb-free solders[26] generally melt at higher temperatures e.g., Sn-Ag-Cu melts >210 °C. The required desoldering temperature therefore vary depending on solder type, along with the specific heat capacity and spatial arrangement of components that might result in non-uniform heating (Xiang et al., 2014). Methods generally operate at 225–265°C i.e., 40–50°C above the solder melting point and close to the critical temperature for dioxin generation from PCBs (270–280°C in the presence of oxygen).

Thermal desoldering therefore requires inert atmospheres and/or off-gas treatment, which adds cost (Kumari et al., 2016). Semi-automated thermal desoldering systems have been developed. Wang *et al.* (2016) created a pilot rotating drum system heated with hot air blowers or electrical resistance heating and with an activated carbon off-gas purification system. By ramping temperature to 265 ± 5 °C in 8 minutes while rotating at 6 rpm all SMDs were removed intact, although issues were encountered when attempting to dislodge THDs and mechanically attached components which required manual removal. Off-the-shelf tunnel type IR based systems are available with dust capture and off-gas filtration which claim to remove >99% of PCB components (Yuxi Machine, 2019). Park *et al.* (2015) developed an IR heating system which heats the solder to melt temperature and then uses a brush and rod to remove SMDs, THDs and mechanically fastened components effectively.

Recycling-Börse used electrohydraulic fragmentation (EHF) to break solder bonds and liberate PCB components, avoiding chemical and thermal desoldering issues (WRAP, 2018). Lee *et al.* (2012) investigated a 3-step separation system of sieving, magnetic and density separation of mixed THDs to simultaneously disassemble components from PCBs via mechanical abrasion and to sort components by type. Xiang *et al.* (2014) developed a 'reuse' oriented thermal disassembly system along with a component testing system, an important step to overcome barriers to use of recovered components in manufacturing and increase valorisation of PCBs while retaining technology metals within the circular economy.

Some of the most important developments in desoldering have come about through image processing technology linked to databases of component information, enabling automated selective disassembly of PCB components, which has the advantage of automatically segregating components according to reusability and CRM / hazardous substance content without further separation processes. Hayashi *et* al. (2019) have developed an object recognition algorithm based on optical character recognition (OCR) analysis of captured product label images for high throughput processing of cameras, enabled by rapid determination of make and model. The Austrian Society for Systems Engineering and Automation (SAT) developed an image analysis system which identifies PCB components based on size, shape, colour and surface markings to direct selective disassembly with dual-beam laser desoldering and vacuum system component removal (Kaya, 2019; Kellner, 2009).

## 13.3 Proprietary Commercially Available Hydrometallurgical PCB Recovery Systems

Alternative hydrometallurgical approaches are also adopted industrially throughout the world. These rely on leaching of metals from PCBs into solution so that they can be separated and refined. These processes have distinct advantages over pyrometallurgical processes. Recovery is viable at far smaller scales, and expansion of capacity when necessary is far easier. Such plants lend themselves to decentralised, local recovery. The issue of TM losses in slags is also avoided. Hydrometallurgical PCB recovery has been more commonplace in Asia than Europe. Traditionally, these systems have used

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[26] Pb-free solder is becoming more prevalent in WEEE since the EU RoHS Directive ban of Pb-solder (EC, 2011).

aggressive cyanide, chloride or inorganic acid-based leaching systems to oxidise and solvate gold and other target metals for subsequent recovery.

### *13.3.1 EMAK Refining & Recycling*

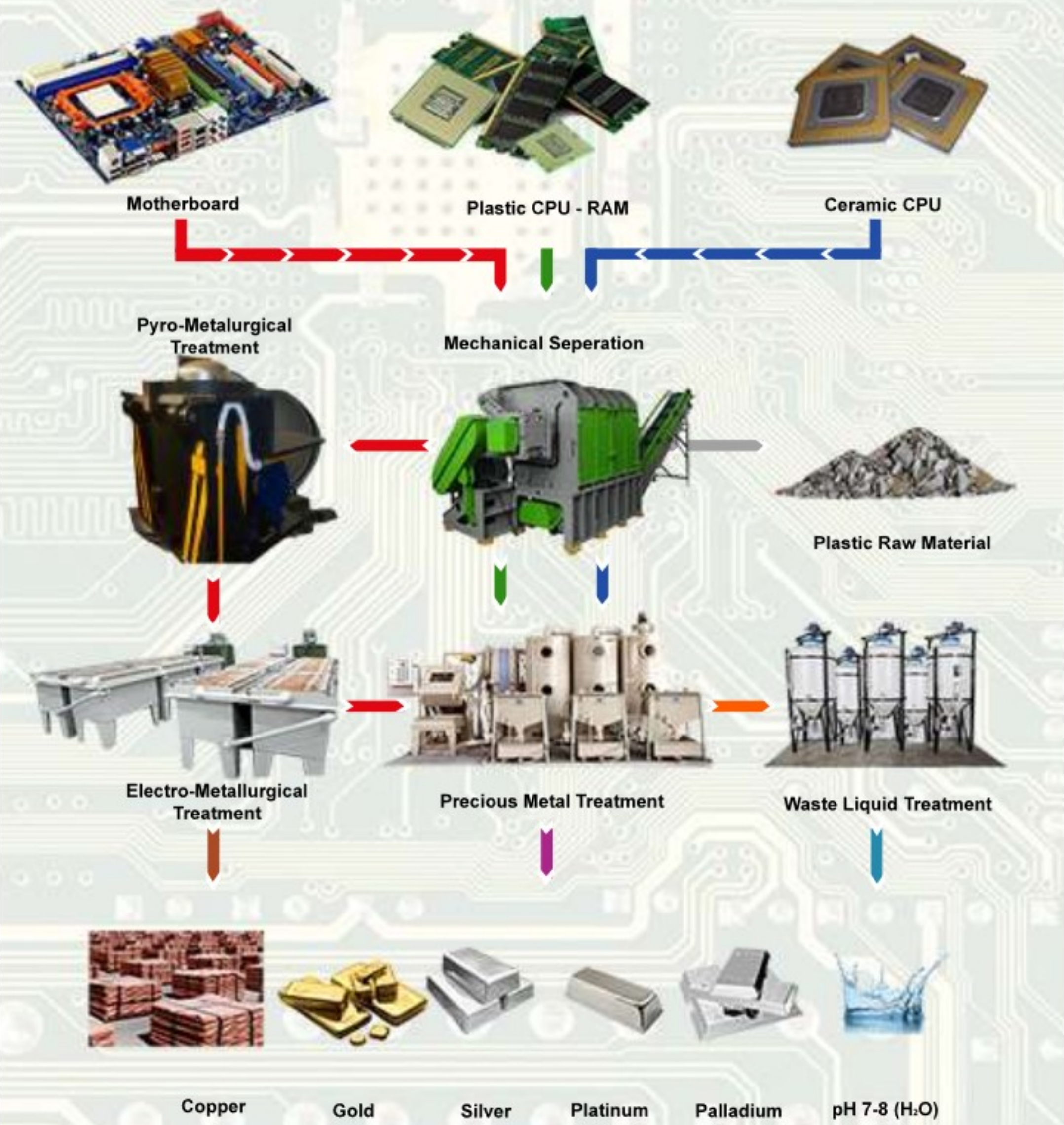
EMAK Refining & Recycling are a Turkish based company claiming to offer turn-key WEEE recycling solutions to recover Au, Ag, Pt, Pd, Cu, Sn, Pb, Zn from PCBs and other PM-rich WEEE components (EMAK Refining & Recycling, 2016). Although these plants only target PMs and PGMs, they can recover a greater range of metals than a typical large smelter. Systems are capable of refining Au, Ag and copper to 99.99% purity and Pd, Pt and Rh to 99.5% purity.

Plants include:

- Mechanical pre-treatment systems to crush and separate plastics;
- Ferrous and non-ferrous metallic fractions with dust capture to limit precious metal losses;
- A mini-smelting unit with fume treatment systems for non-ferrous metal enrichment via smelting (which can run on fuel oil, gas or electricity depending on which is most suitable) and enables low boiling point metals such as Pb, Sn and Zn to be collected, whilst the remaining molten copper alloy can be cast as bars or anodes for subsequent electrolytic refinement;
- Electrolysis systems which enable refinement and recovery of copper as high purity cathodes and collection of PM-rich anode slime for subsequent PM recovery;
- A precious metal recovery system consisting of a rotating drum leaching tanks with precipitation tanks for PM recovery from solution; and
- An effluent neutralisation unit which precipitates out residual metals from solution to prevent losses.

Figure 33 shows the processing stages in an EMAK plant.

*Figure 33: EMAK e-waste recycling systems (EMAK Refining & Recycling, 2016).*



In addition, the potential exists to augment such processes as technology evolves, to be capable of recovering more of the TMs present in PCBs. For example, sequential leaching in different leaching systems could enable more selective extraction of metals present for separate recovery, e.g., an initial leach with sulfuric acid to remove base metals to reduce the volumes of reagents required to leach PMs, PGMs and other more noble metals in a subsequent step, for example with thiourea.

The smelting process used for non-ferrous enrichment will suffer from the same issues as traditional smelting in terms of losses of TMs in slags, however pre-treatment using various technologies previously discussed can potentially segregate any TMs likely to be dissipated in the process prior to comminution for treatment in alternative processes. In addition, materials recovered at the anode will contain a range of TMs carried through the process in copper bullion which may also be recoverable in addition to PMs with further R&D. In addition, an EMAK plant could be repurposed to utilise alternative chemistries as research into alternative leaching and recovery chemistries reaches maturity.

Each of these units can be acquired at various capacities to suit requirements, starting at a minimum throughput of 1 tonne of PCBs per day. The indicative operational costs, revenue from sales of recovered metals and profits at different plant capacities are shown in Table 22.

*Table 22: Indicative costs, revenue and profit for EMAK gold and silver metals recovery systems for PCB treatment (EMAK Refining & Recycling, 2016).*

| **System capacity (Tonnes PCBs/day)** | **USD ($)** | | |
|---|---|---|---|
| | **Monthly costs**[27] | **Monthly revenue**[28] | **Annual profit** |
| 1 | 154,247 | 224,145 | 688,417 |
| 3 | *undisclosed* | *undisclosed* | 3,459,439 |
| 5 | *undisclosed* | *undisclosed* | 4,865,720 |
| 8 | *undisclosed* | *undisclosed* | 7,780,567 |

The running costs include $14,059 in chemicals, $9,706 in electricity and $6,156 in labour per month for the 1t per day plant.

The exact recovery efficiencies (yield) of technology metals from PCBs using this system will vary according to how it is optimised for economic performance, the nature of the boards being fed to the process (i.e., TM content and PCB configuration) and the degree of grinding in the pre-processing steps to enable efficient extraction. Depending on geographically specific factors such as overheads, optimum economic performance may be achieved with a below-maximum recovery efficiency. However, the grinding systems are fitted with various dust capture filters to ensure any technology metals that might be dissipated during shredding are captured, which should limit economic losses in this stage of the process. The dust capture may therefore encourage the use of grinding to produce smaller, higher surface area particles for enhancing treatment efficiencies and yields. Once the non-ferrous fraction from which technology metals are recovered is isolated, it is possible to concentrate 99% of gold in anode mud following electrolytic recovery of copper, acid digestion of the PMs and cementation.

EMAK staff will also support installation and training of staff to use the plant, as well as to optimise process parameters for best economic performance. The entire plant for PCB treatment with throughput of up to 1 tonne per day can be purchased and installed for ~ $2 to €2.5 million, which also includes a fire assay lab to allow essential analysis and assay of materials received. The plant also includes the necessary wastewater treatment, and any necessary extraction to achieve environmental compliance. This enables entry into the PCB recovery arena, for recovery of PMs and copper at considerably lower CAPEX than is required for a traditional pyrometallurgical refinery.

As previously discussed, many of the recycling operations in the UK are pre-processing which isolates PCBs from whole items. To deliver these PCBs to smelters for refining, sufficiently large quantities must be accumulated to i) be of interest to the refiners; and ii) enable economically viable export of PCBs to refineries. Often brokers perform the function of accumulating large quantities of PCBs from suppliers around the UK, to ship to these refineries. Refining terms for larger quantities of boards are also favourable compared to small quantities. Systems such as EMAK's provide the opportunity for smaller suppliers of PCBs and UK based brokers to realise greater value from metals in PCBs, which is important to ensure that items remain above cut-off-grade for manual (and semi-automated) recycling which avoids dissipative losses of TMs.

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[27] Costs include purchasing medium grade PCBs, chemicals, electricity, land and labour

[28] Revenue from sale of recovered precious metals (150 g Au, 250 g Ag), PGMs (30 g) and Cu (150 kg)

### *13.3.2 Enviroleach*

Enviroleach are a Vancouver-based science and technology company that has developed technologies that specifically cater to the mining and e-waste industries. Recently, Enviroleach has developed a patent-pending formula for a water-based leaching system using water and 5 additives which are all FDA approved for human consumption (Enviroleach Technologies Inc, 2020). Enviroleach claims this solution reduces the potential harm caused to the environment or human health by using alternatives to alternative strong inorganic acid and cyanide leaching which have been used to-date both in mining and WEEE recycling for PM recovery. Enviroleach has two new plants utilising their technology, one in Vancouver, and a joint venture with Jabil Inc. Memphis, Tennessee; a 650,000 square foot plant which, following comminution of PCBs, will use the new proprietary leaching technology to recover PMs and PGMs from the WEEE. Jabil will manufacture electronic components for Dell, HP and other clients from these recovered metals, essentially closing the loop.

The process uses pulverised WEEE mixed with ordinary water containing the five proprietary ingredients and the solution is pumped through cells of small, man-made electrically charged diamond plates. The gold and other precious metals separate and are extracted from the solution, which can be recharged and used again. The use of such leaching systems overcomes issues of treating large volumes of hazardous liquid wastes produced in alternative processes and avoids ongoing procurement costs for chemicals. The leaching process has also been used to produce concentrates for sale to Glencore for further refinement. When applied in this way, the process effectively increases concentrations of metals, making them more appealing to refiners, reduces transport costs, and may result in favourable terms at refineries and reductions in mass-based refining charges applied.

### *13.3.3 Mint Innovation*

Mint Innovation limited have developed a proprietary PCB recycling system that relies on bio recovery of precious metals from solutions (Mint Innovation, 2020). The process involves grinding the PCBs to powder in order to create an appropriate feed for the process, which involves two leaching stages. The first is a base metal leaching stage, the second leaching dissolves precious metals using a proprietary leaching system, which is proposed to be environmentally safer than traditional leaching systems such those based on cyanide or acidic chloride media. Once dissolved, solutions containing gold are then blended with a micro-organism which adsorbs the gold present in solution, gaining weight as it does. Once the process is complete, the microbes are centrifuged to separate the now target metal rich microbes from the solution which is now depleted of gold. The isolated microbes are then ashed and refined to yield recovered metallic gold. Mint Innovation holds a patent for this process that covers the application of bio-recovery to most traditional forms of hydrometallurgical treatment of PCBs i.e., with a number of pre-processing options including grinding, component removal following desoldering, and leaching conducted with traditional organic acid lixiviants (Crush, 2019). Mint is currently installing a plant for recovery of PMs from WEEE in Cheshire which is dedicated to WEEE treatment.

### *13.3.4 HydroWEEE*

The HydoWEEE mobile plant was developed under the EU FP7 Environment work program to recover TMs from fluorescent lamps, CRTs, LIBs, PCBs and LCDs (Amato et al., 2016). The project focused on the recovery of base and precious metals from WEEE using innovative hydrometallurgical processes. The initiative built on an earlier EU project that developed processes using sulfuric acid to extract valuable metals from lamps and cathode ray tubes (CRTs), liquid-crystal displays (LCDs), lithium-ion batteries, printed circuit boards (PCBs) and industrial catalysts.

The HydroWEEE plant is mobile (in a container), enabling several SMEs to benefit from the same plant at different times and which limits the necessary quantities of waste being produced to make such a plant viable, as well as minimising the necessary investment in such a plant for recovery. By making the processes universal several fractions (lamps, CRTs, LCDs, printed circuit boards and Li-batteries) can be treated in the same mobile plant in batches. Thus, by treating several waste streams sequentially, a single smaller capacity plant can replace several industrial processes. Further details on this plant in recovering TMs from batteries are in Appendix C.

### *13.3.5 Other examples of recovery plants*

There are other sites in the UK such as Johnson and Matthey in Royston, JBR in West Bromwich and Phoenix County Metals in Northampton that can recover PMs and PGMs from other types of waste such as silver inks, x-ray films and exchange resins.

### *13.3.6 Cost of recovery plants*

In general, setup and running costs for existing TM recovery facilities were not available. However, a capital expenditure of ~\$2 to \$2.5m (£1.4 to £1.8m) is thought to be indicative for a fully operational EMAK recovery and recycling plant (recovering PMs, PGMs, tin and copper) with a capacity to treat 1 tonne of PCBs a day (~250t per annum). Further investment will be required in the developing technologies identified by this research to recover the full range of TMs. At least 56% of the of £133.86m value of TMs in the WEEE resides in the PCBs and an estimated 18 such plants would be needed to treat the estimated 4,500 tonnes of precious metal-bearing PCBs extracted from the computers, laptops, tablets, TVs and smart phones discarded in the UK each year, with an estimated value in excess of £75m. This would require an approximate investment of £25m - £32m in TM recovery plant. Based upon approximate operational costs (as provided by EMAK) for chemicals, electricity and labour of £1,077 per tonne in a 1t a day EMAK plant, the cost to treat the 4,500t would be an estimated cost of £4.85m. This does not include the cost of purchasing the WEEE to feed the plants, permitting costs and various other relevant overheads.

Although this would require significant investment by the WEEE recyclers or others, operational plants, such as that manufactured by EMAK (Table 22), are believed to be commercially viable where market conditions are favourable (e.g., gold bullion price).

Additional specialised plant would be required to treat the other types of TM-bearing WEEE, such as lighting, display equipment and computer hard drives, in particular to focus on the recovery of the TMs beyond the PMs, PGMs and tin.

## 13.4 Section summary

Numerous emerging solutions are available for disassembly of PCBs, in order to increase TM recovery, improve cost-benefit of recovery, and divert these components either to channels of reuse, or specialised recovery processes capable of capturing all TMs contained in the PCBs.

Already, commercially available hydrometallurgical plants for PCB recovery that use lower environmental impact leaching systems, and employ biological recovery of metals such as gold, could be purchased and implemented to begin retention of the valuable technology metals in PCBs from UK WEEE. These processes are not yet perfect in terms of the range of technology metals captured, and are currently limited to Cu, PGMs and PMs, however as hydrometallurgical processes, they are flexible and can be used as bolt on additions for process expansion as strategies to deal more effectively with technology metal recovery in components are established. The revenues generated from such facilities that target the currently recoverable TMs may also help to finance further work to recover other TMs.

These may rely on disassembly, sorting of components and grinding of the components, but the flexibility in plants might mean they can be repurposed to accommodate different leaching and recovery strategies for different components as sufficiently large quantities for economical processing are amassed.

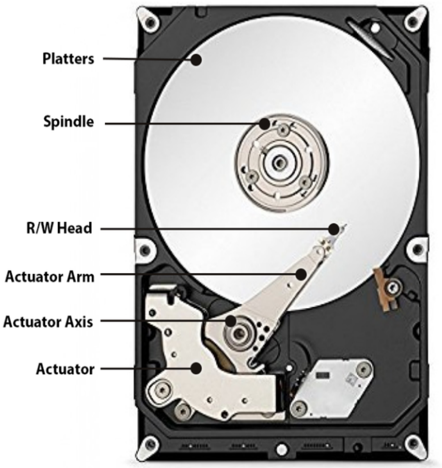
The main disadvantage for some of these processes is the requirement to grind the boards which may mean technology metal losses in the dust, unless these can be recovered and processed.

Appendices B and C details the different PCB disassembly processes and methods to extract technology metals from PCBs.

# 14 Technologies for technology metal recovery from Hard Disk Drives (HDDs)

The recovery of rare earth magnets from hard drives (HDDs) containing the REMs neodymium and dysprosium to-date has been extremely problematic. Data security concerns commonly result in shredding of drives, and prohibitively costly disassembly have meant that these magnets remain unrecovered and technology metals in PCBs are dissipated unless these are manually removed prior to shredding. A common HDD consists of the following components and parts: printed circuit board, aluminium parts (case, platters, lids, and voice coil), steel parts (magnet shoes, lids, and screws), and NIB magnets.

*Figure 34: Hard drive (HDD) parts.*



The concentration of aluminium and iron is more than 80% of the total weight of a typical HDD, whereas the value of materials for recovery is dominated by gold and platinum group metals present at the milligram level in the circuit board. Before any attempt to recover REMs present in HDD magnets can be made, cost-effective means of isolating the magnets from drives are needed, which is not possible with current manual approaches. To overcome this issue, the US Critical Materials Institute (CMI), has developed a system for ultra-high throughput of HDDs, which is exploring automated disassembly, magnet assembly punching and mechanical separation, and value recovery after shredding has taken place (King, 2016). Hitachi has also demonstrated a full HDD recycling plant that automatically disassembles up to 100 HDDs per hour (Hitachi Ltd, 2010), (Baba, k et al 2013). The magnet recovery machine automatically removes and separates the magnets from the demagnetized voice coil motor (VCMs), and the material recovery machine efficiently recovers the different materials from the scrap left over by the HDD dismantler.

The material recovery machine uses a combination of techniques, including a magnetic separator, vibrating filter, and gravity concentration, to separate and recover the different materials from this mixture.

Once isolated, approaches to magnet recycling to recover REMs may be used. Methods to do so reported in the literature vary greatly and include using molten magnesium, electro-discharge and sintering, use of various available acids, thiosulfates, thiourea, chlorides and iodides, and ILs to do so (See Table 23). Manual disassembly by removing the PCBs for precious metal recycling and electrical pad insulators for reuse could be profitable; but a high-throughput, automated HDD dismantling system could substantially reduce the dismantling cost, making PCB, pad insulator, and magnet separation profitable.

Hydrogen decrepitation is a promising method for recovery of NIB magnets. The magnets are exposed to hydrogen gas, which causes them to disintegrate into small particles. Because the nickel coating does not react with hydrogen in the same manner, it can be removed through sieving. The powder can be directly reprocessed into new magnets because the particle size in the powder is almost equivalent to the particle size after jet-milling in primary magnet production. Recycling process efficiency rates of 95% have been reported (Sprecher, Kleijn and Kramer, 2014). This also provides a more circular economy-compatible means to remanufacture magnets for use in HDDs, or manufacture wind turbines and electric motors as demand for these technologies rise. However, the future of the HDD recycling market (and therefore future mass flows of contained elements) is uncertain as solid-state drives (SSD's) increasingly take market share from traditional HDD's.

Table 23 identifies methods for recovering Nd from rare earth magnets.

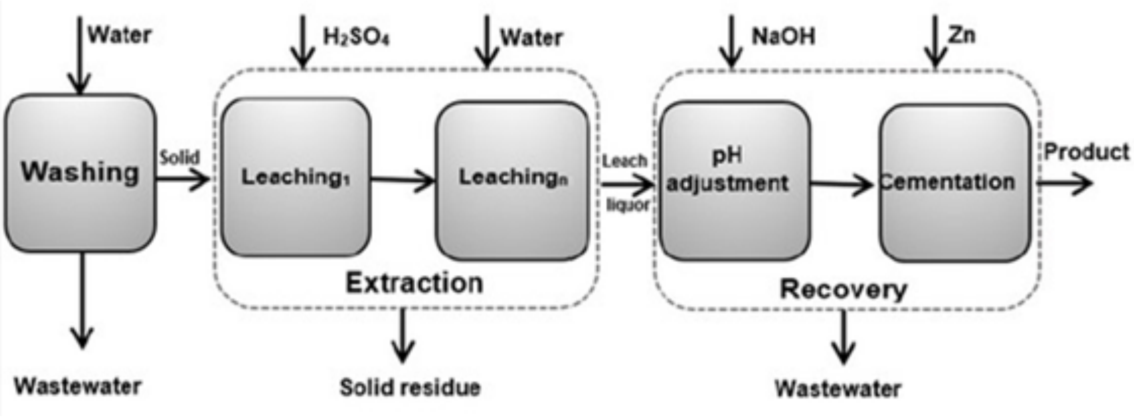
*Table 23: Methods to recover Nd from rare earth magnets.*

| Application | Technology Readiness | Reference |
|---|---|---|
| Recovery of REE from hard drives. | Process fully developed and in use TRL 9 | (Hitachi Ltd, 2010) |
| Prototype reactor for separating NdFeB magnets from WEEE. This used $H_2$ at 1bar to breakdown the rare earth magnets. | Prototype. TRL5. | (Walton *et al.*, no date) |
| Recycling of neodymium magnets using molten magnesium. | lab trials but easily upscaled. TRL 4. | (Anderson, 2016) |
| Simple separation process for neodymium and dysprosium in magnets | Method uses chemicals to separate the two metals. Potential to use on other REE separation. TRL 4. | (Chemie, 2015) |
| NdFeB magnets. Combined leaching/extraction system based on the acidity and thermomorphism of the ionic liquid [Hbet][Tf2N]. | Lab trials. TRL 4 | (Dupont and Binnemans, 2015) |
| NdFeB magnets recovery using oxalic acid. | Lab trials. TRL 4. | (Hoogerstraete et al., 2014) |
| Listing of extraction acids and methods for removal of TMs from NdFeB magnets. | Various available acids, thiosulfates, thiourea, chlorides and iodides. All readily available. TRL 4. | (Sethurajan *et al.*, 2019) |
| Experiments to sort rare earth magnets. The report highlights the issues with different magnets shapes and sizes and magnetism. | Lab trials. TRL 4. | (Holzhauer, Baberg and Spiecker, 2014) |
| Recycling NdFeB magnets by means of electro discharge sintering (EDS). | Lab trials. TRL 4. | (Leich *et al.*, 2019) |
| Roast, leach and solvent extraction from Nd based magnets and by acid dissolution and precipitation. Care as HF acid is used to produce $NdF_3$. | Lab trials. TRL 4. | (Anderson, 2016) |
| NdFeB magnets. A deep-eutectic solvent based on choline chloride and lactic acid (molar ratio 1:2) was used for the leaching of rare earths and other metals from NdFeB magnets. | Lab trials. TRL 4. | (Riaño *et al.*, 2017) |
| Thermal treatment, leaching with sulfuric acid and precipitation of rare earths oxalates using oxalic acid. | Lab trials. TRL 4. | Ippolito *et al.*, 2018) |
| NdFeB magnet leachate Biosorption-based flow-through process for selective CRM recovery from electronic wastes. | Lab trials. TRL3. | (Brewer *et al.*, 2019) |
| Recycling NdFeB magnets by means of electro discharge sintering (EDS). | Lab trials. TRL3. | (Leich *et al.*, 2019) |
| Reviews of Recycling NdFeB magnets by means of hydrogen decrepitation. | Lab trials. TRL3. | (Sprecher, Kleijn and Kramer, 2014) |

# 15 Technologies for TM recovery from flat screens

The current most promising approach to indium recovery from flat screens is the counter current leaching system designed by Rocchetti *et* al. (Rocchetti, Amato and Beolchini, 2016). The process for indium recovery from crushed glass consists of three main steps (Figure 35). A first washing phase with deionised water at room temperature to remove the organic component of LCDs. The second phase for indium extraction was performed using sulfuric acid, at 80 °C for 10 min. This acid was selected because it allowed a lower dissolution of $As_2O_3$, a highly toxic compound, compared to nitric and hydrochloric acids. With the purpose to set up a cross-leaching configuration with two steps, the leach liquor produced in the first step was filtered, restored with a small amount of fresh sulphuric acid and used to treat a second batch of crushed glass. The final step of indium recovery consisted of cementation by zinc powder addition.

*Figure 35: Counter-current leaching system for indium recovery from LCD glass.*



Other methods demonstrated (Table 24) include separation of indium dissolved in nitric acid, hydrochloric acid or sulphuric acid and extraction from mixed metal solutions by solvent extraction with 0.2 M Cyanex 923 in toluene. Another method uses the ground glass plates (with dimension smaller than 1.0 mm) which are mixed with ammonium chloride (50% of glass powder), at reducing pressure, to obtain indium chloride with a purity higher than 99% and an efficiency of ~90%. These two methods are covered in a range of patents reviewed (Amato and Beolchini, 2018)

## 15.1 LEDs

There was only one report on research into the recovery of Ga from LEDs, which is unsurprising considering the previously cited barriers to technology metal recovery from LEDs (Ueberschaar, Otto and Rotter, 2017). A thermal pre-treatment of the chips followed by a manual separation allowed an isolation of gallium rich fractions, with gallium mass fractions up to 35% (Ueberschaar, Otto and Rotter, 2017; Sethurajan *et al.*, 2019). Here, gallium loads per chip were between 0.9 and 1.3 mg. Copper, gold and arsenic were determined as well. Further treatment options for this gallium-rich fraction were assessed. The conventional pyrometallurgical copper route is unlikely to be feasible due to the likely partitioning of Ga into the slag during smelting. Gold is present in the LEDs and so a selective separation prior to processing is necessary to avoid losses of gallium. This may also be necessary to upgrade the gallium fraction to achieve sufficiently high concentration for economic recovery. Diluted with other materials, the gallium content would be too low.

Recolight and partners[29] have received funding in November 2020 to also recover Ga from LEDs.

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[29] https://www.recolight.co.uk/research-project-launched-to-recover-gallium-from-waste-led-lamps/

*Table 24: Methods for recovery of TMs in LCDs.*

| TM recovery | Application | Technology Readiness | Reference |
|---|---|---|---|
| Eu, Gd Tb, Y | Recovery of rare earth elements from powder of CRT, fluorescent lamps and LCDs. | Pilot mobile plant available HydroWEEE TRL 9. | (Altimari *et al.*, 2014)<br>(Beolchini *et al.*, 2014) |
| In and Y | The recovery of CRMs such as indium and yttrium by recycling discarded flat panels. | Pilot plant TRL 8 | (Life-Recumental, 2015) |
| In, Nd, Y | RECYVAL- NANO Develop an innovative recycling process for recovery and reuse of indium, yttrium and neodymium metals from Flat Panel Displays. | Pilot plant. TRL 5. | http://recyval-nano.eu/ |
| In | Production of Indium concentrate from waste LCD Screens. | Production ready method. TRL5. | (Boundy, Spiller and Taylor, no date) |
| In | Indium leaching from LCD screens. Various available acids recovering over 90%. | Lab trials. TRL 4. | (Sethurajan *et al.*, 2019) |
| Co, Ga, Ge, In, | SCRREEN: CRMs from secondary resources from a wide range of WEEE including LEDs, fluorescent tubes, NiMH batteries, ECs etc. | Lab trials. TRL 4. | (Okvist *et al.*, 2020) |
| In | In recovery from LCD panels using acids. Process parameters for high yield recovery of indium from LCD glass were investigated and results indicate that the process can be employed for large-scale sustainable recovery. | Lab trials. TRL 4. | (Felix *et al.*, 2012) |
| In | Recovered indium by means of cementation from a leaching solution of waste liquid crystal display panels. Cementation with zinc powder was optimized through the investigation of the effects of different variables (zinc concentration, pH, cementation time) on cementation efficiency and purity of the solid product. | Lab trials. TRL 4. | (Rocchetti, Amato and Beolchini, 2016) |
| In | The recovery of Indium from LCD panels using acid and zinc cementation. | Lab trials. TRL 4. | (Rocchetti, Amato and Beolchini, 2016) |
| Ce, u, La, Gd, Tb, Y | Patent review of technologies. | Review of patents | (Amato and Beolchini, 2018) |

# 16 Technologies for recovery of TMs from Lighting

REM-based phosphor powders use varying amounts of REMs, resulting in a wide variety of powder compositions with different proportions of yttrium, europium and terbium to generate red, green and blue phosphors. To recover the REMs, the phosphor mixture has to be chemically leached to extract the REMs into solution before they are precipitated by addition of oxalic acid or solvent extracted with solvents. The low recovery efficiency of REMs results from high acid resistance of some type of phosphors, mainly green.

Over 6 tonnes of indium are contained in the flat screens of monitors, TVs and laptops that are generated annually in UK WEEE. Most of this is lost in the processing of the screens. The main method for the extraction of indium is by means of crushing the glass to powder and washing the coating of the glass or leaching it in acid followed by cementation (precipitation of the TMs). This will allow this material to be reused back into new screens.

Over 9 tonnes of lanthanum and 2 tonnes of europium is disposed of annually in the fluorescent tube lighting annually in the UK. To recover the rare earth metals, the phosphor mixture must be chemically attacked to bring the REMs into solution and the REMs are recovered from the solution by precipitation (oxalic acid) or solvent extraction.

The Solvay recovery method first removes the mercury, glass and other components to physically liberate the rare earth concentrate, which is then sent to the chemical separation plant (Solvay and Loop, 2014) in France. There, the halophosphates are removed and the phosphors are cracked resulting in a REM concentrate that can be fed, as in primary REM production, into a solvent extraction process for separation and recovery of the individual REMs. They claim a final yield of REE of ~80%. Currently Solvay recycles several hundred tonnes of rare earth phosphors each year, primarily from Europe.

Numerous methods have been trialled in the literature (Table 25). All these methods require fluorescent lamps to be crushed and phosphors leached using solutions to dissolve the TMs with different methods for recovery from solution.

*Table 25: Methods to recover TMs from lighting.*

| TM recovery | Application | Technology Readiness | Reference |
|---|---|---|---|
| Co, In, Y | HydroWEEE developed innovative plant technology that uses liquid solvents to extract metals including REEs in high purity from electronic waste. | Pilot mobile plant available. TRL 9. | http://www.4980.timewarp.at/sat/HydroWEEE/<br>(Vegliò *et al.*, 2014) |
| Eu, La, Lu, Y | Solvay project: An innovative process of rare earth recycling from used fluorescent lamps. | Lab trials. TRL 8. | https://www.solvay.com/en/innovation/open-innovation/european-life-projects/loop-life-project |
| Eu, La, Lu, Y, and Sc | Separation of CRMs such as Eu and Y from phosphor waste from compact fluorescent light bulbs by using different leaching methods. | Lab trials. TRL 4. | (Bogart *et al.*, 2016) |
| Eu, La, Lu, Y, and Sc | Fluorescent tubes. | Review of potential use of IL solutions only all lab based. TRL 4. | (Bogart *et al.*, 2016) |
| Eu, La, Lu, Y, and Sc | Listing of extraction acids and methods for removal of CRMs from Fluorescent tubes. | Various available acids, thiosulfates, thiourea, chlorides and iodides processes for CRM recovery and various methods for precipitation. All readily available. TRL 4. | (Sethurajan *et al.*, 2019) |
| In, Co, Ga, Ge, No, PGMs | SCRREEN: Production technologies of CRM from secondary resources from a wide range of WEEE including LEDs, fluorescent tubes, NiMH batteries, ECs etc. | Review of methods, various TRLs. | (Okvist *et al.*, 2020) |
| Ce, Gd, La, Tb, Y | Recovery of phosphors in fluorescent lamps by two liquid flotation and leaching processes. | Solutions readily available. TRL 4. | (Binnemans *et al.*, 2013) |
| Eu, Tb, Y | Review of methods for recovery and supply and demand. | N/A | (Machacek *et al.*, 2015) |
| Ce, Eu, Gd, La, Tb, Y | Thermal treatment, leaching with sulfuric acid and precipitation of rare earths oxalates using oxalic acid. | Lab trials. TRL 4. | (Ippolito *et al.*, 2018) |
| Ce, Gd, La, Tb, Y | Fluorescent tubes separation for groups of rare earths by solvent extraction with (2-ethylhexyl) phosphoric acid (D2EHPA), stripping with acid and precipitation with oxalic acid. | Lab scale. TRL 4. | (Ippolito *et al.*, 2018). |
| Eu, Gd, Y, Tb, La | Waste fluorescent phosphor treatment using IL. | Review of potential use of IL solutions only all lab based. TRL 4. | Schaeffer, N. et al. (2018a) 018a) |
| Eu, Y | Lighting, using ionic liquids.<br>Reviews different acid leaching processes. Showed the ability to selectively dissolve REE oxides ($Y_2O^3$ and $Eu_2O3$). | Wide range of different methods the majority could be used immediately. TRL 4. | (Sethurajan *et al.*, 2019) |

# Appendix A: Technologies for the recovery of TMs from PCBs

*Table 26: Technologies for the recovery TMs from PCBs.*

| TM recovery | Application | Technology readiness | References |
|---|---|---|---|
| Potentially all TMs in PCBs. | Plasma arc for shredded PCBs. Output needs further processing – refining/reagents to recover TMs. | Commercially available. TRL 9. | (Tetronics, no date;<br>Tetronics, 2016;<br>Tetronics, 2007;<br>Dowling, 2014a) |
| Pre -treatment. | Plasma arc for shredded PCBs. Output needs further processing – refining/reagents to recover TMs. | Commercially available. TRL 9. | (Johnson, Deegan and Wise, 2012;<br>Tetronics, no date;<br>Dowling, 2014b;<br>Dowling, 2014a;<br>Tetronics, 2007;<br>Tetronics, 2016) |
| PMs and PGMs. | Review of different technologies. | Review of current and developing processes. TRL4 to 9 | (Kaya, 2020b) |
| Pd and PMs. | PCB metal recovery. Various acids trialled. | Could be used<br>in current acid treatment tanks. TRL 4 -9. | (U. Jadhav and Hocheng, 2015) |
| Eu, Ce, Dy, Gd, La, PGMs, Nd, Y. | Listing of extraction acids and methods for removal of CRMs from Fluorescent tubes, NdFeB magnets, NiMH batteries and PCBs. Various available acids, thiosulfates, thiourea, chlorides and iodides processes for CRM recovery and various methods for precipitation. | All leachates readily available. TRL 4. | (Sethurajan *et al.*, 2019) |
| Pre-treatment. | Microwave melting of PCBs. Output was then processed using leach reactor. Mo, Cr, Ti, V, Ta, PMs recovered. | Commercial microwave used. TRL 4. | (Soare *et al.*, 2016) |
| Pt and PMs. | Mixing car catalysts and PCBs to recovery more Pt using Cu as the collector. | Trial using existing recovery methods. Can be scaled. Pt recovery lower than expected. TRL 4. | (Willner *et al.*, 2014) |

| TM recovery | Application | Technology readiness | References |
|---|---|---|---|
| PM and PGMs | Review of TMs on PCBs are treatment. | Lab analysis. TRL 4. | (Bizzo, Figueiredo and De Andrade, 2014;<br>Delfini *et al.*, 2011a) |
| Au, Pd and Pt | Review of where these metals are on PCBs. | Lab analysis. TRL 4. | (Delfini *et al.*, 2011b) |
| Au, Pu. | Recovery of metals from PCBs using different acids. | Lab trials. TRL 4. | (U Jadhav and Hocheng, 2015) |
| PMs and PGMs. | Current status on leaching precious metals from waste printed circuit boards. | Review of current processes. TRL 3 to 4. | (Zhang *et al.*, 2012) |
| Pre- treatment. | IR heating of boards to remove ECs. | TRL 4. | (Park *et al.*, 2015) |
| Pre-treatment. | Removal of solder via heat and centrifugal force and then pyrolysis. | TRL 4. | (Zhou and Qiu, 2010) |
| Ta. | Oxidation of capacitors and recovery of Ta. | Lab trials. TRL 4. | (Matsuoka, Mineta and Okabe, 2004) |
| Ta. | Hydrometallurgical Process for Tantalum Recovery from Epoxy-Coated Solid Electrolyte Tantalum Capacitors. Beware uses of Hydrofluoric Acid. | Lab scale.<br>TRL 4. | (Chen and Ho, 2019) |
| Ta. | Breakdown of organic resins covering the capacitor. Research on time, pressure, and addition of $H_2O_2$. | TRL4. Commercial plant available to operate at the required temperatures and pressures. | Niu, Chen and Xu, 2017) |
| Ta. | Review of recovery from capacitors. | Review of technologies. TRL 3 and 4. | (Ueberschaar, Jalalpoor and Korf, 2017) |
| Ta. | Oxidation of capacitors and recovery of Ta. | Lab trials. TRL 4. | (Matsuoka, Mineta and Okabe, 2004) |
| Ta. | Review of recovery from capacitors. | Lab scale with further research required in purification of Ta. TRL | (Mineta and Okabe, no date) |
| Au, Ce, Eu, Dy, La, Nd, Y. | Recovery of CRMs from WEEE shredding dust. This study investigated the recovery potential of a multi-step leaching process to extract REEs, from the dust produced during the industrial shredding treatment of WEEE using $H_2SO_4$ and $H_2O_2$. | TRL 4. Can be used in existing chemical tanks | (Marra, Cesaro and Belgiorno, 2019) |
| Au, Co. | Bioleaching using 5 different mixed cultures of cyanogenic bacteria were evaluated. | Lab trials but Suitable for use using readily available equipment. TRL4. | (Hursthouse *et al.*, 2018) |
| Co, Dy, Eu, La, Nd, PMs, Se, Yb. | Extraction from different solutions, battery waste and WEEE. Lab trials using different microbial biotechnology. | TRL3. | (Yarlagadda, Mohan and Lens, 2016). |

| TM recovery | Application | Technology readiness | References |
|---|---|---|---|
| Ag, Au, Co. | Bioleaching performed with three types of microbes (acidophiles, fungus and cyanogenic) to determine leaching selectivity. High leaching rate but slow process. | Lab trials using different microbial biotechnology. TRL3. | (WRAP, 2015, 2018). |
| Pre-treatment. | Microwave melting of PCBs. Output was then processed using leach reactor.<br>Mo, Cr, Ti, V, Ta, PMs recovered. | Commercial microwave used. TRL4. | (Soare *et al.*, 2016) |

# Appendix B: Technologies for PCB disassembly processes

*Table 27*: *PCB disassembly processes reported in literature, outcomes of studies, and comments of potential for application at industrial scale.*

| Type | Process | Outcome and Technology readiness | Advantages | Disadvantages | References |
|---|---|---|---|---|---|
| | Removal of components from PCBs using manual methods. | Simple manual methods. TRL6 to 9 | | | (Delfini et al., 2011a) |
| Pilot scale, thermal. | PCB from TVs and PCs tested.<br>This system contains two parts: Ecs automatic disassembly and off-gas purification<br>experimental temperature in this study is adjusted at 265 ± 5 °C.<br>Results show that 8 min was the optimal incubation period (i.e., time to ramp to temperature and complete EC removal)<br>Rotating speed at 6rpm<br>Total electric power of off-gas system is 29 kW. | In summary, a typical thoroughly procedure for TV-WPCBs dismantling is suggested: (1) dismantling LOTs manually from TV-WPCBs firstly. (2) Feeding the TV-WPCBs, which have been already removed LOTs, into the Ecs-ADM, then separating the solder from TV-WPCBs, and the Ecs, which are without pins or whose pins are not bended, could be dislodged simultaneously, under the conditions of 265 ± 5 °C, 8 min, and sufficient vibration. (3) Pulling out the Ecs, which are not dislodged in step (2), from TV-WPCBs (discharged from the Ecs-ADM) manually by pliers or other tools.<br>PC-WPCBs before and after disassembly when the dismantling temperature, rotating | EC recovered intact from FR4 boards (where all the TMs are)<br>Potential for reuse<br>Off-gas system using activated carbon scrubs any toxins evolved by PCB heating (but claim few are evolved at temp below 270-280°C).<br>Cost about $4.61/tonne of boards to process, should be very viable in terms of mass-based refining charge savings on subsequent recovery.<br>Automatic feeding, automatic discharging and continuous production. | The exact and detailed composition of the off gas should be analysed, which is the further work being pursued. | Wang et al., 2016 |

| Type | Process | Outcome and Technology readiness | Advantages | Disadvantages | References |
|---|---|---|---|---|---|
|  |  | speed, and incubation period were 265 ±5oC, 10 rpm, and 8 min, respectively.<br>Almost all ECs are dislodged successfully from the PC-WPCBs. And the solder is also separated completely from the PC-WPCB and shows that no EC was damaged.<br>Different from TV-WPCBs, there are no ECs with bended pins on PC-WPCBs as the technology of wave soldering is employed for almost all of WPCBs (Richard, 2009, Pedro et al., 2003). Hence, ECs on the PC-WPCBs could be dismantled completely by ECs-ADM without extra process.<br>TRL5 |  |  |  |
| Thermal - Solder dissolution using ionic liquid (IL) (lab scale) | Dismantling of WPCBs using water-soluble ionic liquid Submerge PCBs in IL and heat with oil bath and stir in the absence of oxygen/air to avoid generation of hazardous compounds.<br>[BMIm]BF4 was employed as the heating medium due to the wide temperature range | ~90% of component liberated under optimal conditions, Sn solder also recovered.<br>Optimum conditions determined as 250 C heating temperature, 12 min retention time, 45 rpm rotation rate. | Possibilities for industrial scale up<br>Lower environmental impact than alternative thermal approaches used in informal sector.<br>Ionic liquid is lower environmental impact solvent with favourable | This is a collective disassembly approach requiring further sorting of isolated SMDs.<br>Low levels of benzene and methyl benzene generated. | Zeng et al., 2013<br>(Zeng et al., 2013) |

| Type | Process | Outcome and Technology readiness | Advantages | Disadvantages | References |
|---|---|---|---|---|---|
| | of its liquid state, its suitable viscosity and the low expense<br>dismantling efficiency elevated by mechanical vibration from rotor to supplement the bathing.<br>Dissolved solder precipitates upon cooling.<br>Filter and wash solder and ECs with DI water. | Industrial system proposed.<br>TRL4. | properties such as extremely low vapor pressure, low combustibility, excellent thermal stability, and a wide temperature range in its liquid state.<br>Expensive IL can be reused and used sparingly with proposed spray system to reduce costs.<br>Easier to collect solder than with manual dismantling.<br>Only minute organic pollutants as benzene and methylbenzene were generated by the heating process and the pollutants could be captured by the fume cupboard and absorbed by activated carbon.<br>More cost effective than manual and semi-automated mechanical approaches for quantities >1kt and >3kt, respectively. | IL is costly and the biggest bottleneck to proposed process.<br>Less cost-effective than manual processes when processing <1kt of PCBs. | |
| Chemical: Solder dissolution with IL. (Lab scale) | Use IL [EMIN][BF4-] as heating medium.<br>240 °C, 150 rpm stirring for 10 minutes. | Components manually removed prior to study but would be effective at liberating SMDs as total | As above<br>Extending dwell time to 30 mins (@240°C) starts delamination of board, 10 | As above<br>Gas pollutants may be evolved at T>260°C | Zhu et al., 2012; Ping Zhu et al., 2012) |

| Type | Process | Outcome and Technology readiness | Advantages | Disadvantages | References |
|---|---|---|---|---|---|
| | | solder dissolution occurs. (P Zhu et al., 2012)<br>TRL 4. | mins @ 260°C enables Cu foil and fibre glass recovery following delamination and total dissolution of flame retarded epoxy – opportunity to valorise whole board in 1 process. | Some copper wires remain in the fibre glass components. | |
| Mechanical abrasion of joints on revers of boards with subsequent component separation. | Disassembly of EECs from wasted PCBs (WPCBs) and the physical separation of these EECs using a self-designed disassembling apparatus<br>Mechanical abrasion of back side of boards.<br>3-step separation process of sieving, magnetic separation, and dense medium separation | The disassembling efficiencies were evaluated by using the ratio of grinding area (E area) and the weight ratio of the detached EECs (Eweight).<br>97.7% (Earea) and 98% (Eweight) could be accomplished by 3 repetitive treatments at optimum grinder speed of 5500 rpm grinder height of 1.5 mm.<br>most groups of the EECs (except for the diode, transistor, and IC chip groups) could be sorted at a relatively high separation efficiency of about 75% or more.<br>Separation efficiency with regard to the elemental composition, the distribution ratio (Rdis) and the concentration ratio (Rconc) were used. | Higher component liberation efficiencies than those reported for IL processes by Zeng et al. (Zeng et al., 2013).<br>Separation processes enable concentration of TMs into distinct output fractions for downstream recovery. Recyclability of the elements is highly feasible, even though the initial content in EECs is lower than several tens of mg/kg. | Potential evolution of hazardous gases if boards heat in the presence of air.<br>Dust from grinding. (~14% of total board mas as dust, components isolated are only 17.5%) Therefore mitigation plant may be necessary, dust is always explosion risk.<br>May not be so suitable for FR4 boars, this was done on an FR2 from a VCR which had through hole components. | Lee et al., 2012; Lee, Kim and J. Lee, 2012) |

| Type | Process | Outcome and Technology readiness | Advantages | Disadvantages | References |
|---|---|---|---|---|---|
|  |  | 15 elements could be separated with the highest Rdis and Rconc in the same separated division. This result implies that the recyclability of the elements is highly feasible, even though the initial content in EECs is lower than several tens of mg/kg.<br>Most of the EECs (over 95%) can be recovered in a non-destructive state.<br>These EECs contain 17 groups and can be classified into 54 types based on their shapes and sizes.<br>TRL 4. |  |  |  |
| Thermal: centrifugal separation +vacuum pyrolysis (lab scale) | The recycling process contained centrifugal separation and vacuum pyrolysis. Diesel oil was used as heating medium to melt the solder of WPCBs, and then the molten solder was recycled efficiently by the centrifugal force.<br>After centrifugal separation, the two types of WPCBs without solder were pyrolysed under vacuum condition at 600°C for 30 min. | The results showed that the separation of solder from WPCBs was complete when WPCBs were heated at 240 °C, and the rotating drum was rotated at 1400 rpm for 6 min intermittently.<br>The type-A WPCBs (FR2) without solder was pyrolysed to form an average of 69.5 wt% residue, 27.8 wt% oil, and 2.7 wt% gas. Pyrolysis of the type-B (FR2) without | Mass reduction and concentration of TMs.<br>Valorisation of board itself.<br>Complete solder removal so component isolation should be close to ~100% if method was to be used for component removal without vacuum pyrolysis. |  | Zhou & Qiu, 2010 |

| Type | Process | Outcome and Technology readiness | Advantages | Disadvantages | References |
|---|---|---|---|---|---|
| | | solder of WPCBs led to an average mass balance of 75.7 wt% residue, 20.0 wt% oil, and 4.3 wt% gas. The pyrolysis residue contained various metals, glass fibres and other inorganic materials, which could be recycled after further treatment. The results of FT-IR analysis indicated that most of the pyrolysis oil consisted of phenols and substituted phenols. Therefore, the pyrolysis oil can be used for fuel or chemical feedstock for further processing.<br>The pyrolysis gas consisted mainly of CO, CO2, CH4, and H2, which could be collected and combusted for the pyrolysis self-sustain. TRL 4. | | | |
| Thermo-mechanical: IR heating of PCBs with mechanical abrasion (lab/pilot) | Various laptop PCBs treated Disassembly apparatus includes IR heating with wire brushes for mechanical abrasion.<br>Process repeated 3 times for total removal of ECs | 94% EC removal.<br>The removed parts were ECs connected via the THT, rivet and screwed joints, which can be removed regardless of temperature or feeding speed. When the feeding | Deals with through-hole, riveted and other types of components that are difficult to remove with desoldering alone by crushing them. Normally this requires costly manual removal before desoldering.7 | To realise an entire PCBA recycling process, it is also important to sort the ECs based on their metal composition following the disassembling treatment. | Park et al., 2015 |

| Type | Process | Outcome and Technology readiness | Advantages | Disadvantages | References |
|---|---|---|---|---|---|
|  |  | speed was slower than 1 cm/s, the ratio increased exponentially with temperature. On the other hand, if the PCBAs were heated long enough to melt the solder, a complete disassembly could be achieved. Interestingly, the ratio showed a steep rise up to 94% as the heating temperature exceeded 225 °C.<br>No further increase in disassembly ratio above 250°C regardless of solder type (and generates toxics as approach 280°C)<br>TRL 4. | Due to high disassembly performance and time-saving feature of the new apparatus, the current technique is expected to become an essential part of a fully automated recycling system. |  |  |
| Various heat methods | Removal of components from PCBs using different heat methods. | Equipment readily available. TRL4. |  |  | (Charles et al., 2019) |

# Appendix C: Technology Metal Recovery Technologies for Batteries

This section provides an overview of commercial and pipeline technologies for recovery of TMs from lithium-ion batteries (LIBs), highlighting inherent issues with existing processes, and advantageous features to maximise the long-term sustainable recovery of TMs from LIBs for the purpose of providing an evidence base for development of LIB recovery processes for implementation in the UK.

## Background

Li-ion batteries (LIBs) contain a range of TMs including Li, Co, graphite, and REMs such as Y in certain chemistries e.g., lithium-iron-yttrium-phosphate (LFYP) batteries. In general, batteries are composed of a positive and negative electrode, separated by a porous polymer film, filled with electrolyte (Figure 36). Anodes are most commonly composed of carbon/graphite with a binder such as PVDF on a copper foil current collector. Cathodes are composed of a lithium transition metal oxide (e.g., $LiCoO_2$) and binder on an aluminium foil current collector.

Electrodes are separated by a porous polyolefin film. Electrolytes consist of a conducting Li salt (1 M) dissolved in a mixture of linear carbonates and a cyclic carbonate solvent. The electrodes are rolled or folded before being placed in either a plastic or metallic shell to create cylinder, prism and packet LIBs (Figure 37, a-d). Cells may be used individually in devices or connected together to form a battery pack for applications such as laptops (Figure 37, e), EVs and stationary energy storage applications.

Material compositions of typical mobile phone and laptop LIBs reported in literature are given in Table 31, with quantities of materials in a typical $LiCoO_2$ battery shown in Figure 38 Innovation and pursuit of lower cost, higher efficiency, longer battery life and denser energy storage has resulted in use of a great variety of in electrolytes and electrode materials in commercially available LIBs today (Table 32).

*Figure 36: Internal components of a Li-ion battery.*

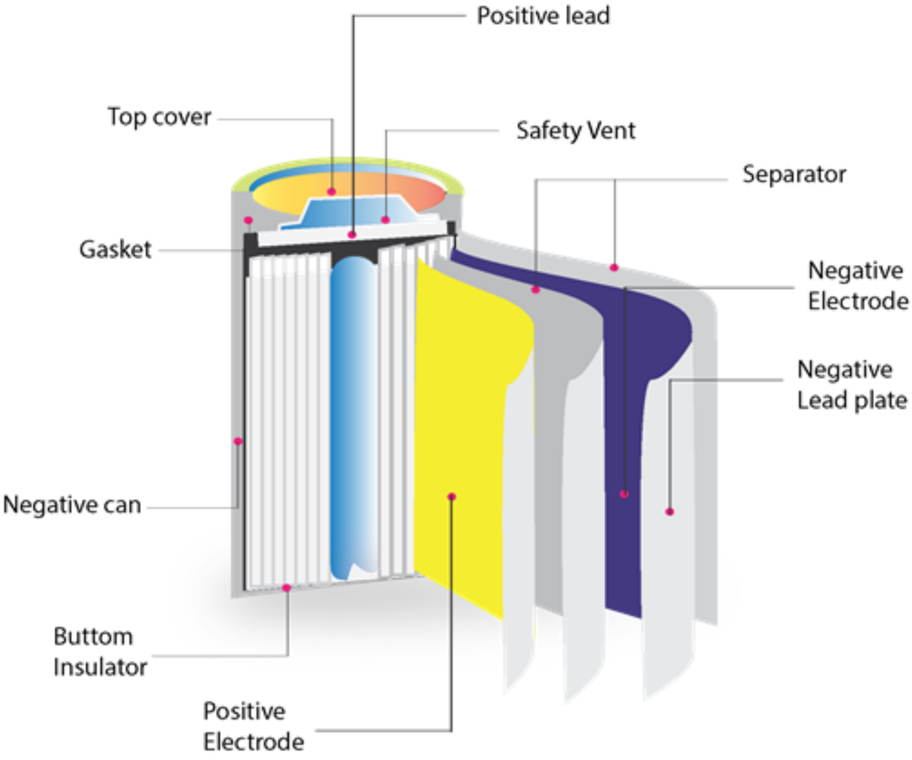


*Figure 37: Different battery types: a) cylinder cell from a laptop battery pack; b) prism cell from a digital camera; c) packet cell from mp3 player; d) prism cell from a mobile phone; e) laptop battery pack with its six contained cylinder cells and PCB shown.*

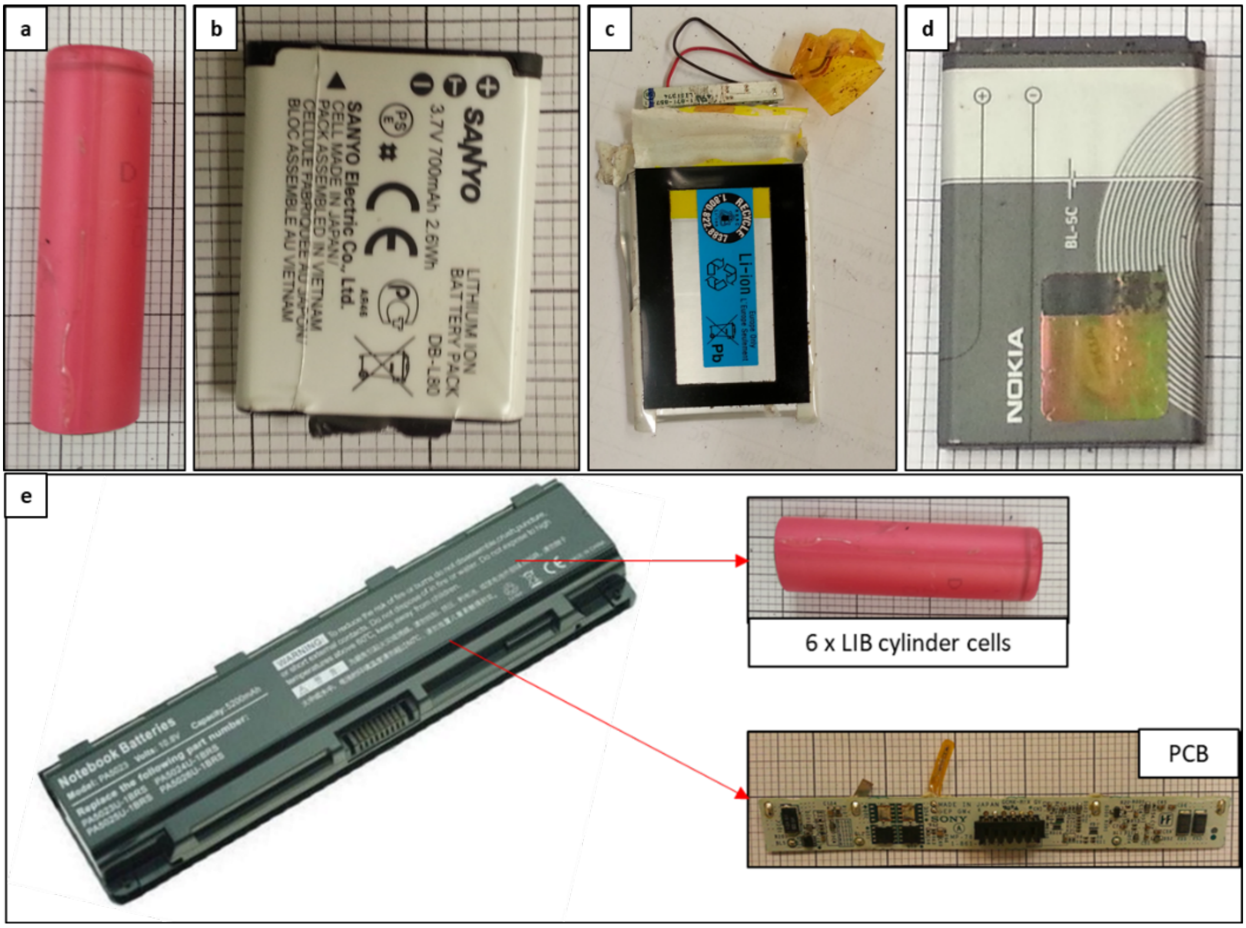


*Table 28: Material composition of Li-ion mobile phone and laptop batteries reported in literature (Meshram* et al.*, 2020a).*

| **Component** | **Mobile phones**<br>(Rojas and Zea, 2016) | | **Laptops**<br>(Qadir and Gulshan, 2018) | |
|---|---|---|---|---|
| | **Mass (g)** | **wt%** | **Mass (g)** | **wt%** |
| Total battery assembly | 17.8 | - | 316 | - |
| Plastic shell | 1.8 | 5.2 | 50 | 15.8 |
| Metallic shell | 4.3 | 12.2 | 51 | 16.1 |
| Cu electrode | 3.5 | 9.8 | 17.2 | 5.4 |
| Al electrode | - | - | 7.5 | 2.4 |
| Cathode materials | 5.4 | 15.1 | 130.9 | 41.4 |
| Polymer | 0.6 | 1.8 | 6.8 | 2.2 |
| Electrolyte | 1.8 | 5.0 | 20.9 | 6.6 |

*Figure 38: Bill of Materials for $LiCoO_2$ battery (wt%) (Larouche* et al.*, 2020).*

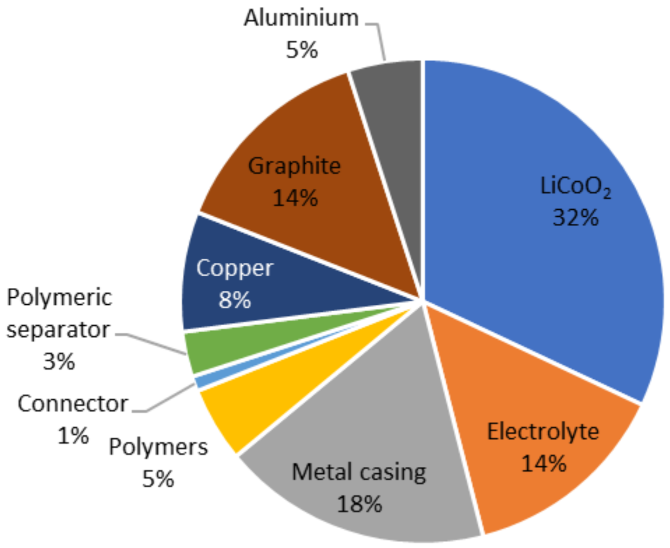


*Table 29: non-exhaustive list of Li-ion battery materials in use today.*

| Cell component | Typical materials in use today |
|---|---|
| Active cathode material | $LiCoO_2$; $LiMn_2O_4$ (spinel); $LiNiO_2$; $LiFePO_4$ (olivine); $Li_2FePO_4F$; $LiCo_{1/3}Ni_{1/3}Mn_{1/3}O$; Li ($(Li_aNi_xMn_yCo_z)O_2$; $LiNi_yMn_yCo_{1-2y}O_2$; $LiMn_{1.5}Ni_{0.5}O_4$. |
| Active anode material | Graphite/carbon; lithium titanate ($Li_4Ti_5O_{12}$). |
| Binders | Polyvinylidene fluoride (PVDF); sodium salt of carboxymethyl cellulose; styrene-butadiene rubber. |
| Electrolyte solvent | Linear organics: dimethyl carbonate (DMC); ethyl methyl carbonate (EMC); diethyl carbonate (DEC).<br>Cyclic organics: propylene carbonate; ethylene carbonate, dioxolane. |
| Electrolyte salt | $LiPF_6$; $LiBF_4$; $LiAsF_6$; $LiCF_3SO_3$; Li $(SO_2CF_3)_2$; $LiClO_4$; LiBETl; LiTFSl; LiTf; LiTFSM. |
| Polymer separator foil | PP, PE, or PE/PP. |
| Shell | Stainless steel; nickel plated steel; aluminium; plastic |

LIBs contain a large variety of TMs, in relatively high concentration in comparison to traces of TMs used in other EEE applications (e.g., Ta in PCBs). This is promising in terms of the economics of their eventual recovery. Demand for graphite, lithium and cathode metals such as Co for LIBs is increasing with wider penetration of EEE in consumer and industrial markets, and deployment of green technologies such as hybrid/electric vehicles ((H)EVs) and vital storage for peak energy saving in renewable energy systems.

The demand from LIB production that has led to significant pressure on primary supply of Co, graphite and Li and the resulting 'critical' status of these materials. Greater recovery of these materials from secondary sources including waste batteries, with a view to reuse and cascade EoL batteries into alternative applications (e.g., EV batteries into stationary energy storage applications) are the main strategies employed to mitigate criticality for these materials. This section considers existing and emerging technologies for LIB recycling, including any limitations or benefits in terms of TM recovery and the future sustainability of these technologies.

Post-consumer batteries present a range of environmental health and safety issues due to their hazardous material content. Toxic heavy metals such as Co, As, F and sulfonated compounds of electrolytes, and numerous highly toxic electrolyte degradation products such as organic fluorophosphates can persist in the environment, and impact on human health through accumulation in the biosphere. These materials, particularly organoflurophosphates also represent considerable risks to recycling workers through ingestion, inhalation and skin contact. In addition, highly flammable Li metal and the presence of flammable organic solvents present additional explosion risks during storage, handling, transport and treatment of waste LIBs.

The toxic degradation products and Li metal dendrites which are not in electrical contact with electrodes of the cell and cannot be oxidised to $Li^+$ during discharge prior to recycling, are generated during the use phase of LIBs and so are absent from virgin batteries. For this reason, used LIBs have unique hazards absent before use in application.

Collected batteries may have leaks, so initial collection may be performed in a dry, cool, ventilated area fitted with non-aqueous fire-suppression equipment at hand. Although leaky cells are unlikely, fire may potentially result when lithium oxidises are exposed to air or moisture. LIBs have a tendency to catch fire and/or explode during mechanical pre-treatment, even after deep discharge, when Li metal produced during overcharging is exposed to air and mechanical shock. In addition, short circuiting of cells with retained charge during mechanical pre-treatment of batteries also results in exothermic reactions which can cause fire and explosion (particularly in the case of $SO_2$ and $SOCl_2$ chemistries).

Recycling must eliminate any risk to human health and the environment posed by these hazardous materials, and for this reason discharge and pre-treatment methods are crucial steps in recycling in which risks of detrimental impacts on human health of workers, the population in general and the environment must be mitigated. Deep discharge of cells and mechanical pre-treatment, which takes account of necessary health and safety measures, must be a priority of any LIB recycling process. Such a process renders LIBs safe for further processing, reduces scrap volume, separates battery components and enriches valuable materials (Shin *et al.*, 2005).

The most valuable cell components are cathode materials, followed by electrolytes and current collector metals (Al and Cu). This is due to the content of valuable metals in cathodic materials and current collector foils and the value of high-grade lithium salts in electrolytes. For this reason, traditional LIB recycling has been conducted via smelting to collect the most valuable cathode transition metals (e.g., Co and Mn) and copper while sacrificing other cell components such as Al and Li which segregate into slag during smelting, and organic materials including electrolytes (despite the value of contained Li salts) and polymer components such as the porous polyolefin separators, which are burned off in the smelting process.

The recycling processes should seek to maximise economic performance to ensure long term viability of recycling, and this is best achieved through closed-loop recycling of all materials present i.e., recovery of materials as added-value raw materials for LIB manufacturing. To achieve this, materials should be recovered in a form suitable for direct inclusion in LIB manufacturing, with a maximum range of materials recovered. This will maximise revenue from sale of recovered materials and may also result in reduced materials costs for manufacturers in comparison with purchasing primary raw materials as recovered material are often obtained at lower economic and environmental cost than they can be acquired from primary sources (Table 33). Recycling in this way is conducive to circular economy, and will therefore result in economic, social and environmental benefits (Foundation, 2013) while mitigating materials criticality issues for the LIB sector through generation of secondary raw materials.

*Table 30: Economic, energy and environmental savings from recycling cathode materials from end-of-life Li-ion batteries relative to the use of virgin materials (data taken from Steward et al. (Steward, Mayyas and Mann, 2019)).*

| | **Cost** | | | | **Energy** | | | **GWP ($CO_{2\text{-}eq}$)** | | |
|---|---|---|---|---|---|---|---|---|---|---|
| **LIB type** | **LCO** | **NMC333** | **NMC811** | **LFP** | **LCO** | **LMO** | **LCO** | **NMC333** | **LMO** | **LCO** |
| Virgin Raw Materials | \$62 | \$45 | \$40 | \$32 | 77 MJ/kg | 34 MJ/kg | 200 kWh/kg | 9 $kgCO_{2\text{-}eq}$/kg cell | 5 $kgCO_{2\text{-}eq}$/kg cell | 11 $kgCO_{2\text{-}eq}$/kg material |
| Pyrometallurgy | 38% | 6% | 5% more | | 35% | | 70% | 78% | 70% | |
| Hydrometallurgy | 41% | 13% | 1% | | 38% | 18% | | | 5% | |
| Direct recycling ($scCO_2$) | 43% | 27% | 16% | 15% | 5% | 76% | | 94% | 10% | |

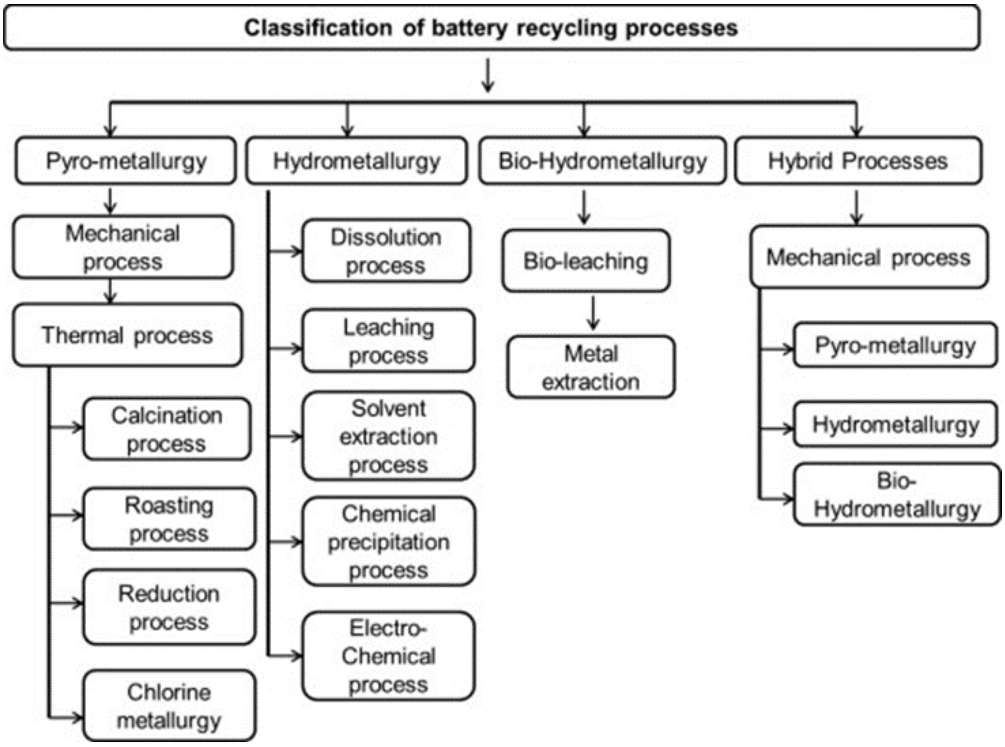
## Current Recycling Practices

This section reviews currently available commercial processes for LIB recycling, evaluating them in terms of suitability to deliver previously mentioned aims of LIB recycling processes. LIBs are commercially recycled around the world by combining technologies which can be generally characterized as physical based technologies for pre-processing and materials separation, or chemical-based technologies for materials extraction and recovery.

Physical based technologies can be sub-categorized as mechanical pre-treatment, and gravity, magnetic and electrostatic based materials separations.

Chemical based technologies can be subdivided further and classified by type as hydrometallurgical processes including leaching, solvent extraction, and precipitation and electrolysis-based recovery; pyrometallurgical processes (battery smelting predominantly); or bio-hydrometallurgical processes in which extraction is conducted by microorganisms (Figure 39).

*Figure 39: Li-ion battery recycling process classification (Swain, 2017a).*



Most processes employed commercially today will be hybrid processes utilise pyro- and/or hydrometallurgy as a means of extracting and refining the valuable components, although mechanical methods of pre-treatment are used for separation of metals, plastics etc., and recovery of materials from solution is conducted via numerous methods including precipitation, electrolysis, solvent extraction and complexation. In general, commercial processes, despite being hybrids of numerous technology types, can be classified according to their use of smelting as pyrometallurgical, or as hydrometallurgical when smelting is avoided.

The commercialised LIB battery recycling processes in terms of their suitability for recovery of TMs from waste LIBs, and any advantageous or disadvantageous features for long term sustainable LIB treatment and their relative pros and cons is given in Tables 34 and 35. All of these are at TRL 9. This is followed by a brief discussion of challenges for future LIB recycling, and pipeline developments which lend themselves to sustainable TM recovery from LIBs into the future, avoiding the pitfalls and other issues encountered with existing commercial options.

*Table 31: General comparison of pyro- and hydrometallurgical recovery processes for LIB recycling and TM/CRM Recovery.*

| Process | Pros | Cons | Comments on recovered materials |
|---|---|---|---|
| Pyrometallurgy (smelting). | Meticulous sorting of batteries by chemistry, disassembly of battery packs, and comminution with subsequent physical separation is unnecessary, avoiding these costs and exposure to health and safety issues associated with these processes.<br>Potential to generate added-value pre-cursor compounds for LIB manufacturing when combined with hydrometallurgical refinement (e.g., Umicore).<br>Alternative to higher environmental impact incineration of batteries.<br>Consumption of graphite and organic portions of batteries offsets demand for coke (coking coal itself is now critical) and other fossil fuel based reducing agents.<br>Can combine treatment with existing smelting operations when advance battery recycling is unavailable.<br>Potential to combine processing of wastes when insufficient quantities of waste LIBs are available for processing to justify capital costs of new plant, or long-distance logistics costs to deliver LIBs to advanced recycling are prohibitively high.<br>Generation of lower volumes of hazardous wastes requiring further treatment than hydrometallurgy. | Direct charging of batteries to smelting results in loss of organics, electrolytes and critical graphite which convert to CO2/VOCs and impacts global warming potential (GWP).<br>Li partitions to slag from which recovery is generally not possible due to thermodynamic/cost barriers.<br>Requires large capital investment and throughput to maintain.<br>Generally unsuitable for smaller scale decentralized (local) treatment with existing plants consuming LIBs from many countries.<br>Impacts and costs of long-distance logistics to deliver batteries to process are high in comparison to decentralized processing.<br>High energy consumption and emissions generation.<br>Gas clean-up required to avoid toxic emissions.<br>Processes generally only economical for batteries with high value cathode metals (Co, Mn and Ni), long term viability in light of innovation in delivering lower cost cathode materials is questionable. | Recovery focused on capture of valuable cathode metals and copper current collectors at the cost of other cell components.<br>Recovered materials:<br>Cathode metals (Co, N, Mn) and<br>Cu current collectors<br>Unrecovered materials:<br>All organic portions including electrolytes and polyolefin membranes are burned off.<br>Graphite of anodes is also burned off.<br>Li and Al segregate to slag rendering recovery uneconomical (although examples of Li recovery exist e.g., Accurec) |
| Hydrometallurgy. | Capable of recovering greater range of materials than pyrometallurgy therefore offers opportunities to valorise batteries to a greater extent (e.g., Lithorec process which recovers electrolytes suitable for reuse).<br>Recovery of materials as added-value raw materials for battery manufacturing is common – greater value generation potential and improved environmental impact of close-loop battery lifecycles.<br>Fewer GHG emissions than pyrometallurgical processes. | Intensive battery sorting by chemistry prior to treatment may be necessary (labour/cost-intensive).<br>Environmental health and safety issues during pre-treatment of LIBs (although mitigation measures such as short circuit discharging, comminution under inert atmospheres, and cryotreatment of cells are commercially adopted).<br>Thermal drying of recovered materials is necessary adding to costs, energy demand, and emissions (although use of low boiling point solvents which are captured and recycled mitigates this). | Recovery of a greater range of materials present is possible, problems of slagging metals or combustion of organics and graphite anodes are avoided.<br>Recovered Materials:<br>Copper, aluminium<br>Li2CO3<br>Cobalt and other transition metals from cathodes |

| Process | Pros | Cons | Comments on recovered materials |
|---|---|---|---|
| | | Use of organic solvents requires consideration, some are hazardous e.g. -acetonitrile, NMP and risk mitigation measure may be necessary.<br>Careful use of protic solvents, if present in recovered electrode materials and electrolytes, may be unsuitable for reuse in LIBs due to risk of reaction with Li metal.<br>Large volumes of liquid wastes may result from processes which can be hazardous and costly to process. | Unrecovered materials:<br>The nature of the process employed may or may not result in recovery of additional materials present including graphite, electrolytes and salts, polymeric materials. |

*Table 32: Summary of evaluated commercial Li-ion battery (LIB) recycling processes.*

| Company | Battery types and target TMs/CRMs | Process | Pros | Cons | References |
|---|---|---|---|---|---|
| Umicore. | LIB (numerous chemistries) & NiMH<br>Co<br>REMs | Pyrometallurgy + hydrometallurgical refinement.<br>Directly charge batteries to smelting.<br>Recovery of Co-Ni alloy.<br>Hydrometallurgical treatment of alloy to recover Mn salts, $NiSO_4$ and $CoCl_2$, $CoO_2$ and $Ni(OH)_2$.<br>Further refinement recovers REMs as oxides. | Direct charging of batteries avoids exposure to hazards during pre-treatment.<br>Simultaneous treatment of numerous battery chemistries.<br>Closed-loop strategy with materials recovered as added-value raw materials for battery manufacturing. | Large capital investment of such processes.<br>Critical Li and graphite are lost.<br>Economics based on recovery of cathode metals which are of diminishing value in newer battery chemistries.<br>Combustion of organics results in GHG emissions and loss of potential value.<br>Only feasible at large scale. | (Buchert, M. Manhart, A., Bleher, D., Pingel, 2012; Elwert et al., 2015) |
| Xstrata. | LIB<br>Co | Pyrometallurgy + hydrometallurgy<br>LIBs charged to furnace with nickel ore for smelting<br>Process recovers Cu, Ni and Co. | Takes advantage of established process for primary production negating capital investment for new recycling process.<br>Scale of process reduces recycling costs.<br>Avoids pre-processing. | Only CRM recovered is Co from cathodes.<br>All other TMs burned off or slagged.<br>Combustion of organics results in GHG emissions and loss of potential value.<br>Further processing of recovered Co required to create raw materials for LIB manufacturing. | (Tollinsky, 2008; X. Zhang et al., 2013; Heelan et al., 2016; Swain, 2017a) |

| Company | Battery types and target TMs/CRMs | Process | Pros | Cons | References |
|---|---|---|---|---|---|
| | | | | Quantity of Co in LIBs in general is reducing resulting in questionable economics over the long term. | |
| Inmetco. | NiCd, NiFe, NiMH, Li ion and Hg-free Zn–Mn batteries.<br>Co. | Combined pyro- + hydrometallurgy:<br>Batteries directly charged to EAF with Fe-Cr pellets from pre-treated stainless steel EAF dust.<br>Co, Cu and Ni recovered in Fe alloy.<br>Further refinement enables recovery of individual metals | Capitalises on established infrastructure (in the UK Celsa Steel in Cardiff operate EAF steel and are currently pursuing resource efficiency options).<br>Dual processing of wastes.<br>Avoids pre-treatment of cells.<br>Lower capital investment than traditional smelting. | Only Co, Cu and Ni recovered, all other components are slagged or consumed as reducing agents.<br>Can only tolerate limited charge of batteries.<br>Quantity of Co in LIBs in general is reducing resulting in questionable economics over the long term.<br>Emissions associated with electricity generation for EAF and from combustion of organic cell components. | (Bernardes, Espinosa and Tenório, 2004; Espinosa, Bernardes and Tenório, 2004; Al-Thyabat et al., 2013; X. Zhang et al., 2013). |
| Accurec | LIB (portable batteries).<br>Co, Li. | Pyrometallurgy + hydrometallurgy.<br>Physical pre-treatment recovers WEEE and plastics from battery packs.<br>Thermal vacuum treatment recovers electrolyte.<br>Physical separation of crushed materials recovery Fe-Ni, Al, Cu and plastics from cells.<br>Smelting of fines produces Co-Mn alloy.<br>Hydrometallurgical treatment recovers $Li_2CO_3$ from slag. | Recovers Co from cathodes (and Mn, although other cathode metals may be lost in slag), and all materials present except for graphite.<br>EAF processes requires lower capital investment than other large-scale smelting processes.<br>Li is recovered from electrolyte and from slag after smelting.<br>Slag treatment may be suitable for Li bearing slags from other recycling operations.<br>Greater proportion of value recouped from batteries than other smelting approaches<br>Capital investment for EAF is lower than large scale smelting e.g., Umicore. | Graphite is lost in thermal treatment resulting in GHG emission.<br>Less valuable cathode metals than Co and Mn are lost.<br>Co and Mn recovered as an alloy requiring further refinement, lower value material than raw materials for manufacturing produced by e.g., Umicore.<br>Higher capital costs than alternative hydrometallurgical approaches. | (Georgi-Maschler et al., 2012a; Meshram, Pandey and Mankhand, 2014a) |
| Retriev Technologies (formerly Toxco). | All Li battery chemistries<br>Co (and other cathode metals), Li | Hydrometallurgy:<br>Cells discharged prior to crushing under liquid solution. | Recovers a greater range of materials from batteries than pyro processes in general inc. Li. | Fate of electrolyte solvents and graphite is uncertain.<br>Pre-treatment via comminution is necessary. | (Lain, 1999a; Retriev Technologies, |

| Company | Battery types and target TMs/CRMs | Process | Pros | Cons | References |
|---|---|---|---|---|---|
| | | Crushed cells reacted with water at pH≥10.<br>Plastic and Cu, Al, Co are filtered from solution, metal components suitable for reuse in new batteries.<br>Lithium salts precipitated from solution and dissolved in $H_2SO_4$.<br>Solution is treated in electrolysis cell to yield LiOH, which is further reacted with $CO_2$ to yield LiCO3. | Cryogenic pre-treatment reduces risks associated with Li metal in pre-processing.<br>Recovery materials in a form suitable for use in other products<br>Flexibility in the way that Li is recovered to create raw materials for different markets.<br>Direct recovery of Li as $LiCO_3$ represents a more closed-loop approach for LIBs.<br>Environmental and economic advantages vs. energy intensive pyro processes that recover metal products rather than raw materials for manufacturing.<br>Lower capital investment than pyro, sustainable at lower throughput.<br>More resilient to reduction in economic value of cathode components in LIBs.<br>Utilises LiOH product to moderate process pH negating purchase of alternative reagents to maintain pH during materials dissolution. | Consumes reagents such as $H_2SO_4$ and liquid nitrogen for cryo-treatment.<br>Use of strong corrosive mineral acids for digestion requires expensive acid proof plant.<br>De-watering and thermal drying of products necessary | 2017a; Swain, 2017b) |
| AEA technologies<br>(process no longer in operation) | LIBs – $LiCoO_2$<br>Co, graphite, Li (from electrolytes and cathodes) | Hydrometallurgy<br>Opening of cells under $N_2$ atmosphere.<br>Dissolution of electrolytes in acetonitrile under $N_2$ atmosphere and recovery following evaporation.<br>Dissolution of binder materials in MNP enables Al, Cu and plastics to be separated. | Far greater range of cell components recovered than pyro in general – all materials recovered.<br>Potential recovery of PVDF electrode material binders through solvent extraction.<br>Critical graphite is recovered.<br>All electrolyte components are recovered in a reusable state (added value). | Inert atmosphere must be maintained during pre-treatment of LIBs.<br>Pre-sorting of batteries to determine which are $LiCoO_2$ based is required – process is specific for single LIB chemistry.<br>Washing and drying of products necessary.<br>Process uses hazardous organic solvents<br>. | (Lain, 1999a, 2001a; Meshram, Pandey and Mankhand, 2014a) |

| **Company** | **Battery types and target TMs/CRMs** | **Process** | **Pros** | **Cons** | **References** |
|---|---|---|---|---|---|
| | | LiCoO2 and carbon particulates are filtered and treated in an electrolytic cell with aqueous LiOH solution to recover Co and carbon. | Process could be easily adapted for treatment of mixed chemistries with treatment of extracted mixed cathode materials elsewhere. | | |
| Recupyl process (France pilot, implemented in Singapore) a.k.a. the Valibat process | All Li battery chemistries including primary Li, all Li-ion and LiPO4 chemistries<br>Co, graphite, Li | Hydrometallurgy<br>Mechanical pre-treatment: 2 stage grinding under inert atmosphere (20: 80 Ar/$CO_2$).<br>Physical separation to recover steel, copper and plastics.<br>Li leached from fines and precipitated as $Li_2CO_3$ or LiPO4.<br>Cathodic mixed oxides, carbon and intercalated Li leached with sulfuric acid.<br>Co(III) hydroxide is precipitated by oxidation with NaClO.<br>Li as aqueous $Li_2SO_4$ is precipitated as $Li_2CO_3$ by reaction with $CO_2$. | Room temperature process.<br>Recovery of cell components as added-value raw materials for battery manufacturing.<br>Automated pre-treatment reducing risks to workers.<br>Tolerates all LIB chemistries.<br>Flexible process.<br>Process iterations can recover valuable PF6 anions of electrolyte salts and $LiPO_4$. | Requires inert atmosphere for comminution and hydrolysis stages<br>Mitigation of risks associated with hydrogen evolution in hydrolysis.<br>Consumes reagents | (Meshram, Pandey and Mankhand, 2014a) |

## Pre-treatment

Due to the safety issues associated with LIBs, special consideration is paid here to methods of pre-treatment prior to further processing. Numerous methods are employed industrially to deal with these issues and are considered here. Discharge of cells is necessary prior to processing in most cases. This can be achieved through submersion in salt water or through use of ohmic resistors (Lain, 1999a), although corrosion issues occur at battery terminals while submerged in brine. OnTo technologies have patented a device to discharge battery cells which connect battery electrodes via an ohmic resistor coupled with a thermochromic material which uses colour change to indicate when current has ceased to flow, and batteries are fully discharged.

The practicality of using such a device on an industrial scale for discharging batteries prior to recycling is questionable due to the large numbers of batteries which require processing and the associated time and manpower required for such an undertaking. Wet crushing has significant safety issues relating to the generation of hydrogen gas during comminution as Li metal hydrolyses on contact with water or other protic solvents. Crushing under a $CO_2$ atmosphere passivates Li metal through formation of lithium carbonate on its surface slowing the rate of further reaction. Inert atmospheres for crushing such as $CO_2$, $N_2$ or Ar drastically reduce exposure to oxygen and water vapour in the air, preventing hydrolysis of lithium. Cryogenic cooling of cells is also employed prior to comminution, as drastic reduction in temperature reduces the reactivity of lithium, enabling safer processing (McLaughlin and Adams, 1999; T. Zhang *et al.*, 2013).

More recently, the use of supercritical $CO_2$ ($ssCO_2$) and subcritical $CO_2$ in LIB pre-treatment to extract electrolytes, electrolyte|electrode interface materials and degradation products formed during battery use have begun to emerge. These hold advantages over current pre-treatment methods and are discussed below in the 'Future LIB Recycling Processes' section of this appendix.

## Pyrometallurgical LIB recovery processes

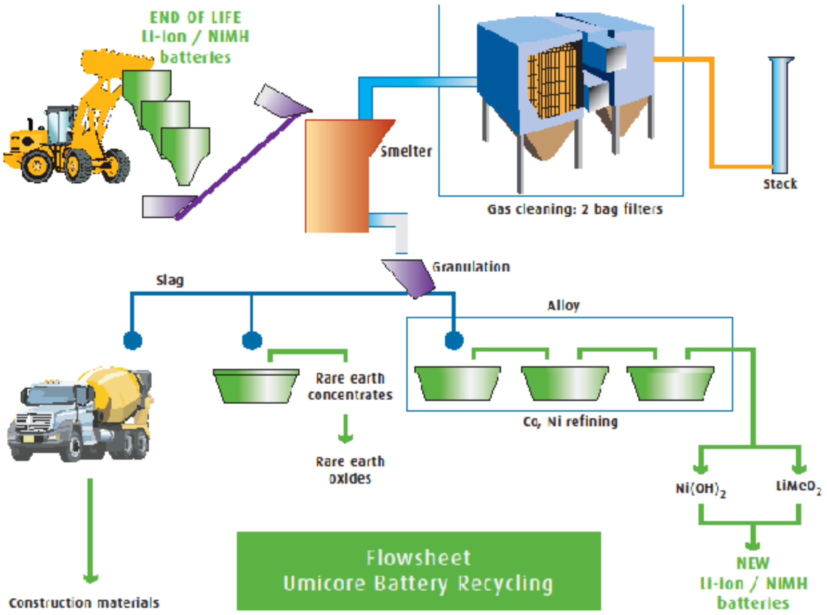
Pyrometallurgical processes involve smelting of LIBs to recover the valuable metallic components as an alloy. This alloy then requires further refinement, usually in a hydrometallurgical process to obtain pure metal secondary resources or materials suitable for direct reuse in LIB manufacturing. Often pyrometallurgical recycling of LIBs is conducted by inclusion in synergistic smelting operations the primary function of which is to process ores or other materials. This is likely to become increasingly unnecessary as available quantities of EoL LIBs grow.

### Umicore

Umicore operate the Val'eas process: a combined pyro-and-hydro-metallurgical process involving smelting of batteries with subsequent electrolytic refinement of resulting nickel-cobalt alloy. This process is designed to accept mixed charges of various types of LIBs and NiMH batteries without any pre-treatment. This avoids pre-processing costs and potential exposure to hazardous materials during pre-processing and costs as, an advantage in comparison to hydrometallurgical processes. Once charged to a purpose-built shaft furnace, plastics, solvents and graphite act as reducing agents during the smelting process and are converted to $CO_2$. Smelting reduces the target metals Cu, Co, Ni, Mn and Fe which are collected in an alloy. The cooled solid metal alloy is then sent from the smelting site in Sweden for hydrometallurgical refinement in Belgium by digestion in sulfuric acid ($H_2SO_4$).

Solvent extraction of Mn salts, $NiSO_4$ and $CoCl_2$ from leach solution is conducted before $LiCoO_2$ and $Ni(OH)_2$ are electrolytically recovered and sold for use in manufacturing new batteries. In this way the process is 'closed-loop'. However, lithium and other ignoble metals are not recovered but partition into slag during smelting. The slag has been specially formulated to suit applications in the construction sector and in this way the material can be valorised (Meskers, Hagelüken and Van Damme, 2009; Meshram, Pandey and Mankhand, 2014a) although downcycled and locked up forever where this slag finds application in the construction sector. The derived value from these metals will be considerably less than if recovered in a usable form for manufacturing and lack of recovery does nothing to mitigate the impacts of primary lithium and aluminium production for manufacturing new LIBs. Figure 40 shows the Umicore battery recycling process.

*Figure 40: Flowsheet of Umicore Battery Recycling Process (Elwert* et al.*, 2015).*



The ability to recycle different battery chemistries at once is an advantage of this process, providing an EoL solutions for various chemistries in a single process unlocking OPEX advantages through economies of scale with larger quantities of materials processed simultaneously. However, the CAPEX for such a large-scale pyro process creates significant pressure to continue to acquire sufficiently large quantities of batteries to repay this investment over the lifetime of the plant, and so combined treatment of batteries of differing chemistries is necessary to sustain the operation. REMs are recovered in this process from NiMH batteries, although the available volumes of NiMH in the future will diminish as they lose market share to LIBs. Additional disadvantages of this process include loss of materials such as carbon, plastics and lithium which can be reclaimed in numerous other LIB recycling operations.

Achieving efficient recycling relies upon the ability to valorise LIB materials in order that a positive cost-benefit to materials recovery can be derived. The economic driving force for LIB recycling via pyrometallurgical processes such as Umicore’s has been recovery of cathodic materials, the most valuable component, at the expense of other cell components in pyrometallurgical operations.

The cost of these materials is determined by market values of individual metals used in cathodic compounds, and the cost of producing these compounds (Nitta *et al.*, 2015). It has been shown these compounds can be created at lower cost through recovery from LIBs than from primary sources (Steward, Mayyas and Mann, 2019), and this strategy of direct production of ‘added-value’ materials for LIB production improves the cost benefit of LIB recycling and so maximum recovery of cathode metals has been the primary goal while sacrificing other critical cell components e.g., Li in electrolytes and cathodes, and graphite in anodes.

Consumption of organic materials including electrolytes, binders and polymers in smelting through combustion or their use as a reducing agent and slagging of lithium and aluminium means that as the proportion of recoverable value present as copper foils and cathode metals decrease, the importance of valorising all cell components for viable recycling will increase. The urgency of action to tackle the climate emergency also casts doubt on the long-term sustainability of any process which converts potential secondary raw materials to GHG emissions. As such, the long-term economic and environmental performance on this process and similar smelting operations is questionable.

## Xstrata

Xstrata operate a process which combines pyrometallurgical and hydrometallurgical processing steps. This process was not designed for LIB recycling, but for processing nickel ores. Cobalt, nickel and copper content of LIBs are recovered, with all other battery materials being 'slagged' or used as an energy source in pyrometallurgical treatment. In this way, the process is similar to Umicore's with potentially recoverable materials lost (Tollinsky, 2008; X. Zhang *et al.*, 2013; Heelan *et al.*, 2016; Swain, 2017a).

The economics of recovery and the long-term viability of inclusion of LIBs in this process for recovery are perhaps more questionable with copper and cathode metals recovered as pure metals rather than added-value raw materials for LIB manufacturing. There is also the question of whether future battery chemistries will be compatible with the primary ore smelting operation, and whether the losses of potentially recoverable materials, particularly those converted to $CO_2$ can be sustained in view of the climate emergency.

## Inmetco

In the USA, International Metal Reclamation Company (INMETCO), a subsidiary of the International Nickel Company (INCO), recycles batteries with a pyrometallurgical process. The process was developed for recovery of Fe, Zn and Pb from electric arc furnace (EAF) dusts from the stainless-steel industry. However, the process is also applied to recover metals from other Fe bearing wastes including NiCd, NiFe, NiMH, Li ion and Zn–Mn batteries free of Hg, which are charged as secondary materials to the process.

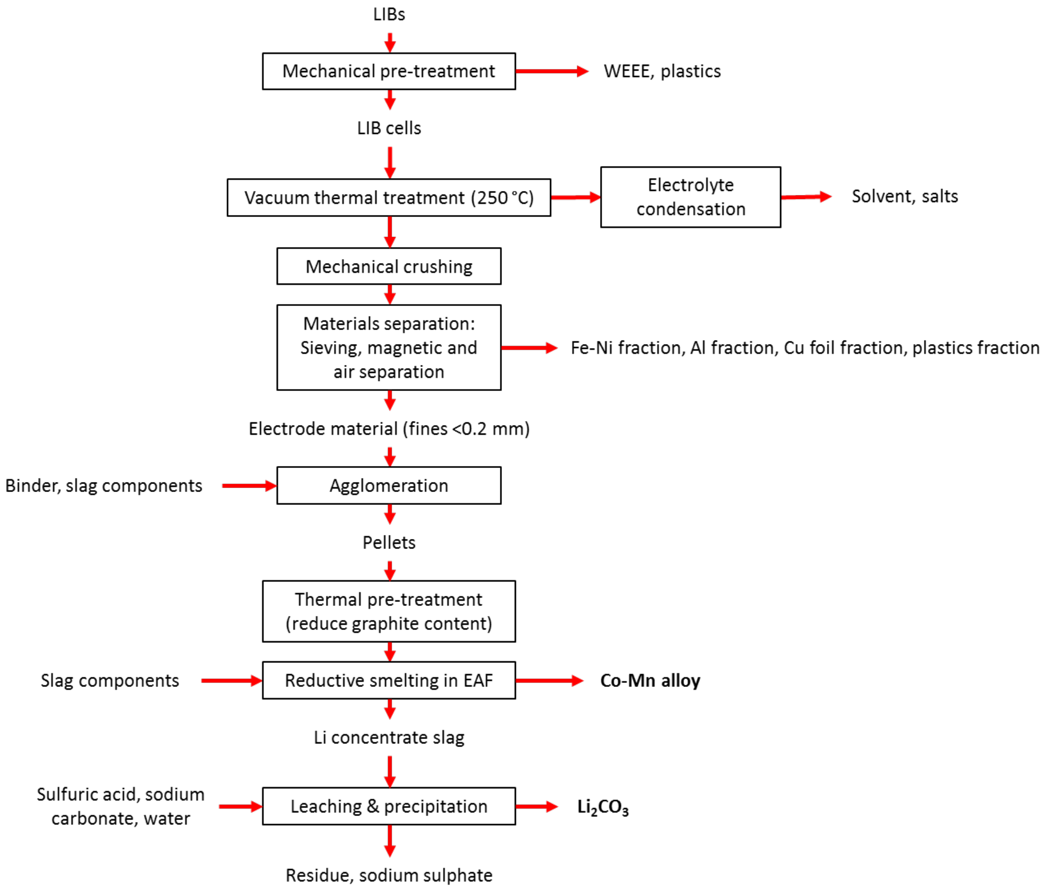
EAF dust is agglomerated into self-reducing pellets with a carbon-based reducing agent, which are then treated for 15 mins at up to 1350°C causing volatile metals such as Zn and Pb to be driven off and collected in a gas treatment system, and oxides are reduced to give Fe-Cr pellets which are then charged with other Fe bearing wastes, including a small proportion of LIBs to an EAF. The aim being recovery of Co, Ni and Cu in an Fe-based alloy. Ignoble metals are slagged and organic materials as well as carbon are burned and used as reducing agents. In the Inmetco process, most of the battery components including the lithium are lost as is typical in pyrometallurgical processes discussed so far (Bernardes, Espinosa and Tenório, 2004; Espinosa, Bernardes and Tenório, 2004; Al-Thyabat *et al.*, 2013; X. Zhang *et al.*, 2013).

## Accurec GmbH

Accurec have developed a process for portable LIB recycling (Figure 41). This process uses pyrometallurgy in combination with hydrometallurgical refinement to recovery Co and Mn from cathode materials as an alloy. However, unlike other pyrometallurgical LIB recycling processes, the process aims to recover other components of LIBs including Li, which is recovered as LiCl salt. Carbo-reductive smelting of LIB fine fractions containing Co and Li electrode materials is conducted in an EAF. Outputs from the EAF process include a cobalt alloy and lithium concentrates which are further treated hydrometallurgical to yield pure lithium carbonate (a precursor for LIB manufacturing), showing an advantage over alternative pyrometallurgical approaches in the ability to valorise lithium through recovery as an added value raw material for LIB manufacturing.

Electronics and plastic casings of battery packs are removed leaving individual LIB cells for further processing, and a WEEE and plastic fraction which can be sold on for recycling. Cells are treated by vacuum thermal treatment at 250 °C which removes electrolyte (solvents and conductive salts) and pyrolyzes cells allowing electrolyte to be captured by condensation. Although recovered, electrolyte cannot be reused in new cells due to contamination with degradation products from the thermal treatment. Safely deactivated cells are then mechanically crushed, and the resulting material undergoes physical separation via sieving, magnetic separation and air classification to yield aluminium, iron-nickel, copper and plastic fractions. The remaining fine fraction (<0.2 mm) contains the valuable cathodic materials of LIBs with Co and Li present as oxides in combination with graphite. This fraction is agglomerated into pellets using a binding agent (molasses), which undergo a thermal pre-treatment in a rotary kiln to reduce graphite content, before charging to an EAF in which they are smelted to obtain Co-Mn alloy and a lithium containing slag. The slag undergoes hydrometallurgical leaching to recover lithium as $Li_2CO_3$.

*Figure 41: Accurec GmbH LIB recycling process (Georgi-Maschler* et al.*, 2012a; Meshram, Pandey and Mankhand, 2014a).*



Just as in other pyro-processes, the critical graphite component of the cells is lost. However, despite being unsuitable for direct reuse in batteries, the electrolyte materials and lithium components are recovered in addition to the valuable metals from cathodes. The leaching of lithium from the slag to recover lithium carbonate represents an additional phase of recovery which is not performed on slags from other pyrometallurgical LIB recycling processes. This element could be extremely important in the future to mitigate lithium loss in other pyrometallurgical LIB recycling processes and reduce primary lithium demand through provision of secondary lithium compounds at lower economic and environmental cost than through primary production.

## Pyrometallurgical LIB Recycling Overview

One major advantage of pyrometallurgical treatment is that meticulous sorting of batteries per their chemistries, disassembly of battery packs, and grinding with subsequent physical separation is not necessary to recover the most valuable metals of cells. This negates the cost and safety issues associated with these processes which are necessary when hydrometallurgical processing is employed. These processes also offer a means to generate added-value pre-cursor compounds for

LIB manufacturing e.g., Umicore's process, if appropriate hydrometallurgical treatment of resulting alloys from smelting is employed. In this way closed-loop battery recycling can be achieved.

When closed-loop recovery is not employed, valuable secondary resources can still be generated whilst providing a means of diverting waste LIBs from landfill or higher environmental impact incineration processes. Utilisation of organic components of cells as reducing agents in smelting also offsets demand for coke and other fossil fuel based reducing agents and energy for the process. However, as exemplified by the Accurec process, a dedicated process for LIB recycling which embraces appropriate pre-treatment with subsequent leaching and recovery of Li from slags can increase the range of materials which are recovered from LIBs considerably in comparison to other pyrometallurgical processes thereby mitigating materials criticality issues.

The prospect also exists to integrate LIB recycling into existing pyro processes such as that used by Inotec when advanced battery processes are unavailable. Additionally, this presents an opportunity to process LIBs along with other wastes when sufficiently large quantities of LIBs alone cannot be accumulated to justify investment in a new process to and the long-distance logistics costs to deliver batteries to advanced recycling processes elsewhere in the world. Pyro-processes on the whole result in generation of far lower volumes of hazardous wastes which require further treatment than purely hydrometallurgical processes. However, direct charging of LIBs to smelting processes means that potentially valuable materials such as electrolytes and plastics from batteries are lost rather than recovered, eventually being burned off.

The resulting direct GHG emissions may be avoided through use of alternative processes which seek to capture rather than consume these materials. Generation of additional toxic gaseous emissions must also be tackled through use of costly abatement plants. Critical lithium and other valuable metals are lost as they partition to slag during smelting, rendering them thermodynamically difficult and therefore prohibitively expensive to recover (although this problem has been addressed by Inotec who recover Li from slag). In addition, far larger capital costs are involved with building pyrometallurgical processes which require large throughput to sustain. This means that pyro processes are generally unsuitable for smaller scale local treatment. The economic and environmental costs of transporting large quantities of end-of-life LIBs from across large geographical areas to recycling processes are considerable compared to those that may be achievable with smaller scale localised treatment of batteries in hydrometallurgical processes.

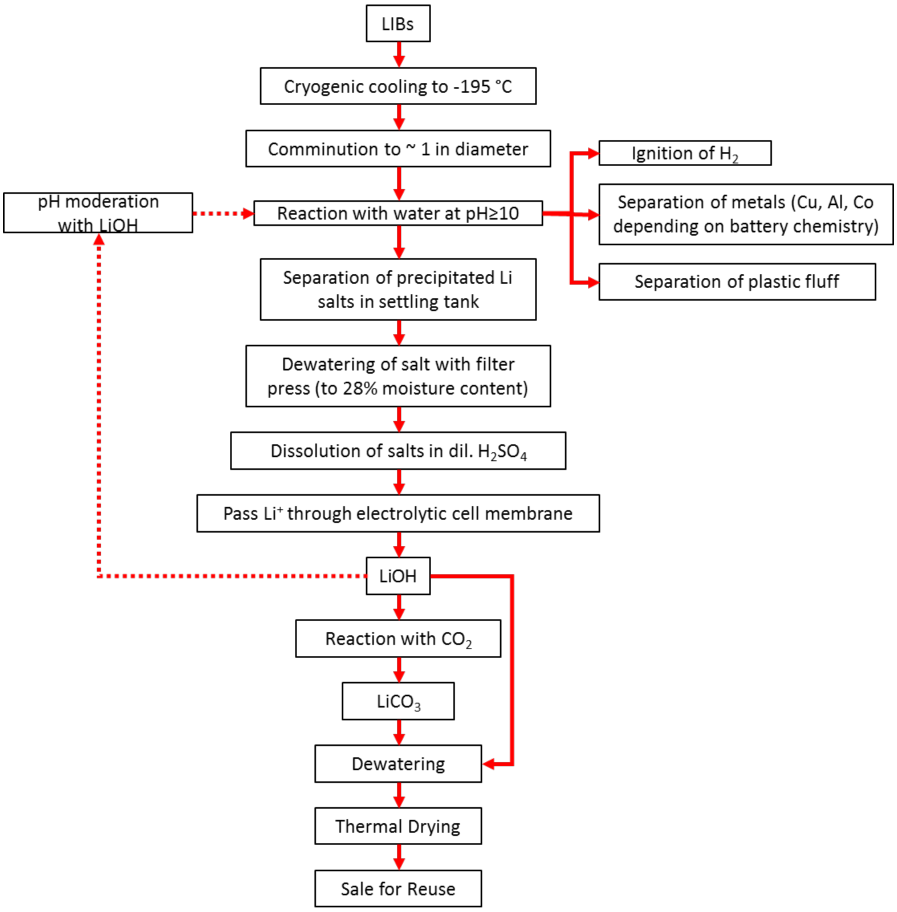
## Hydrometallurgical LIB Recovery Processes

Hydrometallurgical processes are solution based, involving the digestion or dissolution of LIB materials for subsequent separation and recovery, usually by precipitation or electrolysis. This section outlines currently commercialised hydrometallurgical LIB recycling processes, evaluating their advantages and disadvantages in terms of long-term sustainable CRM recovery from waste LIBs.

### Retriev technologies, Canada (formerly Toxco)

This hydrometallurgical process (Figure 42) can accommodate all lithium batteries (inc. Li metal primary cells). Following discharge, cells are cooled in liquid nitrogen to -195°C thereby reducing the reactivity of Li metal by 5-6 orders of magnitude (McLaughlin, 1994). This reduces the risk of explosion due to oxidation of Li present in cells during comminution to a uniform size of 1 inch which enables dissolution of Li in water as a variety of salts dependent upon battery chemistry. A pH ≥10 is maintained by addition of LiOH to prevent formation of toxic $H_2S$. Generated hydrogen is burned off as $H_2O$ and remaining solid metal cell components and plastic fluff are removed. Recovered metals Cu, Al, and Co (depending on LIB chemistry) are suitable for reuse in new products. As Li continues to oxidize and dissolve, the solution becomes supersaturated with Li salts begin to precipitate. These are collected, dewatered and dissolved in dilute sulfuric acid within an electrolytic cell. $Li^+$ ions migrate through a membrane to the basic side of the cell where they precipitate as LiOH. Some of this is used to moderate the pH in the earlier dissolution process, some is dried and sold on for reuse in new products, and the rest is further reacted with $CO_2$ to produce $LiCO_3$ which can used to manufacture new LIBs (Lain, 1999a; Retriev Technologies, 2017b; Swain, 2017a).

*Figure 42: Retriev Technologies (formerly Toxco) process for Li recovery from LIBs.*

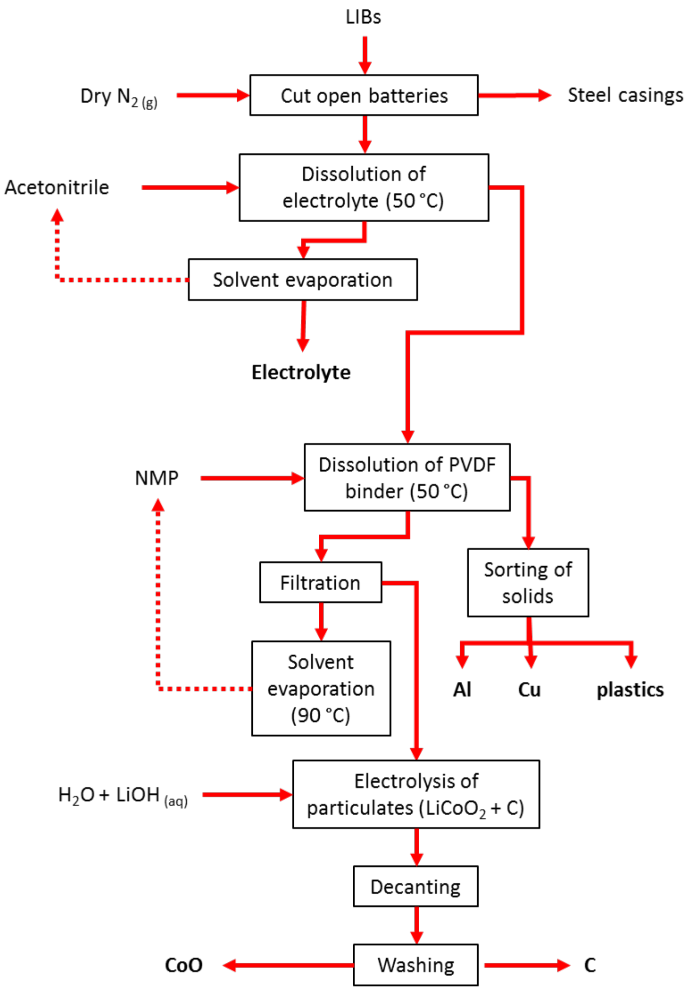


This process holds advantages over pyrometallurgy processes. In general recovery of more of the materials present in LIBs is possible. Direct recovery of electrode materials in a form suitable for re-use in new LIBs represents a more closed-loop recycling strategy which will have greater associated economic and environmental benefits than those pyro-processes that utilise large amounts of energy and produce only secondary metal products rather than pre-cursor compounds for manufacturing. Efficient recovery of lithium as lithium carbonate is also an advantage over pyro processes in general, with the Accurec process as the only pyro example which recovers Li once slagged in smelting.

## AEA technologies

AEA technologies operated a LIB recycling process in Scotland in which the first stage of recycling was to cut open cell casings and remove the internal spiral wound cell components (foils, electrodes and membrane) in a dry inert $N_2$ atmosphere (Figure 43). Cell components are then transported in a mesh basket within a sealed container containing a dry $N_2$ atmosphere to a vessel which also contains dry $N_2$ atmosphere in which acetonitrile solvent at 50°C dissolves the electrolyte and its solvent. Acetonitrile is then removed and evaporated at reduced pressure to leave the electrolyte and its solvent in a suitable form for reuse. Binder materials are dissolved by MNP at 50°C, and the resulting suspension is filtered before evaporation at reduced pressure. This leaves plastic, Al and Cu solids for collection and separation. Filtered particulate material ($LiCoO_2$ + C) is transferred into an electrolysis cell containing $LiOH_{(aq)}$ electrolyte. $LiCoO_2$ is reduced at the cathode to cobalt (II) oxide and LiOH solution is then decanted from the cell leaving cobalt oxide and carbon which is washed and stored (Lain, 1999a 2001a; Meshram, Pandey and Mankhand, 2014a).

*Figure 43 AEA-technologies LIB recycling process (Lain, 1999a)*



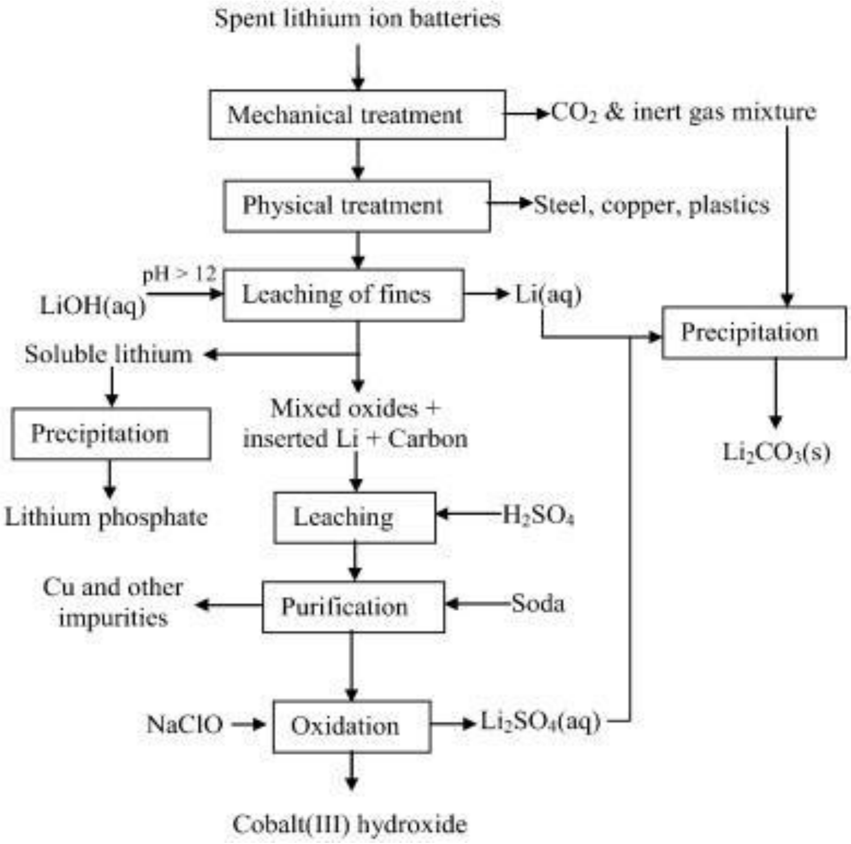
Issues with this process include safety related to the evolution of hydrogen. The use of acetonitrile also introduces health and safety implications for recycling workers and concerns over environmental emissions. However, once again this hydro process holds advantages over pyro processes in terms of the range of materials that can be recovered from LIBs. This process achieves closed-loop recovery of electrolytes (Li salts and electrolyte) in a state suitable for reuse in new batteries, unlike electrolytes recovered in the Retriev Technologies process. This is advantageous enabling greater valorisation of waste LIBs, as electrolytes are the most valuable materials in LIBs after cathode materials. All components of LIBs are recovered including critical graphite of anodes.

However, since the introduction of this process, LIB chemistries have altered, and various cathode materials containing different amounts of Co, Mn and Ni form part of the waste stream. This suggests that for this process to be employed, intensive sorting of batteries by cathode chemistry is necessary to ensure only LIBs containing $LiCoO_2$ cathodes are processed. This highlights a disadvantage of the process, that only a single LIB chemistry can be accommodated. The process could however easily be adapted to recover combined cathode materials from mixed LIB chemistries for processing elsewhere. To the best of the Authors' knowledge, this process is no longer in operation.

## Recupyl process (France) a.k.a. the Valibat process

This is a room temperature hydrometallurgical process designed to treat all types of Li based batteries (primary and secondary including Li-ion and $LiPO_4$ chemistries) (Figure 44). The process recovers metallic casings, electrode contacts, cathode metal oxides and lithium salts from LIBs. The process also aims to recover materials as added-value compounds, suitable for use in manufacturing new products.

*Figure 44: Recupyl LIB recycling process (Meshram, Pandey and Mankhand, 2014a).*



Mechanical grinding of LIB cells is carried out under inert atmosphere (20% Ar/80% $CO_2$). This is conducted in 2 steps with mills arranged in series: the first uses a rotary sheer machine at low rpm (>11 rpm) to release the internal stresses of cells; the second processes the shredded material in a rotor system at no more than 90 rpm. Batteries and cells are continuously fed to the grinding process via a double airlock. The crushed material is then separated on a vibrating screen with a mesh size ≤3 mm and subsequent magnetic and density separation to recover stainless steel, a non-ferrous metal fraction (Cu), and a plastic fraction. Fines rich in metal oxides and carbon are then suspended and stirred in water for hydrolysis and leaching. The turbulence causes metals to be released and Li is hydrolysed with the solution reaching pH>12.

The rate of hydrolysis is controlled by the charge rate of fines to the solution, and turbulence above the solution reduces the risk of evolved hydrogen exploding. An air atmosphere containing >0.5 vol% of $O_2$ is also maintained to limit the risk of explosion. Upon completion of the hydrolysis reaction, filtration yields a solution of lithium salts and a solid fraction of carbon and metal oxides which are separated. Lithium is precipitated as $Li_2CO_3$ using $CO_2$ from mechanical pre-treatment. Mixed metal oxides including their lithium component and cobalt are then leached with sulfuric acid. Carbon is filtered from the resulting solution, prior to cementation of copper by addition of steel shots. Purified solution is then oxidized with sodium hypochlorite to yield cobalt (III) hydroxide as a precipitate, leaving a solution of lithium sulphate which is sent back for precipitation of lithium as $Li_2CO_3$ by reaction with $CO_2$ (Meshram, Pandey and Mankhand, 2014a).

Additional process iterations indicated in the patent for this process include recovery of Li as $Li_3PO_4$ from solution by pH adjustment with phosphoric acid in place of recovery of $Li_2CO_3$; and electrolysis of purified sulphuric acid leach solutions to recover cobalt alloy cathodes. This adds some amount of flexibility to the recovery process and enables users to recover materials in the most appropriate form to suit demand (Tedjar and Foudraz, 2010). The additional opportunity to recover $PF_6$ anions from LIB electrolytes following leaching of shredded battery materials in water also exists. Precipitation of these anions by complexation with appropriate large cationic species (e.g., alkyl ammonium species) is possible, enabling the valuable electrolyte salts to be recovered.

### Hydrometallurgical LIB Recycling Process Overview

In general, hydrometallurgical recovery processes can recover a greater range of materials from LIBs than is achieved with pyro processes, with a greater proportion of the masses of LIBs recovered. This presents opportunities to valorise more of the material content including valuable electrolytes which can be recovered in a form suitable for reuse in new batteries. This is exemplified by the AEA Technologies process. In general, dissipation and loss of LIB components beyond recovery, an inherent problem of pyro processes, is avoided.

Recovery of added-value pre-cursor materials for battery manufacturing is common in hydrometallurgical recovery which enhances the environmental and economic benefits resulting from LIB recycling in comparison to pyro processes. Additionally, organic components are not converted into GHGs as is the case for most pyro processes. There is a necessity to address the previously discussed safety issues in pre-treatment of LIBs prior to hydrometallurgical recovery. However, a breadth of methods to achieve this are demonstrated across available hydrometallurgical recovery processes. Recovered materials generally need to be thermally dried which increases energy consumption, cost and environmental impacts of processes.

The use of solvents with lower boiling points than water such as acetonitrile reduce energy requirements for drying and recovery through evaporation. The use of solvents does however require careful consideration and many of those employed in existing processes such as acetonitrile and NMP are hazardous. Another issue requiring careful consideration in hydrometallurgical processes that utilise protic solvents is the quality of recovered materials. If not properly removed, recovered cathodic, anodic materials and electrolyte materials may be unsuitable for reuse in new LIBs.

Hydro processes hold the disadvantage that often, intensive sorting of batteries by chemistry prior to processing is necessary. This is a potentially costly process which may impact the overall cost-benefit of recycling. Additionally, large volumes of potentially hazardous liquid waste can result from hydro processes.

## Future LIB Recycling Processes.

Development of the next generation of LIB recycling processes must take account of the following criteria for comparison with existing processes: energy savings; alleviation of material supply constraints; economics; compliance with regulations; feed requirements; utility of products; scale (Gaines and Nelson, 2010). Designing future recycling processes also requires an informed and accurate outlook on the changing nature and future composition of the waste stream (Charles *et al.*, 2017). Newer cathodic materials are generally of lower value than those with high Co and Ni content, which brings the economic viability of current pyrometallurgical LIB recycling processes into question (Gaines, 2014). Those now used in LIBs include $LiCoO_2$, $LiNi_yMn_yCo_{1-2y}O_2$, spinel $LiMn_2O_4$, $LiMn_{1.5}Ni_{0.5}O_4$ and olivine $LiFePO_4$ materials. Future generations of cathodes will probably be more complex containing more than one metal.

The following cathodes may be used in the next generation of lithium-ion batteries: $LiNi_{1-x}Co_xO_2$, $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$, $LiMn_{1.5}Ni_{0.5}O_4$, $LiMn_{1.42}Ni_{0.42}Co_{0.16}O_4$, $Li_3V_2(PO_4)_3$, $LiMnPO_4$, $LiCoPO_4$ and $LiFePO_4$. The recovery of cobalt, nickel and lithium from these materials seems to be particularly economically attractive (Chagnes and Pospiech, 2013). However, the proportion of these more valuable metals in cathodic materials of LIBs is decreasing as Co and Ni are replaced by Mn and other less valuable species. This should be of concern to operators of pyrometallurgical recovery processes which valorise only the metallic constituents of cathode materials and fail to recover lithium, electrolytes, aluminium current collectors and other organic components.

In regions, such as Europe where recycling is necessary to comply with legislation, and extended producer responsibility ensures that producers finance the costs of EoL treatment, the reduction in the derivable value of materials in LIBs does not necessarily present an obstacle to future recycling. Particularly if producers can offset lower derivable value from recycling with increased material cost savings for manufacturing through recovery of raw materials for direct use in new LIBs. However, many regions around the world do not have such legislative and regulatory mandates which drive recycling, and so ensuring profitability of recycling for such regions is essential for driving diversion of LIBs from landfill or other destinations where they pose risks to the environment and human health.

As the value of metal species in cathodic materials reduces, adoption of processes which recover cathode materials in a suitable form for reuse or added-value pre-cursor compounds for inclusion in LIB manufacturing will become increasingly important to maintain profitability, as will valorisation of all LIB components. In this regard, hydrometallurgical processing seems to offer the greatest potential. One such process developed by Worcester Polytechnic Institute is in the process of being commercialised by Battery Resources LLC. This hydrometallurgical process can accept any LIB regardless of chemistry and can directly synthesise new cathode materials ($LiNi_xMn_yCO_zO_2$,) as well as recover all components of LIBs with the exception of electrolytes (Zou *et al.*, 2013; Gratz *et al.*, 2014; Sa *et al.*, 2015). Such processes negate the requirement for pre-sorting LIBs by chemistry prior to processing, significantly reducing the overall costs of recycling through avoidance of this manually intensive pre-processing.

*Table 33: Ratio of Co, Ni and Mn in batteries recovered from WPI's recycling centres (Heelan* et al.*, 2016).*

| Year | %Co | %Ni | %Mn |
|---|---|---|---|
| 2012 | 90 | 6 | 4 |
| 2014 | 60 | 25 | 15 |

Lower cost recycling processes will also be important. Much progress has been made in minerals processing in terms of biological recovery processes and biological treatment of conventional municipal solid waste is widespread. Biological processing offers potential lower cost processing routes for LIBs, however, to date no such processes have been commercialised. Recovery of cathodic materials using biological treatment has been demonstrated (Mishra *et al.*, 2008).

In the interests of circular economy, recyclers should begin working with LIB manufacturers now to create effective circular product lifecycle for the next generation of LIBs. This should seek to enable effective reuse and refurbishment of LIBs, with efficient closed-loop recovery of materials during recycling (Larcher and Tarascon, 2015). Cost and environmental impacts should also be minimised. Several key developments in LIB recycling are of note to inform next generation recycling. These are exemplified in lower TRL processes discussed below.

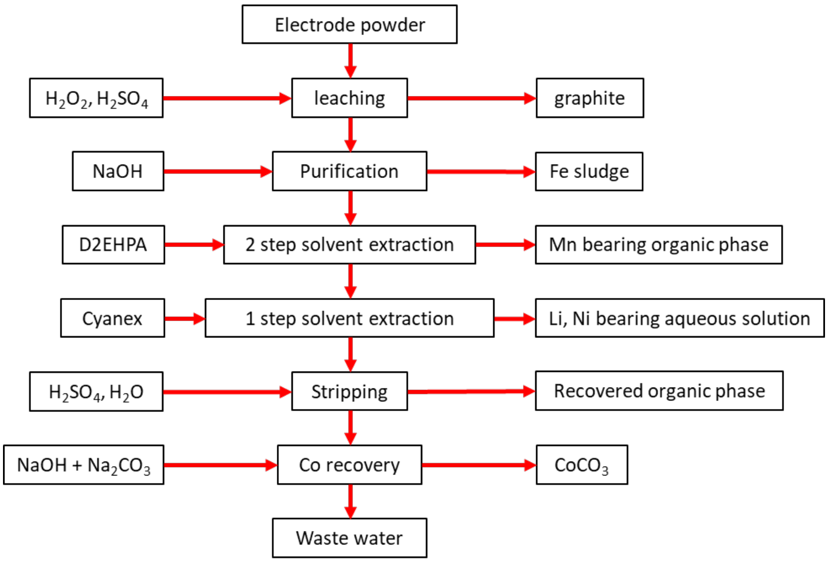
### HydroWEEE

The HydoWEEE mobile plant was developed under the EU FP7 Environment work program and the plant has been used to recover cobalt from electrode powder of LIBs (Figure 45). This necessitates mechanical pre-treatment to isolate this powder from other battery materials prior to treatment in the hydroWEEE process. Once isolate, the 'black mass' from LIBs is leached using sulfuric acid and hydrogen peroxide (34.5-36.5%) using a S/L of 1:10 and 15%v/v of the hydrogen peroxide solution with +100% acid excess leaving only graphite which is also recovered.

The leach solution is then purified by removal of Fe as sludge by precipitation with NaOH at pH 3.8. The Fe-containing sludge could be re-used for the production of added-value nanoproducts. In particular, magnetic nanoferrites were synthetized by a surfactant assisted hydrothermal route using iron and copper bearing solution. Produced nanoparticles could also be obtained using Fe-iron sludge as adsorbent material in the refining operation of waste waters emerging from the LIB recycling process. This is a novel attempt to maximise valorisation of Fe present, which although not a critical material owing to its Earth abundance, should still be recovered with maximum value for environmental reasons and to maximise the economic performance of the recycling system overall.

Solvent extraction is carried out on the leach solution following removal of Fe. First D2EPHA is used in a 2-step solvent extraction to remove manganese impurities. This is followed by a single extraction with Cyanex 272 to remove cobalt, leaving lithium and nickel in the aqueous phase for potential recovery. Cobalt ions are then stripped from the organic phase using 3M sulfuric acid and recovering by addition of NaOH to adjust pH to 3, followed by addition of sodium carbonate with stirring for 30 minutes which caused cobalt to precipitate as $CoCO_3$.

*Figure 45: The HydroWEEE process for recovery of Co from waste Li-ion batteries (Amato* et al.*, 2016).*



The HydroWEEE plant has recently found new application in the EU Life+ funded Direct production of New Electrode materials from battery recycling (DRONE) project which aims to recover high quality graphite, cobalt, nickel and manganese using the HydroWEEE plant, and demonstrate their suitability for production of new lithium-nickel-manganese-cobalt oxide (MNC) LIBs.(Eco Recycling S.L.R., 2020) This approach would enable generation of added-value precursors for manufacturing new LIBs in line with circular economy and increasing derivable value from the batteries.

## Organic acids

Much research is underway into new hydrometallurgical processes for LIBs which use organic acids in place of traditional strong inorganic acids. The field of cathode material recycling has recently been reviewed by Meshram *et al.*, (Eco Recycling S.L.R., 2020) who highlighted the need for green techniques, particularly those which are benign and avoid the use of high temperature and oxidative acid leaching conditions.

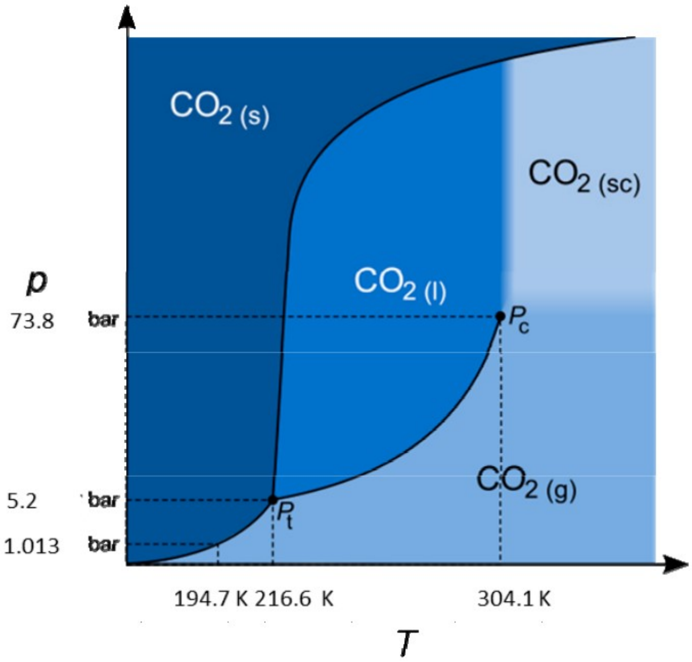
Current and emerging methods were examined in terms of various factors (selectivity, cost, etc.) that govern the use of organic acids in battery recycling. It was ascertained that the GHG emissions to extract Co using organic acids were an 1/8th of that using an inorganic acid leaching process. The review also highlights progress is required in terms of development of processes for separation of these metals from these lixiviants ensuring economics and environmental benefits are delivered. In light of previous comments on the environmental impacts of battery recycling processes, organic acids present an exciting lower environmental impact alternative to pyro and tradition hydrometallurgical recovery processes employed to date.

## Supercritical and subcritical $CO_2$

The application of supercritical $CO_2$ to EoL treatment of Li-ion batteries presents interesting potential for circular economy and safer treatment of batteries. When pressurized and heated above its critical point $CO_2$ is a single-phase supercritical fluid with tuneable liquid-like density (Figure 46). The primary advantage of the supercritical fluid for use in recycling is that during removal of the supercritical $CO_2$ (scCO2) by depressurization, it enters the gas phase without crossing a phase boundary, thus avoiding solvent residue in recovered materials which make liquid solvents problematic, often requiring thermal treatment of recovered materials to ensure residual solvents are driven off and that recovered materials are suitable for use in new applications. $CO_2$ is also abundant, low-cost, non-flammable, non-toxic, recyclable and has high permeability enabling rapid penetration of porous nanostructures such as Lithium-ion batteries (LIB) electrodes (Annohene and Tepper, 2019).

$CO_2$ has a relatively low critical temperature and pressure (critical point is 304.1 K, 7.38 MPa) and is therefore the most popular supercritical fluid used industrially since 1950 for applications including decaffeination of coffee, extraction of hops and production of aerogels. It can act as a solvent, anti-solvent, solute and reaction medium for materials processing. It is a non-polar solvent with large quadrupole moment and a polar carbonyl (C=O) bond which makes some materials soluble with the options to include additives to tune solubility of polar species.

*Figure 46: Pressure and temperature phase diagram of $CO_2$ [158,159]. K: Kelvin, p: pressure, T: temperature, sc: supercritical, l: liquid, g: gaseous, s: solid.*



## OnTo Technologies

OnTo Technologies have patented a method which utilises supercritical $CO_2$ to remove electrolyte from Li-ion batteries and built-up materials at the electrode│electrolyte interface that may cause excess internal impedance in cells (Sloop, 2010a, 2013), and which are highly toxic. This process was designed considering perceived issues with existing hydrometallurgical processes i.e.:

- The use of cryogenic liquid nitrogen to cool the batteries prior to mechanical treatment may be expensive and time-consuming.

- Hydrogen gas evolved in the reaction of lithium with water may pose an explosion hazard.
- Toxic $H_2S$ gas may be produced in the reaction mixture.
- Thermal removal of moisture from the carbonate product may be energy intensive.
- The process produces aqueous waste that may require disposal under expensive permits.
- Water may compromise the functionality of the electrolytes, cathodes and anodes as recycled battery materials for use in new batteries.

A recycling process utilising supercritical $CO_2$ treatment of cells was proposed, but to the best of the authors knowledge, never commercially implemented (Sloop, 2010a). The process has been adopted for pre-treatment of batteries to render them safe prior to further mechanical treatment by OnTo technologies in the US (Sloop, 2013). Collected LIBs are fully discharges before sorting according to chemistry and suitability for refurbishment and reuse. These are processed in batches with whole batteries placed into a vessel to which liquid $CO_2$ is added. Solubility enhancers (e.g alkyl ethers) may be added to the liquid to improve performance in the extraction of electrolyte from LIBs. Following addition of the fluid, temperature and pressure within this vessel are altered to generate a non-aqueous, aprotic supercritical fluid which dissolves the electrolyte within the LIBs. Pressure is increased by pumping liquid $CO_2$ into the vessel which breaches battery casings permeating the components of the cell interior.

Liquid $CO_2$ has a low surface tension enabling it to penetrate sub-micron pores. For this reason, it is ideal for permeating the porous structure of LIB electrodes without prior grinding of waste batteries for extraction of electrolytes, their degradation products, and electrode|electrolyte interphase materials. Electrolytes extracted and dissolved within $ssCO_2$ are simply recovered by evaporation and $CO_2$ is captured and recycled in the process. Battery 'shells' then remain for further processing, in a safer manner than would otherwise be possible. A second sorting of these shells can then be carried out to identify any cells damaged during the extraction. Those which are found to be no longer suitable for refurb can be sent for recycling via shredding under inert conditions to avoid contamination of materials, which are separated using sifting and standard density separation methods.

The opportunity to reuse entire cells once reconditioned and refilled with electrolyte as is possible with OnTo Technology's $ssCO_2$ process represents something more conducive to circular economy than simple recycling. This introduces an opportunity to refurbish and recondition whole cells, which according to the principles of circular economy, should yield greater environmental and economic benefits than any available recycling process, particularly pyrometallurgical ones. Refurbishable cells can potentially be re-filled with electrolyte in a moisture free atmosphere (though careful re-growth of electrode|electrolyte interphase may be necessary to ensure correct and safe functionality), and any breach in the cell sealed with epoxy resin. This also represents a method of rendering ruptured cells safe for further handling.

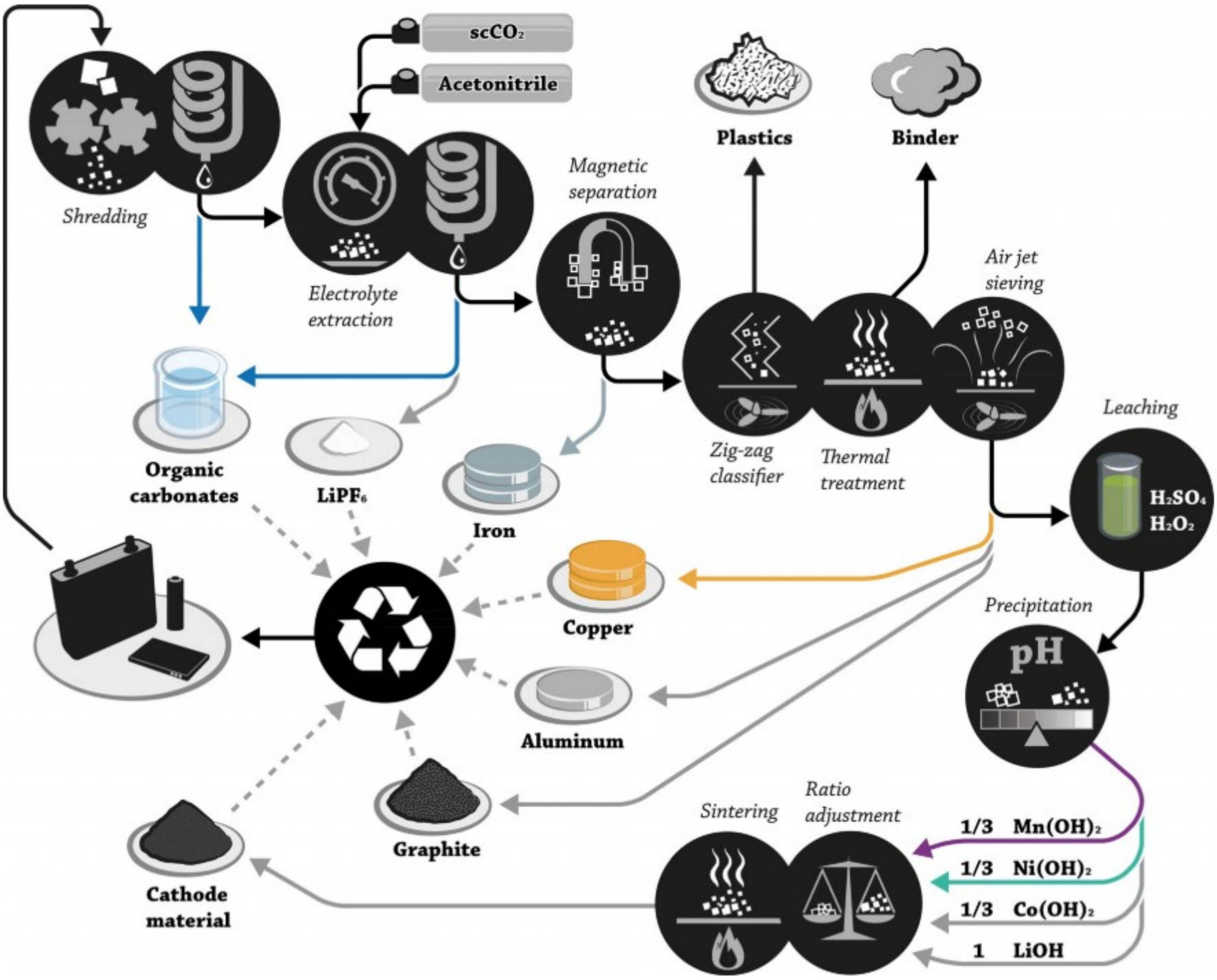
## Lithorec

The LithoRec projects were funded by the German Federal Ministry of the Environment, Nature Conservation, Building and Nuclear Safety and VDI/VDEInnovation+Technik GmbH. Considering issues with existing processes, the project aimed to develop a new recycling process for LIBs from hybrid electric vehicles ((H)EVs) with a focus on energy efficiency and a high material recycling rate. The Lithorec process uses mechanical, mild thermal and hydrometallurgical treatment to reclaim all materials of a battery system (Diekmann *et al.*, 2018; Kwade and Diekmann, 2018).

The follow-on project, LithoRec II, focused on optimization of each of the process steps and creation of a 100 tonnes/year capacity pilot plant. Economic assessment of the proposed system revealed investments in corresponding recycling equipment could be profitable in most scenarios but there are also high uncertainties regarding the main influencing factors: the future amount of spent battery systems and the achievable prices for the recycling products. Because of these reasons, the pilot plant was not produced but a smaller temporary demonstration plant was established in Braunschweig. The realized process chain (Figure 47) was composed of the process steps:

discharging, disassembly, crushing, drying, separation, and sieving. During its operating time of four months the process recycled 1.4 t of battery systems and reached a material recycling rate of 75–80% per battery system.

*Figure 47: Flowsheet for the LithoRec battery recycling process (Rothermel* et al.*, 2016).*



Following discharge, several options were investigated to remove electrolytes from crushed batteries to reclaim valuable electrolytes (solvents and salts), with $scCO_2$ proving to be the most effective way of recovering the salt intact and at maximum yield. Removal of electrolyte at this stage enables dry batteries to be processed preventing agglomeration of material particles for more effective separation. Subsequent mechanical treatment then enables isolation of plastic components of batteries, current collector metals and battery black mass. Black mass is subjected to hydrometallurgical treatment via leaching with sulfuric acid and peroxide results in dissolution of cathode materials allowing graphite to be sieved from solution. pH control then allows selective precipitation of Co, Ni, and Mn from solution, thereby enabling the process to accommodate various LIB chemistries without prior sorting of batteries according to their chemistry. Li remains in solution and is also recovered.

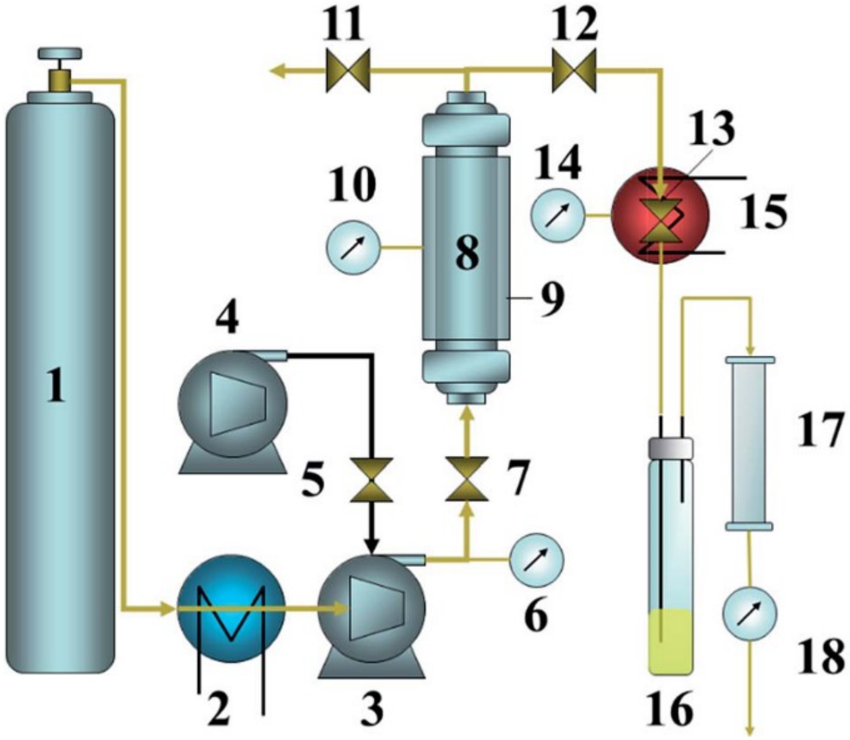
The process overall overcomes many of the issues with existing processes with regards to collective recycling of numerous LIB chemistries, recovery of graphite, Li and electrolytes. The use of $scCO_2$ post grinding differs from the approach of OnTo technologies which aim to render cells safe for grinding with $scCO_2$, whereas this process shreds discharged batteries under an inert atmosphere as a first stage of treatment in which evaporated electrolyte is collected by condensation, electrolyte remaining deeply incorporated within the shredded material may then be treated with $scCO_2$ and co-solvents to recover all remaining electrolyte components including the Li salts (e.g. $LiPF_6$) which are the most valuable component of the electrolyte. Thermal drying would be a more conventional treatment with capture of solvent by condensation, however this fails to extract the Li salts which are subsequently lost in processing, an issue which is avoided via the $scCO_2$ extraction route.

### Application and advantages of subcritical $CO_2$.

Since the publication of Sloop's first patent (Sloop, 2010b), a number of research groups have investigated the use of $ssCO_2$ for electrolyte extraction in recycling applications, and also for quantitative extraction of electrolyte, degradation products and electrode|electrolyte interphase materials for forensic study of electrolyte degradation processes during battery use (Nowak and Winter, 2017). The requirement for total quantitative removal of these materials for accurate study has provided progress in terms of the recovery efficiency using $CO_2$. Further investigations of electrolyte extraction with $scCO_2$ revealed that recovery efficiencies were sub-optimal and that use of flow-through experiments with additional co-solvents should be conducted towards process optimisation (Grützke *et al.*, 2014a).

Commercial extraction systems have been used to extract electrolytes from separators with a recovery rate of ~85% (Figure 48) (Liu *et al.*, 2014a). The Li salt ($LiPF_6$) was not recovered in sufficient quantities, and it was proposed that the salt hydrolyses during the $scCO_2$ treatment to $PF_6^-$. Reaction parameters (extraction pressure, temperature, and static time) were optimised (23.4 MPa, 40 °C, 45 minutes) revealing extraction pressure to be the most important factor for electrolyte extraction. Further research revealed the importance of matching the polarity of the extraction solvent with that of the non-polar carbonate solvents, which should be extracted with non-polar of weakly polar co-solvents, and that polar carbonates should be extracted with the addition of co-solvents to increase polarity of the $ssCO_2$ extraction media (Liu *et al.*, 2014b). The ability to tune the $scCO_2$ polarity by inclusion of co-solvents is an important tool for future recycling processes in order to optimise the efficiency with which electrolytes can be extracted from battery materials to maximise the value and utility of recovered materials in manufacturing.

*Figure 48: Schematic diagram of supercritical $CO_2$ extraction apparatus (Liu* et al.*, 2014a).*



*Key: 1 - CO2 cylinder; 2 - cooling bath; 3 - air driven fluid pump (gas booster pump); 4 - air compressor; 5 - air regulator; 6 - CO2 pressure; 7 - inlet valve; 8 - extraction vessel; 9 - heating jacket; 10 - vessel heat; 11 - vent valve; 12 - outlet valve; 13 - flow valve; 14 - valve heat; 15 - heating jacket; 16 - collecting vial, 17 - alumina filter; 18 - gas flow meter*

Rothermel *et al.* applied the methods used by Grützke *et al.* (Grützke *et al.*, 2014b; Liu *et al.*, 2014c) to extract electrolytes and investigate the effect on graphite anode recycling efficiency (Rothermel *et al.*, 2016). They used 3 approaches: i) thermal evaporation of volatile electrolyte components without recovery, ii) extraction of electrolyte with subcritical $CO_2$ and acetonitrile (ACN) co-solvent prior to thermal treatment; and iii) extraction with $scCO_2$ prior to thermal treatment. Thermal treatment is necessary to burn off the binder of the anode in order to separate graphite from current collector. The term subcritical was used in place of liquid $CO_2$ as the used parameters were subcritical and not liquid conditions, and this approach was found to recover 90% of electrolyte including Li salt and graphite with best electrochemical behaviour when reused in combination with a recycled $LiNi_{1/3}Co_{1/3}Mn_{1/3}O_2$ cathode, outperforming even fresh commercial synthetic graphite used as a benchmark.

Using the methods of Grützke *et al.* (Grützke *et al.*, 2014b; Liu *et al.*, 2014c), Friesen *et al.*, used subcritical $CO_2$ to extract electrolytes including their salts, and ageing products quantitatively from aged commercial 18650-type LIB cells to study the effects of temperature on aging of the batteries (Friesen *et al.*, 2017). This confirmed that the use of subcritical $CO_2$ could achieve effective removal of all electrolyte components and aging products regardless of polarity of the target materials. This process holds several advantages over alternative processes. Pre-cooling of LIBs is not necessary as Li metal present slowly reacts with $CO_2$ and a small amount of oxygen present to generate $LiCO_3$. In addition, no mechanical pre-treatment of LIBs is necessary as the subcritical $CO_2$ penetrates the structure of LIBs. Conversion of residual Li following discharge of cells to lithium carbonate eliminates the fire hazard associated with lithium metal, and no hydrogen gas is formed in the reaction which represents a potential explosion hazards in processes which hydrolyse Li in water or other protic solvents. Electrolytes and oligo-carbonate waste products formed at the electrode|electrolyte interface is soluble in subcritical $CO_2$ allowing them to be removed from batteries leaving cells along with their anhydrous character intact, potentially for reuse.

Electrolyte can simply be recovered by reducing the pressure in the collection vessel. This enables recovery of electrolyte and reuse of the $CO_2$ in a closed-loop system, saving the cost of new fluid for each extraction. In addition, the 'critical' carbon (graphite is now critical, supply at high grade dominated by China) is recovered with other organic components such as the polymer which separates battery electrodes, which are all burned off in smelting operation to generate energy or act as reducing agents in the process.

An interesting possibility from the use of this process is the refurbishment of batteries perceived to be functional by re-filling battery 'shells' with new or even recycled electrolyte, once they have undergone the subcritical $CO_2$ process. This presents an opportunity to generate additional value from suitable EoL batteries than is afforded through materials recovery (Sloop, 2010b). This could be very useful in terms of deriving sufficient value from waste LIBs to finance recycling as the trend in LIB chemistry shifts further towards the use of less valuable cathode materials. Additionally, all separated materials are suitable for re-use in new LIBs or alternative applications. The graphitic materials isolated from electrodes are potentially more valuable than the graphite which went into the original battery as these materials have established pathways for $Li^+$ movement and their surfaces have been stripped of reactive functional groups. Additional valuable materials recovered include salts, solvents and metal oxides.

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