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# Towards Increased Recovery of Critical Raw Materials from WEEE– evaluation of CRMs at a component level and pre-processing methods for interface optimisation with recovery processes





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# ABSTRACT

Increasing recovery of critical raw materials (CRMs) from waste electrical and electronic equipment (WEEE) is a strategic priority to mitigate supply risks. Today, CRM recovery rates are generally low, with increases requiring new recovery processes and interface optimisation with pre-processing to ensure appropriate material flows for efficient recovery are generated. Here, results from an industrial trial to increase CRM recovery from WEEE are presented to inform development of pre-processing strategies which generate such material flows. Au, Ag, Co, Ga, Mg, Nb, Ru, Pd, Ir, Y, Nd, Sb, Ta and W are identified with XRF in components of a range of WEEE samples including within individual printed circuit board (PCB) components. CRM distribution in PCBs is mapped by visual inspection with reference to this data. Cost-effective methods to disassemble WEEE; isolate CRM bearing components, and upgrade/concentrate CRMs are evaluated for industrial adoption. A guillotine is found most suitable for LCD disassembly and separation of Au edge-contacts from PCBs, while cryocracking is best for isolation of internal components of digital media devices. Thermal PCB disassembly with a solder bath for simultaneous SMD removal and subsequent sieving to sort SMDs thereby concentrating CRMs for recovery is a promising approach. Microwave ashing of PCBs to concentrate CRMs is promising although off-gas treatment would be required. Recovery potential of identified CRMs from material streams generated is found to be poor due to lack of suitable recovery infrastructure except for precious and platinum group metals in PCBs, but available pyrometallurgical recovery permanently dissipates other CRMs present.

AATF	approved authorised treatment facility	HDD	hard disk drive
BAT	best available treatment	IC	integrated circuit
CCFL	coldcathodde fluorescent lamp	ICT	information and communications technology
COG	cut-off-grade	IL	ionic liquid
C-Si PV	crystalline silicon photovoltaics	IoT	internet of things
CRMs	critical raw materials	IPR	individual producer responsibility
DEM	discreteelement method	LCOE	levelised cost of energy
DES	deep eutectic solvent	LIBs	Li-ion batteries
EEE	electrical and electronic equipment	MFA	material flow analysis
DfRR	design for reuse and recycling	MLCC	multi-layer ceramic capacitor
EHF	electrohydraulic fragmentation	NIB	neodymiumiron-boron alloy
EPR	extended producer responsibility	OEMs	original equipment manufacturer
EV	electric vehicle	PCI	peripheral component interconnect
GHG	greenhouse gas	PGMs	platinum group metals

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PMs	precious metals
PV	photovoltaics
REMs	rare earth metals
RMI	European Raw Materials Initiative
R&I	research & innovation
SMDs	surface-mount device components
SSD	solid-state drive
THDs	through-hole device components
TRL	technology readiness level
WEEE	waste electrical and electronic equipment
XRF	X-ray fluorescence spectroscopy

# 1. Introduction

The global availability of raw materials is increasingly under pressure due to growing demand and the increasing likelihood of supply bottlenecks. Global resource security is a growing concern, and access to 'critical' raw materials (CRMs) has become a priority issue for governments around the world. CRMs are linked to all industries and stages of supply chains, and economies which rely heavily on imports are particularly susceptible to supply issues. Accelerating technology innovation cycles and growth in emerging economies has steadily increased demand for raw materials with predictions showing that global resource use could double between 2010 and 2030 (Gislev and Grohol, 2018).

CRMs are widely used in electrical and electronic equipment (EEE), particularly in emerging technologies (Table 1). Demand for CRMs will inevitably increase with wider penetration of consumer and industrial markets, and adoption of industry 4.0 and internet of things (IoT) technologies (Ku, 2018). Low-carbon technologies currently account for 20% of global CRMs consumption, and demand will increase with widescale deployment to meet the goals of the Paris Agreement (Gradin et al., 2018; Hache et al., 2019; Hernandez et al., 2017; Månberger and Stenqvist, 2018; Miyamoto et al., 2019). All sectors must reduce greenhouse gas (GHG) emissions, and energy efficiency measures contribute the majority of reductions for the transformation, buildings, industry and transport sectors. Such measures require the manufacture and installation of new energy-efficient technologies, and these contain a wide range of CRMs (IEA, 2017). IRENA's 'REmap Case' provides a technology roadmap to deliver 71% reduction in global energy-related CO<sub>2</sub> emissions by 2050 by reducing energy consumption (from 51 to 38 GJ/cap); increasing renewables share in energy generation (from 25% to 86%); increasing electricity share in total energy consumption (from 29% to 49%); deploying ~1.2 billion passenger

Table 1

Major applications of 'technology metals' and their driving emerging technologies (Charles, 2018).

Raw Material	Emerging Technologies
Ga	Thin layer photovoltaics (PV), ICs, LEDs
Li	Li-ion batteries
Nd	Permanent magnets, laser technology
In	Displays, thin layer photovoltaics
Ge	Fibre optic cable, IR optical technologies,
Pt	Fuel cells, catalysts
Та	Micro capacitors, medical technology
Ag	RFID, lead-free soft solder, Si-PV
Со	Li-ion batteries, synthetic fuels
Pd	Catalysts, seawater desalination
Ti	Seawater desalination, implants
Cu	Efficient electric motors, RFID
Nb	Micro capacitors, ferroalloys
Sb	Flame retardant, dopant in silicon wafers, Pb-acid battery alloys
Cr	Seawater desalination, marine technologies
V	Reflow batteries
REMs	Permanent magnets for wind turbines & electric vehicles (Nd,
	Dy, Pr), cathodes of LFYP Li-ion batteries (Y)

electric vehicles (EVs); and achieving 19 EJ of hydrogen production with renewable electricity (IRENA, 2019a). By 2050, 20 TW of installed power capacity must be in place, including 8.5 and 6.0 TW of solar photovoltaics (PV) and wind power respectively, requiring PV and wind installation rates to reach 360 GW/yr (from 109 GW/yr in 2018) and 240 GW/yr (from 54 GW/yr in 2018) respectively (IRENA, 2019b). Battery storage in stationary applications must increase from 2 to 175 GW by 2030 to support 100% renewable mini-grids and solar home systems (IRENA, 2017). Although green-technology deployment reduces reliance on fossil fuels and GHG emissions, it increases reliance on CRM supplies. PV deployment will increase In, Si, Ga, Te, and Ag demand. Production of permanent magnets for wind turbines and EV motors will increase demand for Dv. Nd and Pr. Battery systems for stationary renewable energy storage and EVs will increase V demand for reflow batteries; Li, Co, Y and natural graphite demand for Li-ion batteries (LIBs); and platinum group metal (PGM) demand for hydrogen generation catalysts. To meet climate and energy targets in the EU, CRM demand is expected to increase by a factor of 20 for certain materials (EU JRC, 2016), and access to CRMs may limit technological deployment and our ability to combat climate change (de Koning et al., 2018; Elshkaki and Graedel, 2013; Kleijn et al., 2011; Roelich et al., 2014).

In 2010 the European Raw Materials Initiative (RMI) undertook an evaluation of 41 raw materials and concluded that 14 of these were 'critical' to the EU due to their high relative economic importance and supply risk (EC, 2010a). The list, known as the 'EU14' (Table 2) predominantly consisted of 'technology metals', so called due to their vital use in numerous high-tech applications. Technology metals have remained the core of the EU CRM list through two revisions with expansions of the range of materials evaluated (EC, 2017, 2014, 2011a), and are consistently found critical by other assessments (Frenzel et al., 2017; Hayes and McCullough, 2018; Jin et al., 2016). The strategic importance of securing supply of the precious metals (PMs)<sup>1</sup> Au and Ag has been well explored, with PMs found to be critical by numerous stakeholders including the Chinese c-Si PV industry (Charles et al., 2017; DEFRA and BIS, 2012; Graedel et al., 2012; Li and Adachi, 2019; Morley and Eatherley, 2008; Nassar et al., 2012; Schneider et al., 2014).

In most cases CRM availability is not an issue of geological abundance but access to supply. CRM production is concentrated within a small number of countries, many of which have resource hungry economies of their own. In 2010, >90% of rare earth metals (REMs) and Sb, and >75% of Ge and W were produced in China; 90% of Nb was produced in Brazil; and 77% of Pt originated in South Africa. Today, this picture has altered little, with geopolitical factors threatening supply to the rest of the world (Gislev and Grohol, 2018). For example, export restrictions such as those imposed by China on REMs which caused the rare earths trade dispute, and political instability in producer nations such as the ongoing conflict in DRC affecting Ta and Co supply which also introduces 'conflict mineral' issues. Many producer nations are pursuing strategies through trade, taxation and investment policies; export taxes and quotas; subsidies; price-fixing; and restrictive investment rules to conserve resources for domestic use in contradiction of WTO rules (EC, 2010a). Rapid primary supply solutions to booms in CRM demand do not exists (EC, 2010b). Bringing new mines online is a lengthy process and many CRMs are minor by-products of major metals production, so supply depends upon markets and production of major metals e.g. In is a by-product of Zn and Pb production so In production depends upon market conditions for Zn and Pb (Frenzel et al., 2017). These factors, in addition to stockpiling and

<sup>&</sup>lt;sup>1</sup> 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.

The 'EU14' list of critical raw materials (EC, 2010a)

antimony (Sb)	beryllium (Be)	cobalt (Co)	fluorspar	gallium (Ga)
germanium (Ge)	graphite (C)	indium (In)	magnesium (Mg)	niobium (Nb)
platinum group metals (PGMs) <sup>1</sup>	rare earth metals (REMs) <sup>2</sup>	tantalum (Ta)	tungsten (W)	

<sup>1</sup> PGMs include platinum (Pt), palladium (Pd), ruthenium (Ru), iridium (Ir), rhodium (Rh), osmium (Os)

<sup>2</sup> REMs include: lanthanide elements and yttrium (Y).

supply interruption, cause price volatility, a concern for organisations relying on CRMs when price spikes cannot be passed on to consumers (DEFRA and BIS, 2012a). Price spikes are a concern for renewable energy products for which levelised cost of energy (LCOE) determines viability of deployment (Leader et al., 2019). In light of the global environmental crisis, the impacts of primary production must also be considered.

One solution to materials criticality issues is adoption of circular economy (Krystofik et al., 2018), in which products, and their CRMs, are retained within the economy over multiple product life cycles through product/components reuse and efficient recycling; thus maximising economic productivity of CRMs and reducing global demand. FirstSolar produce CdTe PV modules and offer a takeback and recycling service which recovers the unrefined semiconductor materials for sale to their suppliers for refinement (Tao and Yu, 2015). This circular economy strategy ensures critical Te contained in end-of-life (EoL) modules is recovered, granting access to secondary Te supply for new module production while offsetting raw materials costs and reducing product LCOE and embodied emissions (CO2eq./kWh). In general, circular economy will decouple economic growth from primary CRM production and utilise secondary CRM sources; though today end-of-life (EoL) CRM recycling rates remain <1% for many technology metals, and barely exceed 50% even for the most valuable i.e. Au and PGMs (Charles et al., 2018; Graedel et al., 2011; Deloitte Sustainability et al., 2017). Secondary CRMs supply to mitigate supply bottlenecks is therefore very limited and increasing CRM recycling rates is essential if growing demand is to be met. Waste EEE (WEEE or e-waste) is the largest and fastest growing waste stream on the planet (~50 Mt/yr, 3-5% growth/year), and an important reserve of secondary CRMs for circular economy and reusable products/components that contain them (Charles et al., 2017; Cucchiella et al., 2015). To exploit these CRMs and enhance secondary supply, reuse and recycling rates of WEEE and CRM recovery efficiencies must increase (Buchert et al., 2008; Moss et al., 2013; Valero et al., 2018; Viebahn et al., 2015). To achieve this, WEEE must be collected and introduced into the recycling process chain for pre-processing and materials recovery. In the UK, this occurs when collected WEEE is received by an authorised approved treatment facility (AATF) for pre-processing (Figure 1). During pre-processing WEEE is assessed for reuse potential, and reusable EEE is sold directly to new users or refurbished/upgraded first. Unsuitable WEEE for reuse undergoes separation into material and component fractions which are sent to downstream recovery processes. Materials separation can be conducted with automated, manual or semi-automated processes. Manual pre-processing is relatively costly as it is labour and time intensive. Products are manually disassembled into constituent parts and sub-assemblies with all contained CRMs intact, providing opportunity to isolate reusable parts (e.g. graphics cards from PCs) and components of sub-assemblies (e.g. surface mounted device components (SMDs) of printed circuit boards (PCBs) such as CPUs on motherboards). The average life of a PCB is 20,000 hours, just 5% of the designed lifespan of its components. At EoL, many components are functional and potentially reusable multiple times (Xiang et al., 2014). For viable reuse and CRM recovery, value derived from sale of equipment, components and recovered materials must be enough to filter back through the recycling value chain to recoup costs incurred. These costs include mass-based recycling charges imposed by recovery facilities on materials accepted from pre-processors. If concentrations of CRMs in items are too low to

yield enough revenue to recoup recycling costs these items fall below cut-of-grade<sup>2</sup> for recycling, since recycling then results in an economic loss rather than a profit. Where this is the case, materials separation is conducted with less costly automated processes which shred whole items (comminution) and separate bulk material fractions by differences in physical properties (e.g. magnetic separation for ferrous fraction; eddy current/corona separation for aluminium and copper, density separation for plastics, etc.) (Kell, 2009). This dissipates CRMs as small particles throughout the shredded materials, which are thus permanently lost through unintended co-separation with bulk fractions, and eliminates any possibility of reuse. Component reuse is only viable with access to markets for valorisation i.e. internal reuse operations or sale for use in repair, upgrade/refurbishment, remanufacturing or cascading to manufacture new EEE. Reuse retains all contained CRMs within the economy, offsetting demand for virgin CRMs. However, rapid technological innovation results in obsolescence, preventing reuse, and potential industrial scale users of recovered post-consumer components require quality assurance. Barriers to PCB component reuse are greater still, since this requires cost-effective disassembly which does not compromise functionality while enabling supply at a competitive price compared to new components manufactured at scale (Xiang et al., 2014).

Pre-processing is crucial to overall CRM retention. Decisions made here determine whether CRMs in WEEE will ultimately be retained through reuse/recovery, or lost forever. If output material fractions containing CRMs are sent to recovery processes which do not target them, they are permanently lost. Barriers to CRM recovery from WEEE include: low collection rates; poor product design for EoL; dissipative applications of small quantities of CRMs in WEEE; and losses of CRMs in recycling during comminution and pyrometallurgical recovery - a process which represent current best available treatment (BAT) but targets high value materials in favour of lower value CRMs (Avarmaa et al., 2019; Chancerel et al., 2009; Charles, 2018; Charles et al., 2018; Charles et al., 2017; Hagelüken and Meskers, 2013; 2008; Klemettinen et al., 2019; Marra et al., 2018; Parker and Arendorf, 2012; Sukhomlinov et al., 2019c, 2019a; Tesfaye et al., 2017; Wan et al., 2018). Numerous research programs have been funded to tackle these barriers (Løvik et al., 2018), and a diverse range of alternative recovery technologies are emerging which overcome existing issues with pyrometallurgical processes and hold the potential to increase CRM recovery rates, including: hydrometallurgical, cementation, precipitation, ion exchange, electrochemical, ionic liquid (IL), deep eutectic solvent (DES), biological, and solvent extraction, processes (Dominguez-Benetton et al., 2018; Işıldar et al., 2019; Perez et al., 2019; Ryder et al., 2020; Schaeffer et al., 2018; Sethurajan et al., 2019). In general, these technologies will enable viable recovery of a greater range of CRMs from WEEE in smaller-scale processes with lower capital investment, energy consumption, and waste and emissions produced. Lower processing costs will result in lower COGs for recovery, and new leaching and bio-leaching options will enable extraction of a greater range of CRMs while avoiding use of toxic chemicals (Isildar et al.,

<sup>&</sup>lt;sup>2</sup> 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 (Charles, 2018; Charles et al., 2017).



Figure 1. Decisions tree for treatment of WEEE by an AATF

2018; Ryder et al., 2020). In addition, it is possible to cycle water and reagents in these processes, reducing chemical consumption and water treatment requirements. Ion-exchange, electrowinning, solvent extraction (including IL and DES approaches), precipitation, and cementation, provide greater capability to selectively recover CRMs from complex mixed metal solutions with high efficiency, and can be combined with aforementioned extraction processes into bespoke processes for CRM recovery from specific material/component outputs from pre-processing. Even with increasing availability of promising recovery technologies, increasing CRM recycling rates from WEEE will require interface optimisation between pre-processing and recovery (Hagelüken, 2006). Isolation of components prior to comminution and/or smelting will prevent dissipative losses of CRMs, and generate fractions from which CRMs may be recovered in emerging processes. Dismantling SMDs from PCBs is therefore the first and most important step in a PCB recycling chain which conserves CRMs, enables components reuse, and maximises economic returns and environmental benefits.

Much research is underway to develop pre-processing methods which overcome barriers to CRM recovery and component reuse, including cost-effective processes to disassemble WEEE and access CRM bearing components. Manual isolation of PCBs from WEEE is optimal for CRM recovery as losses in shredding are avoided. However, manual processing is time and labour intensive, and therefore uneconomical in developed countries, except for WEEE with the most valuable PCBs with high PM content. In the future, product design for disassembly will help to overcome this issue. However for current WEEE, automation to achieve cost-effective component separation has been conducted in a number of ways. Mechanical coarse grinding of WEEE with hammer mills to break open housings and access PCBs whilst attempting to minimise damage and dissipation of CRMs is used, but material flow analysis (MFA) studies have shown bottom-line recovery efficiencies of Au from information and communications technology (ICT) equipment using this method to be as low as 70% (Hagelüken and Meskers, 2013). Tsunazawa et al. (2018) evaluated comminution of smartphones, microwave ovens and rice cookers, with a drum-type agitation mill to liberate PCBs whilst minimising damage to them. Process simulation using discrete element method (DEM) with a particle-based rigid-body model allowed equations to be developed which determine optimum process parameters to minimise PCB breakage while achieveing good separation from appliances. An underwater explosion technique has been used by Matsuo et al. (2011) for rapid disassembly of mobile phones. Robotics present automation options for rapid throughput and cost-effective disassembly which avoid CRM dissipation. Apple have introduced 'Daisy' - succesor of 'Liam the robot' for disassembly of devices received through their product take-back program, utilising inhouse 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 (2006) 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. 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 (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 with the same issue of non-uniformity, 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 (Kaya, 2019; Kellner, 2009; Ryen et al., 2018).

Once isolated, manual or automated PCB disassembly can be used to remove PCB components, either selectively or simultaneously, with a 2 step process in which: i) solder joints attaching components to PCBs are broken in some way; and ii) components are dislodged from boards. Selective PCB disassembly allows valuable components to be 'cherry picked' from PCBs. Simultaneous disassembly removes all SMDs enmasse, and these require further sorting into material flows for reuse and CRM recovery. Desoldering methods have been well reviewed in other publications (Kaya, 2020, 2019). A number of processes are employed industrially, most commonly in the US and China, with much research underway to develop more environmentally friendly 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 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. Through-hole device (THD) and mechanically fastened components were not as effectively removed as surface mounted device components (SMDs) (WRAP, 2018). Zhang et al. (2017, 2015) developed fast, economic and environmentally friendly selective leaching systems which achieve  $\sim$ 100% solder dissolution at room temperature in 35 minutes using 2.5 M HBF<sub>4</sub> with 0.4 mol/L H<sub>2</sub>O<sub>2</sub>; and 45 mins using 3.5 M methanesulfonic acid (MSA) with 0.5 mol/L H<sub>2</sub>O<sub>2</sub>. Ionic liquids also hold great potential to avoid consumption of reagents and component damage. Zeng et al. (2013) achieved complete solder dissolution and 90% component liberation by submerging PCBs in the IL  $[BMIm][BF_4]$  for 12 minutes at 250 °C with stirring at 45 rpm in the absence of air/oxygen. Upon cooling dissolved solder precipitates, enabling recovery and reuse of the costly IL and solder alloy. A spray based industrial system was proposed and a cost-benefit analysis conducted which showed the process becomes economical compared to manual and semi-automated alternative processes at >3kt annual throughput. Zhu et al. (2012a, 2012b) achieved complete solder dissolution by treating PCBs with [EMIM<sup>+</sup>][BF<sub>4</sub>] 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 retarded epoxy, enabling Cu-foil and fibreglass recovery from PCBs, which are promising preliminary results towards valorisation of depopulated PCBs. 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<sup>3</sup> generally melt at higher temperatures e.g. Sn-Ag-Cu melts > 210 °C. The required desoldering temperature therefore varies 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  $\pm$  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 a rod and brush type IR heating system capable of removing 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 simultaneously disassembled from PCBs via mechanical abrasion, as a means to sort components by type and thus potentially CRM content. 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. 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 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).

Advanced disassembly and recovery systems require significant capital investment which is a barrier to adoption in the developing world to which vast quantities of WEEE are exported from the developed world and where domestic generation is increasing. Instead, treatment of WEEE by the informal sector in hazardous and sub-optimal processes is common (Işıldar et al., 2018). Manual pre-processing is favoured in developing regions, since far lower capital investment is required. The high labour intensity required creates much needed jobs in regions of high unemployment. Informal treatment uses manual tools such as chisels, hammers, cutting torches or hot plates to desolder PCBs and isolate valuable components for sale to acid strippers. Such processes are hazardous to the environment and human health, and result in dissipation of CRMs (Terazono et al., 2016; Wang et al., 2011; Wen et al., 2008). This ongoing situation represents a global sink for CRMs and must be addressed to mitigate global materials criticality. The prevelence of manual processes present an opportunity to preprocess WEEE in an optimal manner for CRM recovery. With access to appropriate markets in the developed world, value generated from WEEE and CRM recovery rates could increase through a 'best-of-twoworlds' approach to global WEEE management (Manhart, 2011).

More information to inform appropriate pre-processing which avoids CRM losses during recycling is necessary (Hagelüken, 2006; Punkkinen et al., 2017). This will be vital to inform manual recycling and the development of automated disassembly and novel CRM recovery processes. Here, results of an industrial study which investigated methods to enhance the value generated from WEEE recycling through increased CRM recovery, conducted at an AATF in the South Wales Valleys operating manual pre-processing of WEEE, are presented. Manual processing offered the opportunity to investigate methods which avoid dissipative CRM losses and increase recovery potential through interface optimisation with recovery processes. The purpose of this article is to:

- i) explore the occurrence and distribution of CRMs at a component level in WEEE to inform future recycling strategies;
- ii) evaluate methods to augment manual pre-processing and generate suitable outputs for CRM recovery;
- iii) examine current and future recovery potential of identified CRMs considering currently available, and pipeline, recovery technologies.

CRMs (EU14 and PMs) in samples gathered from the AATF were identified, and pre-processing of these items to generate CRM rich fractions for downstream recovery investigated. Pre-processing methods investigated include: product disassembly and thermal PCB desoldering; PCB component separation; and thermal upgrade processes to concentrate CRMs in output fractions which exceed cut-offgrade for recovery. Data gathered on CRMs in PCB components was used to map their distribution in a TV PCB (sample 16) by visual inspection of surface components, and the accuracy of this map examined by subsequent XRF analysis of the sample. This enabled an evaluation of the potential for optical identification of PCB components and their contained CRMs to direct future recycling strategies. The practical

 $<sup>^3</sup>$  Pb-free solder is becoming more prevalent in WEEE since the EU RoHS Directive ban of Pb-solder (EC, 2011).

Samples for analysis supplied by the AATF

#	Sample Item	No. of samples	Туре
1	Domestic microwave magnetron	12	Component
2	Hard disk drives (assorted 31/2 inch drives)	12	Component
3	TV power boards (2 types)	6	Component
4	Set top box (STB) front panel PCBs (2 types)	12	Component
5	Li-ion laptop battery packs	11	Component
6	14 inch laptop screens	12	Component
7	Mobile phones (assorted)	12	Whole product
8	MP3 players (assorted)	6	Whole product
9	RSA security anti-hacking dongle	12	Whole product
10	Digital cameras (assorted)	12	Whole product
11	PCB from TV LCD	5	Component
12	Graphics card	5	Component
13	Server board	5	Component
14	Ethernet card	3	Component
15	Refrigerator power board	2	Component
16	Main PCB from TV	1	Component

applications of these findings to improve CRM recovery prospects are discussed.

#### 2. Experimental

#### 2.1. Sample selection

Samples selected were high-volume items in WEEE received by the AATF, and known, from operator knowledge and literature data, to contain CRMs and therefore represent the highest potential for CRM recovery (Buchert et al., 2012, 2008; Cucchiella et al., 2015; Jandric et al., 2018; Oguchi et al., 2011). Those selected are given in Table 3. Samples 1-6 and 11-16 were generated through existing manual disassembly operations at the AATF. Samples 7-10 are whole products which are not currently disassembled as it is uneconomical due to poor product design for recycling. Samples 11- 16 were chosen to demonstrate/evaluate methods for disassembly of CRM bearing components from PCBs. Duplicates were gathered for repeat analyses and evaluation of multiple recycling methods.

#### 2.2. Identification and quantification of CRMs in samples

XRF analysis for qualitative identification of CRMs was conducted with a Fischer Instrumentation Fischerscope XDAL-FD system and Fischer WinFTM software. To calibrate the instrument and verify its suitability for identification of CRMs in samples, standard spectra of CRMs were obtained from analysis of Pd, Pt, Au, Ge, Co, Ta, Ag, W, In, Mg, Nb and Y metal foils using manufacturers recommended setting for detection of trace metals i.e. 50 keV with a beam diameter of 0.3 mm and Al primary filter applied. Attempts were made to detect CRM foils through i) 0.1 mm Al foil and ii) 0.1 mm 18/8 stainless-steel foil using these settings. All elements were visible through aluminium, with reduced peak intensity in the cases of W, Nb and Y; through the stainlesssteel foil only Ag, In and Ge were visible. Results suggest that, due to matrix effects, it may be necessary to cut open or grind components to powder when encapsulated in steel, aluminium or plastic to detect CRMs. Where this was necessary is indicated in the results section, and, unless otherwise stated, CRMs were detected with surface scanning of components only. Samples were analysed with 16s scan times and beam cross-section of 0.3-1.0 mm, varied as appropriate for the size of component analysed. Only quantitative analysis of Ta-capacitors was performed via XRF, by grinding of 12  $\times$  100  $\mu$ F 10V 107A capacitors isolated from samples, with metallic Zn as an internal standard, using an IKA-A10 cutting mill to yield a homogenous powder. XRF was used to measure relative wt% of elements present, absolute wt% of Ta was

#### Table 4

Analytical technique and digestion procedures for quantitative determination of CRMs in samples

Sample	Method	Element	Sample preparation
Magnetron ferrite core	AAS	Со	Digest whole sample in 1:1 HCl
Magnetron magnet	AAS	Со	Digest 1 g sample in 1:1 HCl
Li-ion battery (LIB) cells	AAS	Со	Cut open cells and digest electrodes in 1:1 HNO <sub>3</sub>
Contacts from PCBs	AAS	Au, Cu	Digest whole samples in 1:1 aqua regia (3:1 HCl/HNO <sub>3</sub> )
LCD display CCFL	ICP-MS	Y	Digest phosphor in 1:1 HCl
MLCCs from PCBs	ICP-MS	Y	Digest whole component in 1:1 HCl
HDD magnets	ICP-MS	Nd	Digest whole sample in 1:1 HCl
Mobile phone speaker magnets	ICP-MS	Nd	Digest whole sample in 1:1 HCl

calculated from the known mass of Zn present.

The analytical techniques and digestion methods used for quantitative analysis of CRMs in samples are given in Table 4. Digests for ICP-MS and AAS analysis were prepared by oxidative digestion of samples by boiling in acids for 3 hours or until total sample dissolution was achieved, while maintaining a acid volume/sample mass ratio of 40 ml/ g (Charles, 2012; Park and Fray, 2009). Three sub-samples were taken for analysis from each digestion and results are taken as the mean with errors reported as the standard error on the mean. ICP-MS was conducted with a Perkin Elmer Elan 6100 ICP-MS instrument and Elan instrument control software (version 3.3) using argon carrier gas and a plasma flow rate of 0.92 L/min. Three replicate readings were taken for each sample with sample uptake delay of 40 s; read delay of 20 s; and rinse time of 15 s. AAS was conducted with a Perkin Elmer AAnalyst 200 instrument with air-acetylene flame and Perkin Elmer Lumina Hollow Cathode Lamps. Three replicate readings were taken for each sample, using an integration time of 3 seconds and read delay of 5 seconds. Calibration was conducted with solutions of known concentrations made from purchased standard solutions. Solutions of known concentration were prepared from standard solutions and analysed alongside samples to verify instrument precision and accuracy. HCl (31.5-33.0%) and HNO<sub>3</sub> (69-72%) were CertiFied AR (analytical reagent) grade purchased from Fisher Chemicals and used as received. All elemental standard solution used were TraceCERT<sup>®</sup> single element solutions (1000 mg/L) purchased from Sigma-Aldrich and used as received.

#### 2.3. Disassembly & separation processes

Manual disassembly of samples into components and sub-assemblies was performed with standard tools and equipment including: cordless power screw drivers, snips, pliers, scalpels, Dremel multi-tool with grinding and cutting attachments, hammer, scissors, precision screw driver set, guillotine, and a Rubi ND-200 wet saw electric tile cutter. Cryocracking was performed by submersion of samples 7-10 in liquid nitrogen for 20 seconds to embrittle plastic housings before striking with a hammer to shatter housings to enable collection of internal components (Gente et al., 2004). Various methods of thermal desoldering were attempted to isolate components from PCBs including using a: Dremel 2200 Versaflame blow torch; soldering iron; 2kW paint stripping gun; 500 W solder bath; and an NIR oven. An Adphos NIR lab unit with 1 m heated length was used to heat PCBs and soften solder, using a power setting of 70%, rate of travel through the heated length of 5-7 m/min, and heating temperature of 200-240 °C. A comparison of the techniques is made to highlight which are suitable for industrial application. The suitability of isolated SMDs for recovery was verified by submission of samples to a refinery for PM analysis by fire assay (results of Au, Ag and Pd content of SMDs given in Table 9).

#### 2.4. Upgrade and concentration processes

To concentrate CRMs and increase grade for recovery, dry ashing of isolated edge contacts from PCBs was conducted at 800 °C for 30 minutes using a CEM Phoenix microwave muffle furnace, (Marlon and Moraes, 2014). In order to concentrate CRMs in mixed simultaneously disassembled SMD PCB components into distinct fractions, sieving with mesh sizes of: 9.52 mm, 2.80 mm, 1.68 mm and 1.4 mm was used. Isolated SMDs were combined with virgin SMDs to give an appropriate quantity for evaluation of the sieving procedure. Components were shaken in the stack of sieves for 60 s.

#### 3. Results & Discussion

#### 3.1. Identification of CRMs in Samples

To gain insight into the occurrence and distribution of CRMs in WEEE to inform pre-processing to generate optimal CRM rich output fractions for downstream recovery, sample disassembly and component analysis was conducted. CRMs identified in samples are differentiated into those which are product specific and those occurring in common components of sample PCBs.

#### 3.1.1. Product Specific CRMs

Details of CRMs identified in samples are given in Table 5 along with literature data from previous studies of CRMs in WEEE items. XRF failed to reveal the presence of the REMs Dy and Pr known to be present in NIB alloy (Buchert et al., 2012) and Ce, La, Eu, Gd and Tb known to be present in CCFL phosphors (Dupont and Binnemans, 2015; Hobohm et al., 2016). Pt in HDD platers and In in LCD screens of all devices were also undetected by XRF although known to be present (Buchert et al., 2012). In all cases this is probably due to the low levels of CRMs present falling below the instrument detection limit. Quantitative analysis of identified CRMs in certain samples, i.e. Co in LIBs, Nd in NIB magnets, and Y in CCFLs, was conducted to give a basis for discussion of recovery potential with industry experts. Co content of packet type LIBs from phones and MP3 players correlates well with literature data (~19 wt%). Co content of laptop battery packs was found to be  $\sim$ 50 g (15.8 wt%) which differs from data presented by Buchert et al. (2012) of 65 g (13.8 wt%). HDD NIB magnets were found to contain ~35 wt% Nd in their alloy, ~5% higher than reported for 21/2 inch HDD magnets by Buchert et al., (2012) and ~12% higher than reported by Ueberschaar and Rotter (2015). Nd content of NIB speaker magnets was found to be  $\sim 28$ wt%, which correlates well with literature data. An order of magnitude discrepancy was found between measured Y content of CCFL phosphors in laptop LCD samples presented here (~13 mg) and that reported by Buchert et al. (2012) (1.8 mg). One likely source of discrepancy is that Buchert et al., (2012) have calculated this quantity from mean measurements of CCFL tube masses from numerous samples, and used assumptions and estimates of mass fractions of luminescent substances in CCFL tubes and Y within the phosphor itself, resulting in considerable uncertainty in the final calculated value, something the authors themselves highlight. Disassembly of components from the security dongle was not possible because all internal components, including a primary Li button-cell, LCD display and PCB were encapsulated in resin. The only viable recycling of this item is via comminution which eliminates any opportunity for reuse, and results in CRM dissipation. Table 6 summarises locations of individual CRMs identified across WEEE samples. Overall, the greatest variety of CRMs are found within PCBs, particularly in those which have been miniaturised. Sb is frequently found in plastics as a flame retardant (Sb<sub>2</sub>O<sub>3</sub>), Co in magnets and ferrite cores of magnetrons, Mg-alloy is used for laptop housings, Y is present in the phosphors of LCD CCFL backlights, Ga is consistently found in LEDs, and Nd is present in NIB magnets of HDDs and mobile phone speakers.

#### 3.1.2. CRMs in PCBs and their components

CRMs identified in common PCB components, including SMDs and Au-alloy coated on PCB surfaces and contacts, are summarised in Table 7. Two main kinds of Au contacts are found on PCBs: those coated directly onto PCBs such as edge contacts on PC PCI cards and those in connectors mounted and soldered onto PCBs e.g. HDMI and USB ports (Figure 2). Pd was found in combination with Au on occasion, suggesting Pd is alloyed with Au in the coating. Many PCBs had large areas of their surfaces coated with Au. This surface Au was prevalent in mobile phones and LCD screen PCBs (Figure 3). XRF confirmed the alloy was similar in composition to that used in contacts i.e. Au occasionally alloyed with Pd. Ag is found in solder of samples as Ag-Sn alloy, prevalent in RoHS compliant equipment with Pb-free solder. Chip resistors were the most commonly encountered SMDs and contained Ag, Ru and Sb; with Pd in approximately half of those analysed; and Co in less common larger resistors. Integrated circuits (ICs) were found in all PCBs to contain Au, Ag, often Sb, and occasionally Pd and W - a known interconnect material in ICs which was identified by XRF in ground samples. Multi-layered ceramic capacitors (MLCCs) are common on PCB surfaces in a variety of sizes and colours. CRM content varies and can be distinguished by component colour. Y is present in brown MLCCs, probably as an additive in BaTiO<sub>3</sub> to control formation of 'core shell' microstructure (Kim et al., 2008). ICP-MS of two common sizes of MLCCs revealed Y content of the  $0.55 \times 0.25~\text{mm}$  and  $0.9 \times 0.6~\text{mm}$ components to be 0.3 wt% and 0.8 wt% respectively. Less prevalent, but common in highly miniaturised electronics e.g. phones and MP3 players, were MLCCs containing Nb, which is used as an additive in BaTiO<sub>3</sub> to control microstructure (Kim et al., 2008). Nb is found in combination with: Pd in purple, blue and red MLCCs; Ag in gold coloured MLCCs; and Ir in off-white coloured SMDs (1.8  $\times 1.1$  mm and  $0.9 \times 0.6$  mm sizes). Transistors appear in two forms: standard small outline transistors, and smaller 3-pin small outline transistors. The former, which contain Au, Ag and Pd were found in PCBs of items such as HDDs and TVs, but not in miniaturised PCBs. In minaturised PCBs two types of 3-pin transistors are common. Both contain Au and Ag, with the more common black numbered components containing Sb, and the less common coloured components (pink yellow, green) on server boards containing Ga. Ta-capacitors of numerous sizes and capacitances were found. Typically, 1-3 Ta-capacitors were found per PCBs in miniaturised samples, where high charge storage density is required, the exception being the server board sample which contained 15. To detect Ta with XRF in these plastic encapsulations, components were cut open or ground into powder. The plastic itself contains Sb (probably as Sb<sub>2</sub>O<sub>3</sub> flame retardant), and Ag was detected at component terminals. Quantitative analysis revealed 100 µF 10V 107A Ta-capacitors isolated from samples contained 62  $\pm$  9 wt% Ta, with 3  $\pm$  1 wt% Ag and 1.6  $\pm$  0.5 wt % Sb. Chip arrays containing Ag with either Ru or Pd and Au were common to all PCBs with the exception of power boards. Inductors of three types were found on mobile phone PCBs, and less frequently on other miniaturised PCBs. These were distinguishable by colour, with Pd, Ag and Nb in brown, Ag in grey, and Ag and Co in purple inductors. Polarized moulded body diodes were infrequently found across samples, appearing similar to Ta-capacitors to the naked eye due to the shape of their plastic encapsulation, these were found to contain Sb, probably as Sb<sub>2</sub>O<sub>3</sub>.

With reference to data presented in Table 7, the distribution of CRMs in a TV main PCB (sample 16) was mapped following visual identification of PCB components (Figure 4). This confirms the commonality of CRMs used in PCB components, and the suitability of optical identification of components with prior knowledge of CRM content to map CRMs in PCBs and direct component disassembly for reuse and recovery. Component level CRM data could be used to inform automated image analysis systems capable of identifying components from their dimensions/shape, colour and surface markings in a similar way to the dual-laser desoldering system developed by SAT (Kaya, 2019; Kellner, 2009). Automation of disassembly based on this approach

<b>Table</b> CRMs	5 identified in component	s of samples with XRF ar	nd quantified	l with AAS or ICP-MS.	
#	Sample	Component	CRMs	Location	Comparison with literature
1	Domestic microwave magnetron <sup>a</sup>	Ring magnets (×2) Filter box ferrite cores (×2)	Co Co	22.1 $\pm$ 0.3 mg per magnet; 0.01 wt% in strontium ferrite magnet material 6.24 $\pm$ 0.03 mg (0.5 wt%) in ferrite ceramic	
2	3½ inch Hard DiskDrives (HDDs) <sup>b</sup>	PCB Voice coil accelerator magnets $(\times 2)^c$	Numerous Nd	SMDs; mounted connectors (see Table 7) 5.7 $\pm0.2$ g per magnet, 34.9 $\pm0.9$ wt% in NIB alloy (Nd_2Fe_{14}B); 11.4 $\pm0.3$ g of Nd per HDD <sup>d</sup>	- ~4-5% more Nd in alloy than reported in 2½ inch HDD magnets (30 %) by Buchert <i>et al.</i> , (2012); Nd (22.9 $\pm 2.8$ %), Pr (2.7 $\pm 2.2$ %) and Dy (1.4 $\pm 1.5$ %) in the manets (Ileberschaar and Rotter, 2015).
ŝ	TV Power boards (2	None identified			
4	types) Set top box (STB) front nanel PCBs	SMDs and mounted	Numerous	SMDs; mounted connectors (see Table 7)	
		Blue LEDs	Ga, Ag	Ga in LED e.g. InGaN or GaN; Ag in wiring or solder.	Blue LEDs based typically contain 17-25 $\mu g$ of Ga, 28 ng of In (Gassmann <i>et al.</i> ,
		Green LEDs	Ga, Ag, Au	Ga in LED e.g. GaP; Ag in wiring or solder; Au in connecting wire.	Green LEDs known to use GaP (Marwede <i>et al.</i> , 2012); LEDs often contacted via Au bond wires $\sim 200 \text{ mg per LED}$ (Gassmann <i>et al.</i> , 2016)
Ω.	Laptop Li-ion battery (LIB) packs	PCB Cylinder LIB cells (×6)	Numerous Co	SMDs; mounted connectors (see Table 7) Cathode materials e.g. LiCO <sub>2</sub> , LiCO <sub>1-(x+y)</sub> Mn <sub>x</sub> Ni <sub>y</sub> O <sub>2</sub> , 8.30 $\pm$ 0.01 g per cell (18.94 $\pm$ 0.02 wt% of cell); 49.82 $\pm$ 0.06 g per battery pack (15.8 wt% of LIB hattery neck)	- 65 g per notebook battery pack, 13.8 wt% (Buchert <i>et al.</i> , 2012). Lower mass per battery pack and higher wt% of Co than previously reported
9	Laptop screens <sup>e</sup>	PCB Housing CCFL	Numerous Mg Y <sup>f</sup>	SMDs; mounted connectors, surface gold (see Table 7) SMDs; mounted connectors, surface gold (see Table 7) AZ91D magnesium alloy (90% Mg, 9% Al, 1% Zn) 13.0 $\pm 0.1$ mg in Y <sub>2</sub> O <sub>3</sub> :Eu <sup>3+</sup> phosphor (1.7 wt% of CCFL with connecting wires removed)	- Industry standard alloy composition 1.8 mg of Y, 0.13 mg Eu, 0.11 mg La, 0.076 mg Ce, 0.038 mg Tb and 0.011 mg Gd ner screen (Bucher <i>et al.</i> , 2012)
~	Mobile Phones <sup>e</sup> (assorted)	LIB PCB SD card	Co Numerous Au, Sb	Cathorem meterials e.g. LiCoO <sub>2</sub> , LiCo <sub>1-(x+y)</sub> Mn <sub>x</sub> Ni <sub>y</sub> O <sub>2</sub> ; 4.17 $\pm$ 0.01 g per cell (19.44 $\pm$ 0.07 wt%) SMDs; mounted connectors (see Table 7) SMDs; mounted connectors (see Table 7) Au coated notio surface of Ni-Cu alloy of contacts; Sb in plastic as SbO <sub>3</sub> flame	Equivalent to literature data: 3.8 g Co per 20 g cell (19 wf%) (Buchert <i>et al.</i> , 2012)
		Speaker Magnets LCD screen connector	Nd Numerous	Present in NIB alloy (133 $\pm$ 3 mg/magnet; 27.9 $\pm$ 0.7 wt%) SMDs, coated connectors, surface gold (see Table 7)	31 wt% REMs in magnet alloy with Nd and Pr present in 5:1 ratio (Buchert $etal.,$ 2012)
ø	MP3 players (assorted) <sup>e</sup>	iou LIB PCB (×2)	Co, Li Numerous	Cathode materials e.g. LiCoO <sub>26</sub> LiCo <sub>1-(x+y)</sub> Mn <sub>x</sub> Ni <sub>y</sub> O <sub>25</sub> (2.46 $\pm$ 0.02 g per cell; 20.8 $\pm$ 0.2 wt%) SMDs, coated connectors (see Table 7)	Correlates to packet battery data in literature, see entry for mobile phone LIB above. -
9 10	Security dongle Digital Cameras	None identified LIB	, °C ;	cathode materials e.g. LiCoO <sub>2</sub> , Li	- See entry for mobile phone LIB above
11	(assorted) <sup>e</sup> PCB from TV LCD	PCB PCB	Numerous Numerous	SMDs, mounted connectors (see Table 7) SMDs, mounted connectors (see Table 7)	
12	Graphics card Server board	PCB	Numerous	SMDs, coated connectors (see Table 7) SMDs. coated connectors (see Table 7)	
14	Ethernet card	PCB	Numerous	SMDs, coated connectors (see Table 7)	
15 16	Refrigerator power board Main PCB from TV	None identified PCB	- Numerous	- SMDs, mounted connectors (see Table 7)	
O Z A Z H Ž	o content totals 56 mg $_{\rm I}$ o Pt identified in HDD I r and Dy were undetecte d content ~5 wt% high t undetected by XRF in I o other REMs in phospho	er magnetron latters by XRF d but known to be used $r$ than found in $2y_2$ inch CD screen, 39 mg on a 1 rs identified e.g. Eu and	in the NIB al (HDD magne (4" display re (Gd, levels a	loy (München et al., 2018) ts (Buchert et al., 2012) eported in literature (Buchert et al., 2012) re comparatively low vs. Y and below XRF detection limits (Dupont and B	innemans, 2015).

8

Summary of identified locations of CRMs in WEEE samples

Critical materials	Identified location in samples
Antimony (Sb)	Within plastics & numerous SMDs, probably as $\rm Sb_2O_3$
Cobalt (Co)	LIB cathodes; magnetic ferrite materials in magnetrons;
	SMD chip resistors and inductors.
Gallium (Ga)	LEDs
Magnesium (Mg)	Mg-alloy laptop housings
Niobium (Nb)	Some MLCC and inductor SMDs
Tantalum (Ta)	SMD Ta-capacitors
Tungsten (W)	ICs
Yttrium (Y)	CCFLs phosphors and MLCC SMDs
Neodymium (Nd)	NdFeB (NIB) alloys of HDD magnets and mobile phone
	speakers
Iridium (Ir)	Some MLCCs
Ruthenium (Ru)	Chip resistor and chip array SMDs
Palladium (Pd)	Au-alloys of contacts, solder, numerous SMDs (chip
	resistors, ICs, MLCCs & chip arrays)
Gold (Au)	LEDs, alloy coating on contacts and directly on PCBs,
	numerous SMDs (ICs, transistors, chip arrays)
Silver (Ag)	SMDs: ICs, chip resistors, MLCCs, transistors, Ta-capacitors,
	chip arrays & inductors; Pb-free solder

would negate the requirement for further component separation.

#### 3.1.3. Recovery potential of CRMs in PCBs

Disassembly in pre-processing to isolate PCBs must be conducted as economically as possible to ensure PM recovery cut-off grades remain low. Higher pre-processing costs require higher returns from refineries for economically viable recycling. If PCBs are of insufficient grade to justify the expense of PM recycling, they are smelted for Cu recovery only, resulting in loss of CRMs. If PCB grade is sufficiently high (i.e. above 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. CRMs recovered in these processes are limited to those of highest value i.e. PMs and PGMs. There are economic and thermodynamic limitations to viable recovery of other CRMs due to the way in which metals partition between liquid copper and slag phases during smelting, which ultimately determines 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 (Holland et al., 2018; Klemettinen et al., 2019, 2019, 2017; Reck and Graedel, 2012; Sukhomlinov et al., 2019a,b,c; Wan et al., 2018). Umicore's integrated smelting facility in Hoboken, Belgium, is the most advanced plant of its kind, and, in addition to the normal metals recovered, the CRMs Sb and In are targeted (Hagelüken, 2006). Comparing unrecovered elements with component level CRM data (Table 7) highlights that removal of SMDs containing non-target CRMs prior to smelting is essential to avoid permanent loss. These SMDs include: certain chip resistors and inductors containing Co; MLCCs containing Y or Nb; ICs containing W; transistors containing Ga; Ta-capacitors; and inductors containing either Co or Nb. Many PCBs find their way to this process, although there is fierce competition for waste PCBs from other European and Asian smelters that do not recover In and Sb. The costs of recovery are passed on to pre-processors as mass-based refining charges applied to materials received. To increase the chance of viable CRM recovery from PCBs, cost-effective disassembly of WEEE is necessary, and use of additional 'upgrade' processes which conserve and concentrate CRMs in PCB components would reduce recovery charges and enable CRM recovery from PCBs which otherwise fall below recovery cut-off-grade. Desoldering is important in this regard, enabling CRMs in SMDs to be collected while eliminating the majority of the PCB mass and therefore refining charge. This is also essential to be able to divert components containing non-target CRMs from PCB smelting to appropriate alternative means of recovery when they are developed. Removal of edge contacts and portions of PCBs with Au-coating and/or highmass low-value parts like steel brackets would also improve cost-benefit of recovering CRMs from PCBs.

### 3.2. Product disassembly and CRM recycling potential

Table 8 gives times and additional cost considerations of methods trialled for components isolation from samples, along with general comments on recovery potential of CRMs contained.<sup>4</sup> Due to the similar nature of the digital media device samples (i.e. mobile phones, MP3 players and cameras) disassembly processes for these are considered together. As no CRMs were identified in power boards, these are not considered further in this section. Equipment costs for manual disassembly are low, but it is time intensive. This approach may be viable in the developing world but prohibitively costly elsewhere. More costeffective methods for product disassembly will be essential to avoid preprocessing via comminution and CRM dissipation. Cryocracking enabled rapid isolation of high-grade PCBs and LIBs from digital media devices (Figure 5) and was well suited to items with plastic housings provided there were no major metallic structural features which remained ductile following cooling and impeded access to PCBs following housing removal. 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<sub>2</sub>. 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. In the case of HDDs, this method is unsuitable for rapid isolation of magnets due to their cast aluminium housing. Much research is underway to develop automated NIB magnet recycling processes for rapid access to the magnets at high throughput. Pipeline solutions include an automated system under development by the US Critical Materials Institute (CMI) which snips away the corners of HDDs containing the magnets (King, 2016); and a hydrogen decrepitation process which infiltrates the drives with hydrogen gas causing the magnets to expand and disintegrate, so that NIB alloy powder can be shaken from drives in a rotating drum and collected for re-sintering into new magnets, without disassembly (C. Li et al., 2015; X. Li et al., 2015; X.T. Li et al., 2015; Lixandru et al., 2017; O'Connor et al., 2016; Szymański et al., 2016). The use of a guillotine to open LCD housings by slicing through edges of the bezel was effective at reducing time to isolate internal components, all identified CRM containing parts were isolated in 65 seconds, compared to 180 seconds just to open the housing with manual disassembly. The method is low cost, with PCB guillotines readily available, but CCFL lamps were fractured, releasing Hg, each time this method was trialled on LCD samples. For this reason, environmental monitoring would be required, adding considerable cost to the process, and the method is therefore not recommended for use by the informal sector in the developing world. The use of the wet tile cutter enabled isolation of components intact, but it is more time intensive than manual disassembly, consumes electricity, and produces hazardous dust and slurry requiring costly disposal. For this reason, the method is generally unsuitable for WEEE treatment.

For details of current and future recovery prospects see Table 8. In general, a lack of available markets for component reuse prevents valorisation and retention of contained CRMs in tighter highest value

<sup>&</sup>lt;sup>4</sup> To determine true viability of implementing disassembly methods, a costbenefit analysis based on time in motion studies, capital and consumable costs as well as plant overheads must be conducted to take account of varying costs and employee abilities in individual recycling plants. Material revenues from refineries based on assay must be considered which will vary with quantities of materials sent for refining and market values of metals which show temporal variation. Logistics costs will also be unique to individual facilities depending on distance to refineries and costs of complying with import/export regulations.

Summary of CRMs identified in PCB component

(	Component	PI	Ms		PGMs		REMs			Other (	CRMs		
		Au	Ag	Pd	Ru	Ir	Y	Co	Ga	Nb	Sb	Та	W
Gold contacts	e.g. Figure 2	✓		(√)									
Surface Au	e.g. Figure 3	✓		(✔)									
Solder	Connects all SMDs to PCBs		✓										
Chip resistors	e.g. Table 10		✓	(✔)	✓			(✔)			✓		
Integrated Circuits	e.g. Table 10	✓	✓	(✔)							(✔)		✓
				~			✓			✓			
<u>MLCC</u> s						~				~			
			~							~			
				~						~			
	E obszaz E obszaz	~	~								~		
<u>Transistor</u> s		~	~								~		
		~	~						~		~		
Ta-capacitors	A 107A L 50 (M K8 2)		~								~	~	
Chip arrays	828	(√)	~	(✔)	(√)								
			~					~					
Inductors	HQH		~										
	ad		~	~						~			
Polarized diodes											~		

 $\checkmark$  - consistently present in component;  $(\checkmark)$  - sometimes present in component



Figure 2. Au coated mounted contacts of HDMI ports on a TV PCB (left) and coated edge contacts of a PCI graphics card (right)



Figure 3. Photograph of the coated Au on the surface of a mobile phone PCB

loops of the circular economy. Although manufacturing from recovered components occurs in Asia, the relatively high cost of isolation and logistics may prevent component reuse in parts of the world that are distant from these processes. A distinct lack of CRM recovery processes represents a major barrier to increasing CRM recycling rates from WEEE, except in the case of those previously discussed as recoverable in PCB smelting and those contained in LIBs, although Li is lost in slag and graphite is burnt off in smelting operations which target higher value cathode metals (e.g. Co, Mn) (Gaines, 2014; Tirronen et al., 2017). No commercial process for recycling post-consumer HDD magnets was identified although recycling of production scrap is known if sufficiently large quantities can be accumulated (~300 tonnes, or 184,000

magnets). The feasibility of this recycling route is questionable considering cost and time required to isolate sufficient quantities of magnets for refining, coupled with long delays in payment on material received at refineries, which may create cash flow issues for recyclers. These issues may be partially addressed through adoption of the process for rapid magnet isolation being developed by the US CMI (King, 2016). Although no reuse markets were identified, reuse of HDD magnets may be possible and generate more value than recycling. However, SSD is superseding HDD technology so any reuse market may be short lived. Hydrogen decrepitation could enable collection of the alloy for sintering into magnets for alternative applications such as electric motors and wind turbines, allowing CRMs to be cascaded into lifecvcles of these products to meet growing demand as SSD technology supersedes HDDs. This approach is more closed-loop than traditional recycling, and remanufacturing the magnets in this way could generate greater value, thereby increasing viability of recovery. No recovery of REMs from CCFL phosphors could be identified, although pipeline technologies are in development with the HydroWEEE demonstration plant in Europe at the highest TRL (Dupont and Binnemans, 2015; Favot and Massarutto, 2019; Hu et al., 2017; Innocenzi et al., 2014). Coatings on Mg-alloy laptop housings prevent recycling in general, although an inhouse process has been developed by Fujitsu to de-coat the alloy for recycling (Horikoshi et al., 2003; Kimura et al., 2002; Meskers, 2008; Meskers et al., 2007, 2006). No process for CRM recovery from LEDs exists, with little prospect for future recovery given the low value of the CRMs contained and the presence of a polymer encapsulant which



Figure 4. Colour coded CRM distribution in TV main PCB

to CRM bearing components and considerations of their recovery potential.	CRMs present Disassembly method Cost considerations Comments on recovery potential	Time (s) Other	CoManual120• No specific recovery for ferritic magnetic materials which target Co was identified.• No positive cost benefit can be derived from manual isolation of these components in comparison to treatment of whole magnetron in comparison with treatment of whole magnetrons with other small domestic appliances (SDA) in automated shred and sort processes.• Materials will be collected in ferrous output fractions directed to the collected in ferrous output fractions directed to	Numerous <sup>a</sup> Manual 55 • • PCB is probably suitable grade for recovery in traditional pyrometallurgical	<ul> <li>Manual 185 PM recovery processe.<sup>5</sup></li> <li>Manual isolation of magnets is prohibitively time consuming, but research is underway to automate isolation process (Grag, 2016).</li> <li>Manuel isolation of magnets is prohibitively time consuming, but research is underway to automate isolation process (Grag, 2016).</li> <li>No commercial process for recycling of REMS from HDD magnets has been identified to date, but several technologies are in development (Yang et al., 2017).</li> <li>Industrial recycling of NIB production scrap exists, provided a sufficient quantity, ~ 301 tomes, can be accumulated this may be a viable recovery route, although this equates to ~ 184,000 NIB magnets, requiring disascembly of ~ 92,000 HDDs assuming 2 magnets, requiring disascembly of ~ 92,000 HDDs assuming 2 magnets, requiring disascembly of ~ 92,000 HDDs assuming 2 magnets, requiring disascembly of ~ 92,000 HDDs assuming 2 magnets, requiring disascembly of ~ 92,000 HDDs for re-sintered into new magnets. Closed-loop approach would reduce pre-processing costs and potentially derive greater value from whole HDDs for re-sintered into new magnets. Closed-loop approach would reduce pre-processing costs and potentially derive greater value from whole HDDs for re-sintered into new magnets. Closed-loop approach would reduce pre-processing costs and potentially derive greater value from whole HDDs for re-sintered into new magnets. Closed-loop approach would reduce pre-processing costs and potentially derive greater value from whole HDDs for re-sintered into new magnets.</li> </ul>	<ul> <li>Ga, Ag, Au Manual 20</li> <li>LED recycling is problematic, CRMs are encapeulated within LED and at prohibitively low concentration for economic recovery; no implementation of commercial recycling is likely in the near future (Ueberschaar et al., 2017); no economic benefit derivable from isolation of LEDs.</li> <li>Few LEDs of low mass per item result in high costs of isolation of minimum refinable quantities were a suitable process available.</li> <li>CRMs are lost through unintended co-separation in automated recycling (CPAM) 2013.</li> </ul>	<ul> <li>Co, (Li, Manual 157</li> <li>e. LiB packs are disassembled from laptops as part of the standard recycling procedure to comply with depollution aspects of WEEE legislation (EC, 2012).</li> <li>e. Compliance schemes were found to be the only possible route to recycling for LIBs.</li> <li>The value of the batteries to recyclers varies greatly depending on the type of battery and its Co content (Gaines, 2014); Coi sefficiently recovered with similary. 2017).</li> <li>Potential for refurbishment and reuse of cells in new battery packs which would reduce CRM demand, retaining CRMs in higher value, tighter loops of</li> </ul>	Numerous     291     291     • PCB is of relatively low grade with no surface gold and low density of SMDs on its surface.       • Relatively high disasembly time and associated cost mean that item is probably below cut-off grade for PM recovery in pyrometallurgical
mponents and cons	Disassembly methoo		Manual	Manual	Manual	Manual	Manual	Manual
to CRM bearing co	CRMs present		S	Numerous <sup>a</sup>	РИ	Ga, Ag, Au	Co, (Li, graphite) <sup>g</sup>	Numerous
ıple disassembly in	Component		Magnetic ferrite materials	PCB	NIB magnets	LEDs	LIB cells	PCB
siderations of sam	ıple		nestic microwave gnetron	rd disk drives	(st	top box front el PCBs	top LJB packs	
80 <del>–</del>	an		1ag	a.	E	an	ap	

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# Sample	Component	<b>CRMs</b> present	Disassembly method	Cost considerations		Comments on recovery potential
				Time (s)	Other	
6 Laptop screens	Mg housing	Mg	Manual Wet tile cutter Guillotine <sup>d</sup>	180 141° 65	Electricity -	without alternative cost-effective methods of pre-processing and isolation of components from PCB to concentrate and upgrade CRM fractions. • The recycling of coated Mg-alloy is problematic due to difficulties encountered with complete removal of coatings, and coatings are likely to form dioxins during standard Mg recycling processes. (Meskers, 2008; Meskers et al., 2007, 2006),
						<ul> <li>Problematic to remove all wires, and fixings which contaminate Mg-alloy, decreasing its value to recyclers.</li> <li>Material is problematic for Mg recyclers due to risk of introduction of contamination from post-consumer scrap, only small volumes can be accommodated.</li> <li>The only company to process painted Mg-alloy laptop housings identified is Fujits Group in Japan treating only in-house products and production scrap (Horkoshi et al. 2002).</li> </ul>
	Minor PCB	Numerous <sup>a</sup>	Manual Wet tile cutter Guillotine <sup>d</sup>	224 154 <sup>e</sup> 65	- Electricity -	$\bullet$ PCB is of low grade, given time for isolation, unlikely to be above cut-of-grade for PM recovery.
	Major PCB	Numerous <sup>a</sup>	Manual Warie Wet tile cutter Guillotine <sup>d</sup>	248 154° 65	- Electricity -	<ul> <li>PCB has surface gold although SMD density is low; isolation of PCB sections with surface gold and isolation of SMDs may enable upgrade and concentuation of CRMs into material streams of sufficient grade for PM recovery.<sup>b</sup></li> </ul>
	CCFL backlight	*	Manual Tile cutter <sup>c</sup> Guillotine <sup>d</sup>	390 241 65	Electricity	<ul> <li>No commercially known process for recovery of REMs from CCFLs at time of trial,</li> <li>Several processes in development for recovery of REMs from lamp phosphors (Dupont and Binnemans, 2015; Hu et al., 2017; Innocenzi et al., 2014); HydroWEEE is at highest technology readiness level (TRL) with demonstration plant in operation (Favot and Massarutto, 2019).</li> <li>Presence of Hg poses health &amp; safety and environmental issues (Cryan et al.,</li> </ul>
7 Mobile phone <sup>f</sup>	SD Card	Au, Sb	a) Manual b) Gryocracking	8 8 %	<ul> <li>a) -</li> <li>b) Liq. N<sub>2</sub>, dewar, PPE;</li> <li>O<sub>2</sub> deficiency monitors and ventilation system</li> </ul>	2009; McDonnell and Williams, 2010) complicating and adding cost to recycling, requiring CCFL backlit screens to be processed by specialists. Grade may be suitable for PM recovery via pyrometallurgical routes with other PCBs. <sup>b</sup> • Sb not generally recovered in pyrometallurgical PM recovery (Umicore's process is an exception - Hagelüken, 2006).
	PCB	Numerous <sup>a</sup>	a) Manual b) Cryocracking	87 30 <sup>h</sup>	may be required	<ul> <li>Boards are high-grade and suitable for downstream PM recovery.</li> <li>Manual disassembly is prohibitively costly, greater recovery rates of CRMs from mobile phones may be possible if cryocracking or alternative accouncient with accounter property.</li> </ul>
	LCD screen	hn <sup>8</sup>	a) Manual b) Gryocracking	87 30 <sup>h</sup>		<ul> <li>economical reput usasseminity are used to parater F.Cos.</li> <li>A cross-current leaching method has been developed by Rocchetti et al., which achieves economic recovery of In from LCDs (Rocchetti et al., 2016, 2015; Rocchetti and Beolchini, 2015).</li> <li>SMDs and Au-contacts on the plastic substrates of flexible PCB type</li> </ul>
	Speaker magnet	Nd	a) Manual b) Cryocracking	98 30 <sup>h</sup>		<ul> <li>No current commercial recovery process for these magnets. May be suitable for inclusion with HDD magnets for recycling in emerging processes outlined above.</li> <li>(continued on next page)</li> </ul>

# Sample	Component	CRMs present	Disassembly method	Cost considerations	Comments on recovery potential
				Time (s) Othe	
8 MP3 player	LIB cells	Co, (Li, graphite) <sup>g</sup>	a) Manual b) Cryocracking <sup>i</sup> b) Cryocracking <sup>k</sup>	52 30 <sup>h</sup> 240 <sup>k</sup>	• See previous comment on LIB CRM recovery potential
	Major PCB	Numerous <sup>a</sup>	a) Marual b) Cryocracking b) Cryocracking	62 30 <sup>†</sup> 240 <sup>†</sup>	<ul> <li>PCB is densely populated and relatively high-grade<sup>b</sup>.</li> <li>Device with metallic housing required repeat submersions and hammering resulting in higher costs than manual disassembly, high value ICs detached from PCB resulting in problematic recovery from other debris from the process.</li> <li>Cryocracking to isolate PCBs when no metallic housings or structural components are present may reduce recovery cut-off grade sufficiently for economic PM recovery.</li> </ul>
	LCD screen <sup>k</sup> Minor PCB	ln <sup>8</sup> Numerous <sup>a</sup>	a) Manual b) Cryocracking <sup> </sup> b) Cryocracking <sup> </sup> a) Manual b) Cryocracking <sup> </sup> b) Cryocracking <sup> </sup>	94 30 <sup>1</sup> h 105 30 <sup>1</sup> h 240 <sup>k</sup>	<ul> <li>See previous comment on LCD CRM recovery potential</li> <li>SMD density on PCB is lower than main PCB but may still be above COG for PM recovery if rapid disassembly processes such as cryocracking are utilised.<sup>b</sup></li> </ul>
9 Security dongle	Li cell PCB LCD screen	Li <sup>i</sup> Numerous <sup>a, i</sup> In <sup>i</sup>	Following manual removal of hous for removal of resin encapsulating components.	ing, no method available internal CRM containing	<ul> <li>No pre-processing options other than shredding with associated losses of CRMs.</li> <li>The dongle provides a good example of poor design for recycling as a barrier to any CRM recovery. This highlights the necessity of DfRR with careful consideration of end-of-life during product design to facilitate CRM recovery (Akinade et al., 2017; Ochme et al., 2013).</li> </ul>
10 Digital camera	LIB cell LCD screen	Co, (Li, graphite) <sup>8</sup> In <sup>8</sup>	a) Manual a) Manual b) Cryocracking <sup>i</sup> b) Cryocracking <sup>i</sup>	8 120 30 <sup>m</sup> 60 <sup>m,</sup>	<ul> <li>See previous comment on LIB CRM recovery potential</li> <li>See previous comment on LCD CRM recovery potential</li> <li>Cryocracking enables more rapid isolation of LCD than manual disasembly, although to a lesser extent with metallic housings, thereby reducing CPM recovery of the recovery potential</li> </ul>
	PCB		a) Manual b) Cryocracking <sup> </sup> b) Cryocracking <sup>k</sup>	166 30 <sup>m</sup> >240 <sup>m</sup>	<ul> <li>PCB is high-grade with high SMD density on both sides of PCB, and numerous Au contacts present.<sup>b</sup></li> <li>Cryocracking greatly reduces time to isolate PCB when it is near the front or back surface of the device.</li> <li>PCBs located deep within camera structure behind other components require removal of additional screws and internal structural parts to isolate PCB, increasing disassembly times.</li> </ul>

<sup>b</sup> see previous discussion on recovery potential of CRMs from PCBs;

° wet tile cutter used to cut away bottom edge of screen containing CCFL which fractures posing H&S risk;

<sup>d</sup> guillotine was used to remove top and side edges of housing to access internal components, CCFL fractured in process posing H&S risk, method enables simultaneous isolation of all components;

<sup>e</sup> time is in addition to time to isolate CCFL with this method;

sample received with LIB already removed as part of standard depollution procedure at AATF;

<sup>8</sup> element undetected in this study but known to be present (Amato et al., 2017; Buchert et al., 2012, 2008);

 $^{\rm h}\,$  20 s submersion, 10 s to shatter and isolate components;

<sup>1</sup> Liq N<sub>2</sub> is consumable and treats  $\sim$ 200 items/L at  $\sim$ £2.50/L + delivery charges, storage dewar is a one-off cost of  $\sim$ £3k and O<sub>2</sub> deficiency monitors with forced ventilation systems may be required if siting dewar outside is not possible and sudden venting would decrease  $0_2$  content in room to < 18%;

Sample with plastic housing;

k Sample with aluminium housing;

<sup>1</sup> unknown backlight as LCD sandwiches were not further disassembled, may contain additional REMs in CCFL or Ga in LEDs;

<sup>m</sup> LIBs isolated manually prior to cryocracking, time is in addition to 8s for manual isolation of LIB.

Table 8 (continued)

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Figure 5. Cryocracking of mobile phone for isolation of CRM bearing components

prevents leaching of CRMs.

# 3.3. Methods for the isolation of PCB components

Enhancing recovery rates of CRMs identified in SMDs and PCB contacts may be possible by disassembly, thus concentrating them within lower mass fractions to reduce refining costs and increase recovery efficiency and value derived (Chancerel et al., 2009). Details of methods trialled to achieve this, including times and additional cost considerations and impacts on recovery potential of CRMs in isolated components, are given in Table 9. Difficulty was encountered when attempting to find the correct balance of power and travel rate through the NIR-oven to soften solder without burning PCBs or evolving toxic gases, and further research would be necessary to optimize the operational parameters. It is likely that these setting will vary depending on solder alloy present in each sample, the IR absorbing nature of the PCB substrate, and spatial arrangement and varying specific heat capacities of components. Activated carbon off-gas treatment systems and inert atmospheres have been used by others to limit these issues in thermal desoldering systems. Capital requirement and potential for impacts on human health and the environment may render this method unsuitable for application in the developing world. Like other thermal desoldering processes reported, this method suffers from the inability to remove THDs and mechanically fastened components without further mechanical treatment, although 100% of SMDs are effectively removed. The process is therefore best suited to highly miniaturised PCBs without THDs and mechanically fastened components. Other PCBs such as motherboards with a combination of these component types would be best treated with alternative off-the-shelf desoldering system e.g. the Yuxi Machine which removes all PCB component types, includes off-gas treatment system, and is a promising desoldering solution with potential for global adoption, capital and operational costs permitting. Of the thermal desoldering methods trialled, simultaneous disassembly of SMDs with a solder bath is most promising and cost-effective, enabling recovery of intact components with higher reuse potential without evolution of hazardous gaseous emissions. Further sorting of simultaneously disassembled components according to reuse potential and CRM content would be required. Manual removal may be suitable for selective disassembly of high-value SMDs, although this often causes component damage thus preventing reuse (Figure 6). For Au-coated portions of PCBs, including edge contacts, isolation from whole PCBs was achieved rapidly with a guillotine and wet tile cutter in comparable times. However, previously discussed issues with the tile cutter renders it less suitable for this application, with a guillotine proving to be an effective low-cost option which is transferable to the developing world and which avoids consumption of electricity and generation of hazardous dust/slurry. The single advantage of the tile cutter is its ability to more intricately cut around high value parts of PCBs compared with

the guillotine.

#### 3.4. Methods used to concentrate CRM content in the isolated components

Microwave ashing of coated Au contacts achieved mass reductions of ~19-51%, depending on the nature of the sample. The ash of contacts from: TV boards, graphics cards, server boards and ethernet cards were found to have 81%, 67%% 56% and 49% of the original sample masses respectively, with all CRMs retained, showing the process is effective at upgrading materials for recovery without CRM dissipation. Smelters typically accept minimum loads of 500 kg for refining, which would require ashing of >285,000 graphics card edge connectors for example, requiring considerable expenditure before a return on processed material is received. This may present cash-flow issues for smaller recyclers. In addition, the ashing process generates toxic gases through thermal treatment of PCBs at 800 °C in the presence of air. Off-gas treatment is therefore required, adding to the cost of industrial implementation. Although a superior option to open burning and incineration employed in the developing world, alternative thermal approaches such as pyrolysis or gasification which valorise non-metallic portions of PCBs and concentrate CRMs by thermal treatment in the absence of air/oxygen may be superior to ashing from a resource efficiency and environmental health and safety point of view. Capital requirements may prevent adoption of these processes in the developing world, so combining activated-carbon off-gas treatment systems with microwave furnaces may be a good compromise in terms of capital requirements and minimum viable scale of implementation compared to expensive pyrolysis and gasification systems.

The results of sieving simultaneously disassembled PCB components are shown in Table 10. CRMs were found to concentrate according to size of fractions e.g. Ta-capacitors are isolated in the 9.52-2.80 mm fraction, and W in ICs concentrates in the >9.52 mm fraction. Where specific downstream CRM recovery options are available for these the Ta-capacitor recovery proposed components, e.g. hv Ueberschaar et al., 2017, sieving seems a promising low capital method to separate components according to CRM content. This supports findings of Lee et al. (2012) showing sieving to be an effective first step in separation of simultaneously disassembled THD PCB components. Sb is spread over the 9.52-2.80 mm, 2.80-1.68 mm and <1.40 mm fractions in transistors, chip resistors and Ta-capacitors, the latter two component types being plastic encapsulations. As suggested by Lee et al. (2012), magnetic and density separation of these fractions would be a good approach to further segregate CRMs; for example, removal of the plastic encapsulated components would concentrate Sb into distinct output fractions for recovery. This separation strategy was also used by Axion in the 'CRM Recovery' trials (WRAP, 2018). No segregation of PMs and PGMs into distinct fractions is possible as these CRMs occur in all components. Emerging recovery processes for all SMDs must target

Table 9 Cost considerations o	f processes for isolation	of CRM containing co	mponent from PCBs	and CRM	recovery potential from compor	nents.
PCB	Isolated components	<b>CRM</b> content	Isolation method		Cost considerations	Comments on process potential to increase CRM recovery rates <sup>a</sup>
				Time (s)	Other	
Mobile phone	Mounted contacts (Figure 6)	Au (3.82 mg; ~1,300 ppm)	Manual – pliers	60		<ul> <li>Method is inexpensive and suitable for rapid isolation of high-grade mounted contacts from PCBs; suitable to isolate contacts from PCBs which fall below cut-of grade for PM recovery. Commonent damage is common eliminating notential for reuse.</li> </ul>
TV-LCD PCB	Coated edge contacts	Au (1.16 mg; ~340 ppm)	a) Manual –guillotine b) Manual – tile cutter	8 8	<ul> <li>a) Periodic blade replacement / sharpening</li> <li>b) Electricity, slurry disposal, blade replacement</li> </ul>	<ul> <li>Both methods are suitable for rapid isolation of CRM containing contacts from PCBs below cut-off grades for PM recovery.</li> <li>Isolation by both methods is possible in equal time with significant time saving vs manual isolation.</li> </ul>
Ethernet card	Coated edge contacts	Au (4.11 mg; $\sim$ 2,400 ppm)	a) Manual -guillotine b) Manual - tile cutter	4 4	-	<ul> <li>PCB guillotines are available and inexpensive; potential for automation exists, potentially using image recognition to align Au coated parts with cutting blades.</li> <li>Tile cutters incur higher initial equipment costs and ongoing electricity costs, but more suitable for intricate cuting and thicker boards (&gt;2 mm).</li> </ul>
Server board	Coated edge contacts	Au (22.2 mg; ~7,100 ppm)	a) Manual -guillotine b) Manual - tile cutter	6		<ul> <li>More H&amp;S issues with wet tile cutter i.e. noise, generation of slurry containing toxic PCB dust.</li> <li>On a commercial scale, areas identified as containing concentrations of CRMs could be cut away from the rest of the PCB using a guillotine, wet circular saw, laser, snips, a punch or a water ref.</li> </ul>
Graphics card	Coated edge contacts	Au (2.95 mg; ~1,700)	a) Manual -guillotine b) Manual - tile cutter	4 4		ς.
MP3 player main PCB	SMDs	Au (~1,600 ppm) Ag (~2,900 ppm) Pd (~110 ppm)	a) Manual <sup>b</sup> Thermal methods: b) blow torch c) paint stripper d) solder bath	a) 240 b) 36 c) 60 d) 15	a) . <sup>c</sup> b) Gas (butane) c) Electricity d) Electricity and equipment cost	a) Manual isolation for of all SMDs in place on PCBs is time intensive and prohibitively expensive, may be suitable for 'cherry picking' of individual components identified as containing CRMs not targeted in PM recovery (e.g. Ta capacitors; Table 7). So by low torch method has low cost but is a time-consuming process at small scale requiring operators to manually scrape components from PCBs once solder softens, issues may be overcome through automation and scale-uo. Additional H8S issues occur if boards begin to
LCD main PCB	SMDs	Au (~1,150 ppm) Ag (~2,900 ppm) Pd (~140 ppm)	a) Manual <sup>b</sup> <u>Thermal methods:</u> b) blow torch c) paint stripper d) solder bath	a) 600 b) 60 c) 120 d) 15		burn or evolve toxic assess in the process. c) 2 kW heat gun took 15-30 s to soften solder, temperature of heat gun requires careful control to avoid PCB burning SMDs and PCBs liberating hazardous emissions posing a H&S issue; high tunning costs compared to other thermal methods, and the process was time intensive. d) Solder Path – When PCBs were placed on the surface of molten solder, solder thermite. Method to DCRs softened sufficiently for remain commont removal in 10-20 s without hurning.
Mobile phone	SMDs	Au (~1,460 ppm) Ag (~1,310 ppm) Pd (~660 ppm)	<u>Thermal methods:</u> b) blow torch c) paint stripper d) solder bath	b)120 c) 180 d) 20		with the liquid solder bast so the composition of SMDs on the underside of PCBs in contact with the liquid solder surface remained in place until removed from the solder bath. Those that did detach remained floating on the surface of the molten solder and were easily sieved out again; solder bath system could be scaled up for industrial use (many larger sizes available) with the arear numbers for commonents isolated simultaneously to reduce operational costs: todaw
Camera	SMDs	Au (~470 ppm) Ag (~800 ppm) Pd (~40 ppm)	<u>Thermal methods:</u> b) blow torch c) paint stripper d) solder bath	b) 266 c) 300 d) 20		method is common on manual recycling lines in China.
<sup>a</sup> recovery potentii <sup>b</sup> standard tools us <sup>c</sup> method abandon	al is based on feedback f ed to pull or scrape SMI ed prior to trials on mob	ollowing submission o Ds from PCBs; vile phone and camera	of isolated CRM cont PCBs	aining con	aponents from PCBs to a PM ref	inery for assessment by fire assay;



Figure 6. Isolation of contacts from a mobile phone PCB and housing

these high value metals along with additional CRMs present to be economically competitive with established pyrometallurgical recovery options and present viable options to increase CRM recovery rates.

# 3.5. Practical applications of findings

Data on component level distribution of CRMs in WEEE can inform development of next generation recycling systems to minimise CRM dissipation and maximise recovery efficiencies for all CRMs present. Data presented on specific occurrences of CRMs in PCBs can inform disassembly approaches to concentrate CRMs in pre-processing outputs and improve economic and technical viability of recovery. In the developed world this may be through automation of disassembly based on image processing, and in the developing world this may be from manual disassembly and appropriate sorting of components. The potential of greater revenue from pre-processing and the low capital requirement for solder baths and sieves may encourage PCB disassembly in developing nations for sale of components to advanced CRM recovery in the

# Table 10

SMDs and CRMs contained in sieved fractions

developed world in favour of hazardous, polluting, backyard recycling operations prevalent in these regions. This could increase global CRM recovery rates from WEEE via a 'best-of-two-worlds' approach (Manhart, 2011). Application of upgrade and concentration processes evaluated here would reduce pre-processing costs and refining charges, thereby increasing economic value of pre-processing output fractions. Alternative recovery processes may be developed which target all CRMs contained in pre-processing outputs based on component level CRM data presented here. This would help minimise current losses in pyrometallurgical recovery (e.g. loss of W in ICs). Methods proposed also represent a means of upgrading CRMs in pre-processing outputs to exceed recovery cut-off-grades, e.g. cryocracking for phone disassembly to recover CRMs from speaker magnets, screens and PCBs which today are dissipated in comminution due to poor product design for disassembly; and PCB desoldering to concentrate CRMs without losses to yield greater returns from refineries. In these ways, the data presented here can hopefully inform interface optimisation between stages of recycling to increase CRM recovery from WEEE.

Fraction (mm)	SMDs			CRMs contained
>9.52	Litelesny Street Street	Me	mory ICs:	Au, Ag, Pd, W
9.52-2.80		a. b. c. d.	ICs: Ta-capacitors: Small outline ICs: Transistors:	Au Pd Ag Ag, Sb, Ta Au, Ag, Pd Au, Ag, Sb
2.80-1.68		а. b. c.	Chip resistors: MLCCs: Transistors:	Ag, Pd, Ru, Co, Sb Ag, Pd, Ir, Y, Nb Au, Ag, Sb
1.68-1.40	正常報	Ch	ip Arrays	Au, Ag, Pd, Ru
<1.40	No la la	a. b.	Chip resistors MLCCs	Ag, Pd, Ru, Co, Sb Ag, Pd, Ir, Y, Nb

#### 3.6. Policy recommendations

Increasing CRM recovery has been a strategic priority of governments for a decade, yet policy intervention has done little to increase recovery rates. Extended producer responsibility (EPR) should incentivise design for EoL by reducing EoL costs for products and compliance fees for original equipment manufacturers (OEMs). Where collective producer responsibility is implemented, the economic incentive for individual organisations to improve design is diluted compared with individual producer responsibility (IPR) systems. One solution is to implement modulated compliance fees which reflect product performance in EoL treatment against those with best practice design for reuse and recycling. A lack of recovery infrastructure for CRMs is a major barrier to recovery. This can be addressed by aligning industrial, economic and environmental strategies of governments. Allocating funding for collaboration, knowledge exchange and R&I to develop required infrastructure would help to address this. Appropriate regulatory frameworks to foster viable secondary materials markets would also create an additional incentive to recover CRMs from WEEE. Price volatility creates risk to investment in any pipeline technologies to address infrastructure gaps. The introduction of national waste tracking systems such as the 'Smart Wastes' system being developed by DEFRA for the UK would enable rapid digital prospecting of the 'urban mine' to generate data which de-risks investment in recovery infrastructure, such as rate of WEEE generation, total available quantities, distribution of material for processing etc. Statutory mass-based reuse and recycling targets have increase WEEE recycling, however they do nothing to incentivise CRM recovery. The value of CRMs contained are low compared with that of PMs, PGMs and Cu, which are the only metals PCB refiners pay returns on, regardless of any additional metals recovered. For these reasons no economic or legislative incentive to increase CRM recovery exists, an issue which must be addressed through adoption of CRM specific/environmentally weighted statutory reuse and recycling targets. Suitable metrics for statutory targets and product evaluation for determination of modulated EPR compliance fees have been proposed by others e.g. Horta Arduin et al., 2020; Huisman, 2003, Huisman et al., 2003. Incentives to stimulate use of secondary components and CRMs in manufacturing would stimulate secondary materials markets, a lack of which hampers recovery. WEEE regulations mandate the provision of product information to recyclers to inform appropriate treatment. Amending such regulations to include the provision of data on the occurrence and content of CRMs in EEE would help to guide recycling and reuse. Additionally, storage of this information in a system similar to the UK national materials datahub would allow rapid access to the data for those able to make best use of it. Linking of waste tracking systems to compositional data for products would enable CRM flows to be mapped, and help regulatory bodies track non-compliance with regulations, preventing illegal exports, fly-tipping and inappropriate treatment. Regulations should also make clear the specific responsibilities of actors in the WEEE value chain with regards to ensuring CRM recovery. 'Right to repair' measures introduced recently by the EU in amendments to the Ecodesign directive should be expanded to include a greater range of products, particularly those items found to be particularly CRMs rich i.e. phones, computers etc (EC, 2009). This would create statutory requirements for appropriate product design for circular economy and provision of information on how to repair/recycle products with spare parts. Requirements to report environmental performance of products which take account of whole product lifecycles and materials criticality metrics would allow consumers to make more informed choices when purchasing EEE. Standards should be developed for assessment to prevent 'green-washing' and ensure a fair basis for product comparison, and these should have a materials criticality component. Appropriate product labelling to empower consumers to make informed purchasing decision based on product performance in this regard would also encourage greater uptake of appropriate measures by OEMs and facilitate wider consumer awareness of this crucial issue.

## 4. Conclusions

Poor product design for EoL and a lack of suitable recovery infrastructure represents a major barrier to increasing recycling rates of most CRMs from all samples investigated, including: ferritic materials in magnetrons, HDD magnets, CCFL phosphors, LEDs, Mg laptop housings, Sb in flame retarded plastics and CRMs identified in PCBs; the exception being the recycling of precious and platinum group metals which are targeted in pyrometallurgical treatment. To improve prospects for eventual recovery, cryocracking is a promising method for rapid disassembly of items with plastic housings such as mobile phones; guillotines can be used to rapidly open LCD housings, and crop CRM rich or low value parts from PCBs; and solder bath treatment is useful for disassembly of PCBs with subsequent sieving to segregate simultaneously disassembled components according to reuse potential and/or CRM content for downstream CRM recovery. Each of these processes requires relatively low capital investment and is potentially suited to the developing world. Optical identification of components for correlation with databases of CRM content is a powerful method to map CRMs in PCB components and direct PCB disassembly to generate suitable output fractions for recovery of specific CRMs as new recovery processes emerge. This may be achieved in the developed world via automated digital image processing to direct disassembly. Emerging recovery processes for CRMs in PCBs must target PMs and PGMs as well as other CRMs to be competitive with existing pyrometallurgical recovery.

# CRediT authorship contribution statement

Rhys G. Charles: Conceptualization, Methodology, Investigation, Writing - original draft, Visualization. Peter Douglas: Conceptualization, Methodology, Writing - review & editing, Visualization, Supervision. Mark Dowling: Conceptualization, Methodology, Investigation, Supervision, Funding acquisition. Gareth Liversage: Resources, Supervision, Funding acquisition. Matthew L. Davies: Writing - review & editing, Funding acquisition.

#### **Declaration of Competing Interest**

None.

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