Review Article

wm&r

Waste Management & Research
31(10) Supplement 3–16
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DOI: 10.1177/0734242X13499814
wmr.sagepub.com
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Data availability and the need for research to localize, quantify and recycle critical metals in information technology, telecommunication and consumer equipment

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Abstract

The supply of critical metals like gallium, germanium, indium and rare earths elements (REE) is of technological, economic and strategic relevance in the manufacturing of electrical and electronic equipment (EEE). Recycling is one of the key strategies to secure the long-term supply of these metals. The dissipation of the metals related to the low concentrations in the products and to the configuration of the life cycle (short use time, insufficient collection, treatment focusing on the recovery of other materials) creates challenges to achieve efficient recycling. This article assesses the available data and sets priorities for further research aimed at developing solutions to improve the recycling of seven critical metals or metal families (antimony, cobalt, gallium, germanium, indium, REE and tantalum). Twenty-six metal applications were identified for those six metals and the REE family. The criteria used for the assessment are (i) the metal criticality related to strategic and economic issues; (ii) the share of the worldwide mine or refinery production going to EEE manufacturing; (iii) rough estimates of the concentration and the content of the metals in the products; (iv) the accuracy of the data already available; and (v) the occurrence of the application in specific WEEE groups. Eight applications were classified as relevant for further research, including the use of antimony as a flame retardant, gallium and germanium in integrated circuits, rare earths in phosphors and permanent magnets, cobalt in batteries, tantalum capacitors and indium as an indium–tin-oxide transparent conductive layer in flat displays.

Keywords

Critical metals, recycling, resources, WEEE, electronic waste, E-waste

Introduction

Improving the efficiency of the use of critical metals is an objective of the European Union owing to their importance in industry, price fluctuations and supply risks. Recycling was recommended to the European Commission as a strategy to reduce and alleviate the supply risks facing critical raw materials (COM, 2010a). UNEP (2011) reported that the recycling rates for critical metals like gallium, indium, germanium and rare earth elements (REE) are presently < 1% owing to product designs that make material separation difficult, the high mobility of products, unclear material flows, low awareness and the lack of an appropriate recycling infrastructure that can keep pace with complex products, such as electronics.

The electronics industry strongly depends on critical metals; for example, about 71% of the gallium consumed in the USA was used for integrated circuits (ICs); the other 29% was used in optoelectronic devices (USGS, 2013). Around 74% of the produced indium is used in liquid crystal displays (LCD) (COM, 2010b). Waste electric and electronic equipment (WEEE) is a potential source of secondary materials. This publication focuses on obsolete information technology (IT), telecommunication and consumer equipment, which contain the most complex electronic systems and, therefore, most of the critical metals used in electric and electronic equipment (EEE) manufacturing. The seven metals, or metal families, antimony, cobalt, gallium, germanium, indium, REE and tantalum were selected for this research owing to their criticality, their relevance for technologies used in EEE and the very low recovery rates achieved by the current recycling infrastructure.

Analysis of the literature dealing with critical metals in WEEE shows that the scientific community has so far focused either on specific metals or on selected equipment types. Oguchi

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et al. (2011) report the results of measurements of the concentration of tin, silver, gold, palladium, cobalt, gallium, tantalum and other metals in printed circuit boards (PCBs) of 21 different equipment types, including televisions, computers, mobile phones and digital cameras. Sander et al. (2012) measured the concentrations of indium, tin, silver, gold, tantalum, gallium, yttrium, europium, lanthanum, neodymium and cerium in 30 devices belonging to six different equipment types, for which the data availability is especially poor (tablet computers, video projectors, multi-function printers, navigation devices, smartphones and MP3 players). Buchert et al. (2012) combined literature data to estimate the content of cobalt, gallium, germanium, indium, platinum-group metals, REE and tantalum in flat displays, notebook computers, smartphone and light-emitting diode (LED) lighting products. Other publications, focusing on specific equipment types, investigated the content of critical metals of mobile phones (Huisman, 2004), mobile phones and computers (Hagelüken and Buchert, 2008), personal computers (PCs) (MCC, 1996) and LED-containing equipment (Deubzer et al., 2012). Further research focuses on product parts, like electronic components (Wichmann et al., 2002) or indium in flat displays (Götze and Rotter, 2012).

So far, the literature does not provide us with sufficient information on the holistic system for further research, for example to estimate the potential of WEEE as a source of raw materials. The data gaps concern all levels of detail, from the analysis of the metal content in relevant assemblies and components, to batch analyses of critical metals in complex equipment mixes received for treatment by the recycling industry.

Beyond the inventory of critical metals, Wang and Gaustad (2012) presented an approach for prioritizing material recovery for end-of-life PCBs. They applied a 'weighted sum method' to evaluate the economic relevance (price criterion), the potential energy savings through recycling (primary production energy – secondary production energy) and the toxic potential of 21 metals, among which were tantalum, gallium, antimony, various precious metals and hazardous substances like lead, cadmium and mercury. The authors concluded that further research needs to be done to obtain 'secondary production' data for uncommonly recycled metals. Their evaluation did not include aspects of multi-element recycling, as practised in modern smelters. Thermodynamic barriers to be considered for the recycling effectiveness, when evaluating multi-element recycling, were introduced, in particular, by the International Resource Panel/ Reuter et al. (in press).

This article systematically assesses the available data to set priorities for further research aimed at developing solutions to enhance the recycling rates of seven critical metals. The specific objectives of the research are to:

1. Develop a methodology to define priorities for further research
2. List and localize the uses of the critical metals in IT, telecommunication and consumer equipment in an exhaustive way, as well as to collect and analyze the available quantitative data on the content of critical metals in the considered EEE
3. Define the metals and applications for which the needs for further research on recycling of the selected metals are high, as well as a research frame for developing separation technologies beyond the-state-of-the-art recovery processes.

Materials and methods

The publication aims at defining 'needs for research'. The term is related to other expressions like 'setting priorities', 'development of an action plan' and 'review of challenges', which have been used in other publications. The methods applied to define needs for research are based on an assessment of the status quo, for example by conducting surveys among stakeholders (Davis and Herat, 2010), by reporting the practical experience gathered in a domain and comparing it with expectations defined by the framework (Friege, 2012), or by assessing the available data (Herat and Agamuthu, 2012).

The first step of this research was to conduct a review of applications using critical metals in IT, telecommunication and consumer equipment using technological understanding based on expert knowledge and literature data. According to the keys published by Wang et al. (2012), 124 product groups belong to IT, telecommunication and consumer equipment. Generally, an 'application' is, according to the Cambridge Dictionary (2013), " … a way in which something can be used for a purpose …". Here, we define a 'metal application' as a technological use of a metal fulfilling specific functions that enable the products to comply with the expectations of manufacturers and consumers. The classification according to 'metal application' was preferred to a classification according to product parts or assemblies (e.g. PCB, casing) because one application can be implemented in different kinds of assemblies. In addition, an 'application' usually shows a typical physical macro- and micro-structure relevant for metal separation technologies, while a 'product part' or 'assembly' may vary in its physical structure. Only the applications established on the market were considered, not the applications under research and development.

For the definition of priorities for further research, five quantitative and qualitative criteria were selected and used as indicators:

1. Metal criticality related to strategic and economic issues
2. Worldwide mine or refinery production for the reference year 2012 and share of the produced metal going to EEE manufacturing
3. Rough estimates of the concentration and content of the metals in the products
4. Accuracy of the data on concentration and content already available
5. Occurrence of the application in specific WEEE groups.

The results of the evaluation based on the aforementioned criteria reflect the complexity of the considered issues. The indicators deal with several dimensions of the problematic and were considered as not comparable because of their diverse information content and units. For this reason, they were not aggregated through weighting into a single score indicator or with clear thresholds. The assessment of the overall priority of an application regarding further research was based on an equal consideration of the five indicators. Only if one of the five indicators for an application tends toward 'zero' or 'not relevant', it means that the need for further research is very low. The objective of further research is to gather sufficiently quantitative information for a prioritization of recovery strategies.

Criticality of the metal

Erdmann and Graedel (2011) provided a survey of the available literature, reviewed the methodological approaches and analyzed the results of studies designated to the criticality of non-fuel minerals. They distinguished two dimensions of the concept of raw material criticality: the supply risks (potential physical interruptions in the supply chain, market imbalances and governmental interventions) and the vulnerability to a potential supply disruption (physical shortages, increasing prices). Erdmann and Graedel (2011) showed, for example, that rare earth elements are frequently singled out as critical, albeit by differing criteria. A meta-study focusing on the selected metals was conducted, considering six studies, which were mostly European studies except for one US study (COM, 2010a; Erdmann et al., 2011; IW Consult GmbH, 2011; Morley and Eatherley, 2008; NRC, 2008; Rückschloss, 2010). The studies used different sets of indicators to assess the criticality of the metals. All studies considered the supply risk as an indicator for criticality. Other indicators, such as environmental impacts, quantitative data on regional or global metal demand, and substitutability of the metals were used in some of the studies. In the meta-study, we summarized the results by classifying if the criticality of the metals was regarded as 'high', 'medium' or 'low' in the six criticality assessments considered. The criticality indicates how high supply risks are and, therefore, how relevant recycling is as a potential strategy to reduce them.

Metal production in 2012 and share of produced metal going to sectors related to EEE

The production of a metal in terms of mine or refinery production (refinery production for metals that are by-products of primarily mined other metals) in 2012 serves as an indicator to estimate the relevance of the metal content in the electronic products. The share of produced metal going to sectors related to EEE shows how relevant the use of that metal in EEE is compared to uses competing with EEE for the supply. The used data are mainly provided by the estimations conducted by the United States Geological Survey (USGS, 2013) and should be considered as indicative.

Rough estimate of metal concentration and content

Owing to the lack of quantitative information on the concentration and content of the metals in the products or product parts, the data from the literature were summarized by indicating orders of magnitude (ranges). Two indicators were considered:

1. The 'concentration' of a metal in a product part (unit, e.g., $mg.kg^{-1}$) is a relevant indicator for the technical feasibility of metal recovery in terms of concentrating through pre-processing and metal refining, and correlates with the energy demand necessary for recycling
2. The 'content' (unit, e.g., mg) indicates the effort required to accumulate a sufficient amount of a metal for recycling through collection and selective treatment.

This evaluation was done per application. The link between concentration and content is the weight of the considered products or product parts (upper part of Figure 1). The content requires knowledge about the weight of the considered products or product parts. Data on the average weight of product and product parts were published by Huisman et al. (2007) and Chancerel and Rotter (2009).

Accuracy of the available data

The literature was first checked to determine whether quantitative data on metal use in EEE were available at all. In case data were available, the methods used to get the data were questioned to determine whether the data were gathered through theoretical estimation or through experimental measurement. Some authors published detailed information on the methods used to take samples, prepare them and conduct the chemical analysis (Jalalpoor et al., in press; Oguchi et al., 2011; Sander et al., 2012); others did not.

The information on data accuracy reveals if further research is needed to gather reliable knowledge on the location of the metals, the amount of metal used and the potential for recycling. Further research is needed in case no, or few, data are published, if methods were not sufficiently documented and if only theoretical data were available.

Occurrence of the application in WEEE

The 'occurrence' is defined as the presence of a metal application across the different WEEE equipment types. The classification of equipment types according to the keys defined and published by Wang et al. (2012) were used.

The occurrence indicates if the metal application is only used in a limited amount of products, or if a broad set of products of IT, telecommunication and consumer equipment contains the application.

The 'physical potential' (unit: $kg.a^{-1}$) can be calculated if the occurrence, i.e. the numbers of units in the waste stream, and the

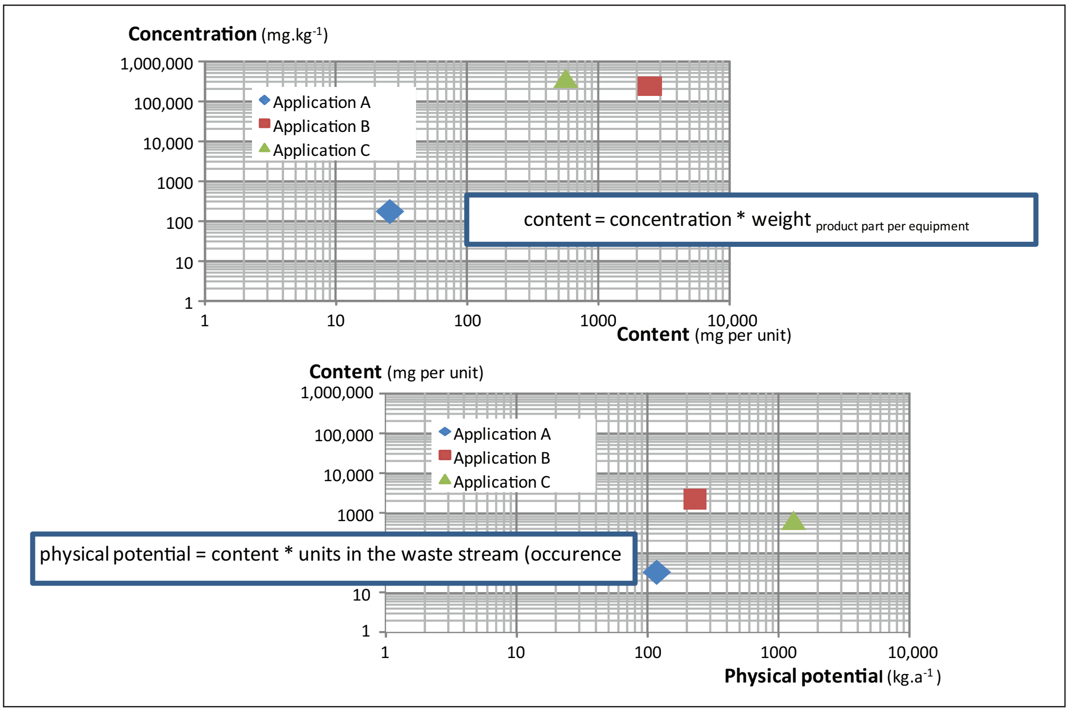

Figure 1. Link between concentration, content and occurrence.

content are known (Figure 1). The ‘physical potential’ indicates the macro-economic potential for recycling beyond a technical feasibility. Regarding the unconsolidated data basis about the quantitative occurrence of these metal applications in WEEE, it was not possible to quantify the ‘physical potential’.

Moreover, the spread of occurrences is an indicator for the diffusion of the metal across different products. A low spread of occurrence facilitates the selective collection of relevant EEE groups; a wide spread of occurrence requires the processing of large volumes of WEEE for recovering a metal used in a particular application.

Results and discussion

Applications in EEE

In this section we present an overview of the applications using the seven selected metals.

Antimony. Antimony, in the form of antimony oxide, is primarily used as metal oxide synergist to enhance the efficiency of flame retardants in plastic casings, PCBs, cable coatings, electrical connectors and other plastic parts. For instance, the polymers polyvinylchloride, acrylonitrile butadiene styrene and high impact polystyrene may be mixed with antimony oxide (European Flame Retardants Association, 2011). The use of antimony in flame retardants has decreased since 2000 (Schlummer et al., 2007). However, it will take time until this will be reflected in the chemical composition of WEEE. Schlummer et al. (2007) showed that the antimony concentration in casings of televisions and PC monitors amounts to 0.2–1.8%, and Huisman et al. (2007) reports, based on a review of literature data, concentration levels of 0.04–0.35% in PCBs of information and communications technology equipment.

Also, packages of capacitors and various electronic components may contain antimony oxide (Angerer et al., 2009; EPCOS, 2012). Solders for microelectronic applications rarely contain antimony, as described in the Restriction of Hazardous Substances Directive-exemption No. 14, which concerns a solder consisting of 82% lead, 10% tin and 8% antimony (Gensch et al., 2009). Moreover, antimony–tin–oxide (ATO) may be used as a transparent conducting electrode to substitute for the commonly used indium–tin–oxide (ITO), which is used as transparent conductive electrode in LCDs. Although the research on ATO began in the 1970s (Peaker, 1971), in 2009 no alternative to ITO could be found in the products (Patel-Predd, 2009). In order to improve the properties of glass of color cathode ray tube (CRT) televisions and monitors, small amounts of antimony trioxide are added (BiPRO, 2006). Méar et al. (2006) report antimony concentrations of 0.09% in color panel glass and 0.12% in color funnel glass.

Cobalt. Cobalt is mainly used in cathodes of secondary batteries for portable devices like mobile phones, smartphones and notebooks. Battery recyclers currently observe a trend towards lower cobalt content in batteries (S. Kross, personal communication). The cobalt concentration was estimated to reach 13–27 % of the battery weight in lithium-ion batteries (Angerer et al., 2009; EPBA, 2007; Hagelüken and Buchert, 2008). In handheld electronics, this battery type usually has a lithium cobalt dioxide ($LiCoO_2$) cathode and a lithium graphite anode. The $LiCoO_2$ powders have a cobalt share of between 18% and 36% (Granata et al., 2012; Mantuano et al., 2006; Petránikova et al., 2011). Cobalt is mainly found in the cathode but, after the use phase, it is also found in lower concentrations in the anode (Dorella and Mansur, 2007). Anode powders have cobalt concentrations of between 0.05% and 3.22%; cathode powders have cobalt

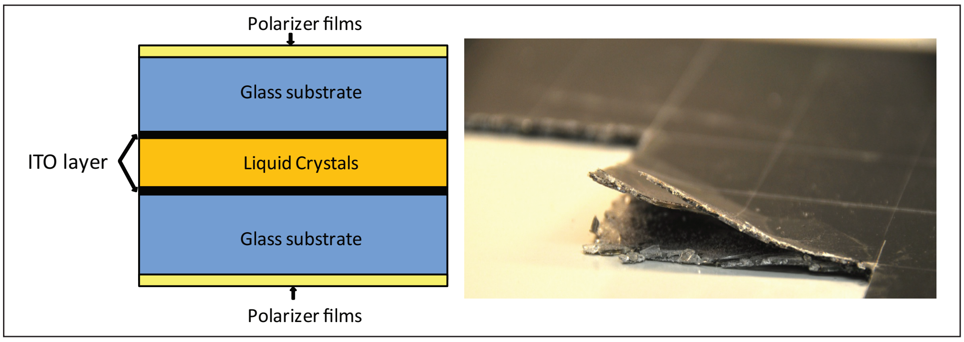

Figure 2. Composition of a liquid crystal display panel (Götze and Rotter, 2012). ITO: indium–tin–oxide.

concentrations of between 43.3 and 47.96% (Dorella and Mansur, 2007; Ferreira et al., 2009).

Nickel–cadmium batteries contain small amounts of cobalt in the nickel hydroxide cathode to enhance the performance (Rydh and Svärd, 2003). The cobalt concentration in the battery was estimated to reach 1–2% (Angerer et al., 2009).

Nickel–metal hydride batteries consist of a cathode plate containing nickel hydroxide as its primary active material and an anode plate. The active material in the anode of nickel metal hydride batteries is hydrogen stored in a metal hydride structure. The most common type of metal hydride alloy (AB_5) refers originally to an alloy made of lanthanum and nickel, which has been altered to an alloy of mixed metals (mainly cerium, lanthanum and neodymium) and a mixture of nickel, cobalt, manganese and aluminum (Kopera, 2004). The cobalt concentration varies between 2% and 4% for the whole battery (Angerer et al., 2009; EPBA, 2007; Nogueira and Margarido, 2012). The cobalt concentration in the anode and cathode powders of an AB_5 battery type range between 4.4% and 6.1% (Innocenzi and Vegliò, 2012; Mantuano et al., 2006).

Cobalt is also used in permanent magnets. The Magnetic Materials Producers Association (MMPA) (Standard No. 0100–00, n.d.) classifies rare earth magnet materials into three families of materials: rare-earth cobalt 5 (l–5 alloys, with the approximate atomic ratio of one rare earth atom to five cobalt atoms), for example samarium cobalt magnets, the rare earth 2 transition metal 17 group (also called 2–17 alloys, which means two rare earth atoms to 13–17 atoms of transition metals) and rare earth iron alloys like $Nd_2Fe_{17}B$. In neodymium magnets cobalt is used as an alloying element to elevate the Curie point of the material (PatentDe, 2005). The MMPA (Standard No. 0100–00) reports cobalt concentrations in permanent magnets of 61–66% in l–5 alloys and 3–15% in rare earth iron alloys. 2 to 17 alloys contain 72–77% of an unspecified cobalt-rich combination of cobalt, iron and copper (Standard No. 0100–00, n.d.). Contacts in ICs can contain up to 15% cobalt (Achzet et al., 2011). Cobalt oxide is used in the active part of electronic components like protection devices (EPCOS, 2012). Oguchi et al. (2011) measured cobalt in PCBs and report concentrations between 8 mg kg^{-1} (radio cassette recorder) and 280 mg kg^{-1} (mobile phone). Traces of cobalt are added to the glass of CRT televisions and monitors to improve the properties of the glass (BiPRO, 2006).

Gallium. About 71% of the gallium consumed in the USA is used in ICs and the other 29% in optoelectronic devices, which include laser diodes, LEDs, photodetectors and solar cells (USGS, 2013).

For instance in high-frequency applications like mobile phones and wireless local area networks hardware, gallium arsenide is used as semiconductor material in ICs. The gallium concentration in the gallium–arsenide semiconductor material amounts to around 50% by weight, but no data are available on the quantity of gallium arsenide used to produce the ICs. Gallium concentrations in PCBs varying from 2 mg kg^{-1} in a tablet and 140 mg kg^{-1} in mobile phones were measured (Oguchi et al., 2011; Sander et al., 2012), showing the presence of gallium in the components.

The semiconductors gallium nitride (GaN) and indium gallium nitride (InGaN) are used to manufacture chips for white LEDs, which are placed in lighting products and backlighting systems for displays of televisions, monitors, mobile phones and other products. Sander et al. (2012) measured gallium concentrations in LEDs between 248 and 690 mg kg^{-1}. Also, laser diodes in DVD, CD and Blu-ray players employ GaN and gallium arsenide (GaAs) (Wochele et al., 2011).

Germanium. Silicon germanium is, like GaAs, employed in ICs of wireless local area network hardware, mobile phones and navigation systems (Angerer et al., 2009). It is also used, combined with magnesium, as phosphor in lamps ($Mg_{28}Ge_{10}O_{48}$ and $Mg_{56}Ge_{15}O_{66}F_{20}$) (Villalba et al., 2012). Quantitative data on germanium in EEE are not available in the literature.

Indium. Most of the produced indium goes to the production of ITO to make transparent conductive thin films for displays, such as liquid crystal, flat panel and plasma. Figure 2 shows the glass sandwich with two ITO layers and the liquid crystal layer in between. The ITO films usually contain 90% In_2O_3 and 10% SnO_2, which corresponds to a mass share of 78% indium (Böni and Widmer, 2011).

Extensive measurements of the indium concentration in displays were conducted by Jalalpoor et al. (in press), showing that the average indium concentration in notebook/PC displays is 175 mg kg^{-1} (SD: 60 mg kg^{-1}) and in mobile phones 320 mg kg^{-1} (with a high SD of 160 mg kg^{-1}). Sander et al. (2012) measured concentrations of 193 mg kg^{-1} in a tablet computer display and 460 mg kg^{-1} in a smartphone display. These experimental results differ strongly from the theoretical estimations made by Angerer et al. (2009), who reported an average concentration of 1000 mg kg^{-1}. ITO may be used in digital cameras as gate electrodes in light sensors using charge-coupled device technologies (Roper Scientific Inc., 1999).

Indium is also contained in the semiconductor materials InGaN, aluminum gallium indium phosphide and aluminum gallium indium nitride, which are used in LEDs. Measurements conducted in white LEDs by Sander et al. (2012) to quantify the indium concentration delivered results below the detection limits of 50 and 200 mg kg^{-1}. Estimations of Deubzer et al. (2012) report an indium content of 0.11 µg per mm² LED die and a die area in products varying between 0.22 (mobile phone) and 77 mm² (direct-lit television). In some special applications, indium may be contained in solders (Gensch et al., 2009; Reitlinger, 2001).

REE. The REEs yttrium, cerium, lanthanum, europium, terbium, dysprosium, lutetium and gadolinium are used to produce phosphors for cold-cathode fluorescent lamps (CCFL), plasma displays, LEDs (lighting products and display backlighting) and organic light-emitting diode displays (Angerer et al., 2009; Buchert et al., 2012; EPIC, 2010). Estimations of the REE content of CCFL and LEDs were published by Buchert et al. (2012) and Deubzer et al. (2012). The REE content in LED products depends on interconnection technology and the phosphor material [yttrium aluminium garnet (YAG) ($Y_3Al_5O_{12}$:Ce) and terbium aluminium garnet ($Tb_3Al_5O_{12}$:Ce) or europium-doped orthosilicate] (Deubzer et al., 2012). Sander et al. (2012) analyzed a CCFL from a tablet computer and white LEDs from various equipment types. Concentrations of 4386 mg kg^{-1} yttrium and 287 mg kg^{-1} europium were determined for the CCFL. The concentration of lanthanum was under the detection limit. Buchert et al. (2012) estimated that the CCFL of a notebook computer contains 1.8 mg yttrium, and between 0.01 and 0.13 mg of europium, lanthanum, cerium, terbium and gadolinium. These numbers were rounded up to 110 mg yttrium, around 8 mg europium, 7 mg lanthanum, and < 1 mg cerium, terbium and gadolinium in televisions with 15 CCFL of 4 g each in the backlighting. In LEDs, the concentrations were under the detection limits for yttrium in LEDs of smartphones and players. In LEDs of navigation devices, concentrations of 526 mg kg^{-1} yttrium, 68 mg kg^{-1} europium and 54 mg kg^{-1} cerium were measured (Sander et al., 2012). Buchert et al. (2012) assumed that the garnet phosphor YAG was used, and estimated that the LEDs of a television contain 4.9 mg yttrium, 2.3 mg gadolinium, and between 0 and 0.3 mg of europium, cerium and terbium.

Two magnet systems contain REE:

1. Neodymium iron boron magnets ($Nd_2Fe_{14}B$)—neodymium, with praseodymium, dysprosium and terbium as alloying elements, are key components of permanent magnets, which are required to produce loudspeakers, microphones, motors and some miniaturized components
2. Samarium cobalt magnets ($SmCo_5$ or Sm_2Co_{17}) with smaller amounts of alloying elements, mainly used for applications at higher temperature and thus of less relevance for WEEE.

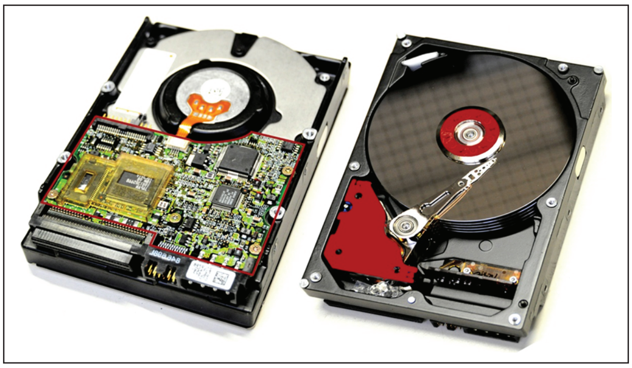

Figure 3. Printed circuit board of a hard disk (left) and opened casing showing the linear and spindle motors (right), which are marked in red (Rotter et al. 2013a).

NdFeB magnets are coated with a nickel, nickel copper or nickel resin plating owing to the corrosiveness of the material. Measurements of separated magnets from hard disk drives delivered a neodymium concentration in the magnets of 23–25 % and concentrations of praseodymium, dysprosium and terbium between 0.01% and 4 % (Rotter et al., 2013a) (Figure 3).

Neodymium concentrations of 8–9% were measured in the loudspeakers of a tablet computer and a smartphone (Sander et al., 2012). Standard No. 0100–00 (n.d.) reports REE concentrations in permanent magnets between 23% and 39%, depending on the family of rare earths magnet materials (l–5 alloys, 2–17 alloys or rare earth iron alloys).

For the cathode material of nickel–metal hydride batteries (metal hydride alloy), hydrogen-absorbing alloys containing REE like lanthanum, cerium and praseodymium in variable shares are used. Increasingly, the REE are substituted by Mischmetall (Mm), which is an unrefined REE mixture (mainly cerium, lanthanum, neodymium and praseodymium) with a composition depending on the ore quality (Berndt and Spahrbier 2001). The overall share of REE in the battery weight varies between 6% and 10% (EPBA, 2007; Müller, 2004; Nogueira and Margarido, 2012). The powders from anode and cathode contain in average around 5.6% lanthanum and 2.6% cerium, as well as neodymium and praseodymium. All REE concentrations are higher in anodes than cathodes (Granata et al., 2012; Innocenzi and Vegliò, 2012).

REE, for example in the form of samarium titanate, can be used in electronic components (EPCOS, 2012). Results of measurements in components or printed circuit boards are not published.

Tantalum. The primary use of tantalum is for miniaturized capacitors, consisting of an anode with a sintered metallic tantalum body covered with a tantalum oxide layer or a tantalum foil as dielectric, a cathode with solid or liquid electrolyte and an epoxy resin packaging (Figure 4).

A tantalum concentration in tantalum capacitors between 24.4% and 42.6% (average: 36.7%) was reported in 2003 by the

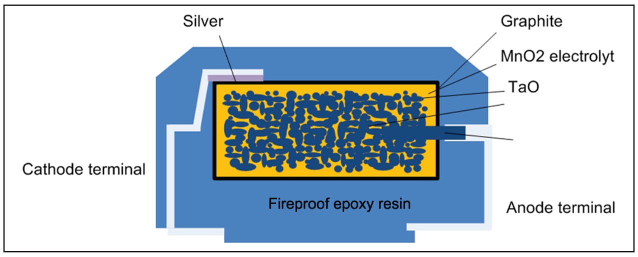

Figure 4. Composition of tantalum capacitor [compact capacitor with manganese oxide (MnO_2) electrolyte]; electrolyte varies depending on the capacitor type. TaO: tantalum oxide.

producers association ZVEI (Buchert et al., 2012), who considers that these data are currently out of date. Updated data were planned to be published on the ZVEI web page in spring 2013. Mineta and Okabe (2005) estimated a share between 40% and 50% of tantalum in tantalum capacitor scrap. Oguchi et al. (2011) experimentally determined tantalum concentrations in PCBs (including the capacitors) between 7 (desktop PC) and around 8000 mg kg^{-1} (digital camera and camcorder). The concentrations measured by Sander et al. (2012) were below the detection limit of 100 mg kg^{-1} for a smartphone and MP3 player, and amounted to 135 mg kg^{-1} in a tablet PC and 470 mg kg^{-1} in a navigation device. Tantalum is used in other electronic components, for example as tantalum oxide in tantalum film resistors (Tu, 2013), optoelectronic semiconductors (PatentDe, 2009) and lithium tantalite in surface acoustic wave filters (EPCOS, 2012).

Quantification of indicators related to the relevance of the applications for further research

The results of the meta-study on the criticality of the selected metals, as well as the data on metal production and share of metal production used by sectors related to the electronic industry, are presented in Table 1.

Figure 5 shows the quantitative data related to the metal content and concentration in the applications presented in the previous section. The numbers should be seen as orientation values and not as detailed data owing to the high uncertainties related to, for instance, the variety of manufacturing technologies (leading to strongly varying concentrations in the products and components), the quantification methods (experimental measurement or theoretical estimation) and the sampling uncertainties. The data presented in Figure 5 show the concentration and content ranges in the product part in which the application is embedded, for example the plastic casing for the application ‘Synergist for flame retardant in casings’, and the LCD panel for the applications ‘ATO transparent conductive layer’ and ‘ITO transparent conductive layer’ [see Figure 5 (notes)]. For most applications, the available data do not permit us to make statements on changes in the metal content and concentration that may be expected in the coming years.

Table 2 shows the occurrence of the applications in information technology, telecommunication and consumer equipment, classified according to the 18 United Nations University (UNU) keys from Wang et al. (2012) related to categories 3 and 4 defined in the WEEE Directive.

Table 3 summarizes the data accuracy, showing (1) if data are available or not, and (2) if the available data can be considered as indicative values giving orders of magnitude or as accurate, i.e. based on measurements using adequate measurement methods that are described by the research team on a representative sample of products. Only the data available for the application ‘ITO in flat displays’ were considered as accurate.

Priorities for research

Regarding the highly critical metal antimony, future research should focus on the recycling of plastics containing antimony as a synergist for flame retardants. This application is by far the most relevant regarding the share of metal used to produce flame retardants, as well as the concentration, content and occurrence in WEEE.

Data on the quantities of gallium and germanium used in ICs are not available, even though the share of the gallium production used in this application is high, and both metals were classified as critical by most studies.

The phosphors of CCFLs require more research owing to the relatively high content of REE in flat displays with CCFL backlighting. LEDs are also a field of interest regarding REE in phosphors. Phosphors use heavy REEs (terbium, europium and dysprosium), which are the most critical REE in regard to supply risks (Elsner, 2011). The gallium and indium content in LED products is so low that it is improbable that more research would deliver findings showing a sufficient potential to open ways to recover the metals.

Although the share of the worldwide production of cobalt used in WEEE is limited, cobalt in cathodes of batteries, for instance lithium-ion batteries, represents a relatively large potential for recovery and should therefore be better investigated. The available data on the occurrence of batteries in WEEE, classified according to product and battery types, are insufficient. The other applications of cobalt have a lower potential owing to lower concentration, content and/or occurrence in WEEE.

REE used in permanent magnets are also a relevant field of research owing to the high share of REE used to produce magnets and the relatively high concentration, content and occurrence in WEEE. Also, REE in nickel-metal hydride batteries and in electronic components should be better investigated because the currently available data are highly insufficient.

Even though tantalum was not classified as highly critical by four out of five studies, the large share of the metal production going to this application, as well as the high concentration in the components, make tantalum capacitors relevant for further research.

ITO in flat displays is the only application for which the available data can be considered as reliable. Also, the other indicators (criticality, metal share going to this application, concentration, content and occurrence) show the relevance of this application.

Table 1. Criticality, production and share used by sectors related to the electronic industry for the selected metals.

Metal	Criticality of the metal[a]	Metal production in tonnes in 2012[b]	Share of metal production going to sectors related to electronics
Antimony	High according to all studies that considered antimony	180,000[c]	USA: 35% used for flame retardants, not just for electronic products (USGS, 2013)
Cobalt	High (COM, 2010a; IW Consult GmbH, 2011; Rückschloss, 2010) Medium (Erdmann et al., 2011; Morley and Eatherley, 2008)	110,000[c]	USA: 16% in metallic applications excepting superalloys (USGS, 2013)
Gallium	High (COM 2010a; Erdmann et al., 2011) Medium (IW Consult GmbH, 2011; Morley and Eatherley 2008; NRC, 2008)	354[d]	18% for LED and 66% for integrated circuits (FEM/IUTA, 2011)
Germanium	High (COM, 2010a; Erdmann et al., 2011; IW Consult GmbH, 2011; Rückschloss, 2010) Medium (Morley and Eatherley, 2008)	128[d]	USA: 15% for electronics and solar electric applications (USGS, 2013)
Indium	High (COM, 2010a; Erdmann et al., 2011; IW Consult GmbH, 2011; NRC, 2008) Medium (Morley and Eatherley, 2008; Rückschloss, 2010)	670[d]	74% for flat display panels, 10% for low melting point alloys used as solders, 2% for semiconductors among others for LEDs (COM, 2010b)
Rare earths elements	High according to all studies that considered rare earths	110,000[c]	11% for hard disk drive and mobile phone magnets (Zepf, 2013), USA: 3% for phosphors (USGS, 2013)—detailed data differentiating 15 rare earth elements is provided by Du and Graedel (2011)
Tantalum	High (COM, 2010a) Medium (IW Consult GmbH, 2011; NRC, 2008) Low (Erdmann et al., 2011; Rückschloss, 2010)	765[c]	USA: 60% for capacitors (USGS, 2013)

LED: light-emitting diode.

[a] COM (2010a), Erdmann et al. (2011), IW Consult GmbH (2011), Morley and Eatherley (2008), NRC (2008), Rückschloss (2010).

[b] USGS (2013).

[c] Mine production.

[d] Refinery production.

Therefore, research needs to be taken to the next level, which is the development of technologies and infrastructure for metal recovery.

An important finding of the assessment of the data is that reliable quantitative data are available for almost none of the applications using the selected critical metals in EEE. Even though some investigations report the results of their respective analyses, most research uses secondary data and theoretical metals based on assumptions to calculate the content of critical metals. Recent findings showed a significant discrepancy between the results of theoretical estimations and of a measurement campaign focusing on indium used as ITO in flat displays (Rotter et al., 2013b). Although orientating values provided by estimates may have been enough for recommendations for policy makers, they are insufficient to estimate the potential for recycling, developing adequate technologies and optimizing infrastructure.

Besides the question ‘What should we investigate?’, the lack of documented data on analytical methods, as shown here, indicates that there is also a research demand for the question ‘How should we investigate?’. Jalalpoor et al. (in press) showed how different digestion methods can decisively influence the amount of detected indium by more than 50%. So far, no standard methods exist for heterogeneous matrices like WEEE. Waste-related standard methods regarding sample size reduction, digestion and detection of metals are only applicable to a limited extent. Standards like IEC/PAS 62596 and IEC 62321 only consider ‘restricted substances’ and are not validated for critical metals like those addressed in this article. In particular, acid digestion methods are very metal-specific. The use of inadequate digestion methods may lead to systematic under-detection of the correct concentration.

Under a recycling perspective, one should also discuss the significance of the parameter ‘concentration’ or ‘content’ as a stand-alone criterion. Van Schaik and Reuter (2010) showed that dynamic process models require, in particular, data on the liberation behavior of material/metals. The data can be gathered through performing dismantling and destruction tests, shredding and recycling experiments, or through determining particle characteristics after shredding. This type of data is work-intensive to collect, and often not available with the product designers or with the recyclers. For materials currently recovered, Reuter and Van

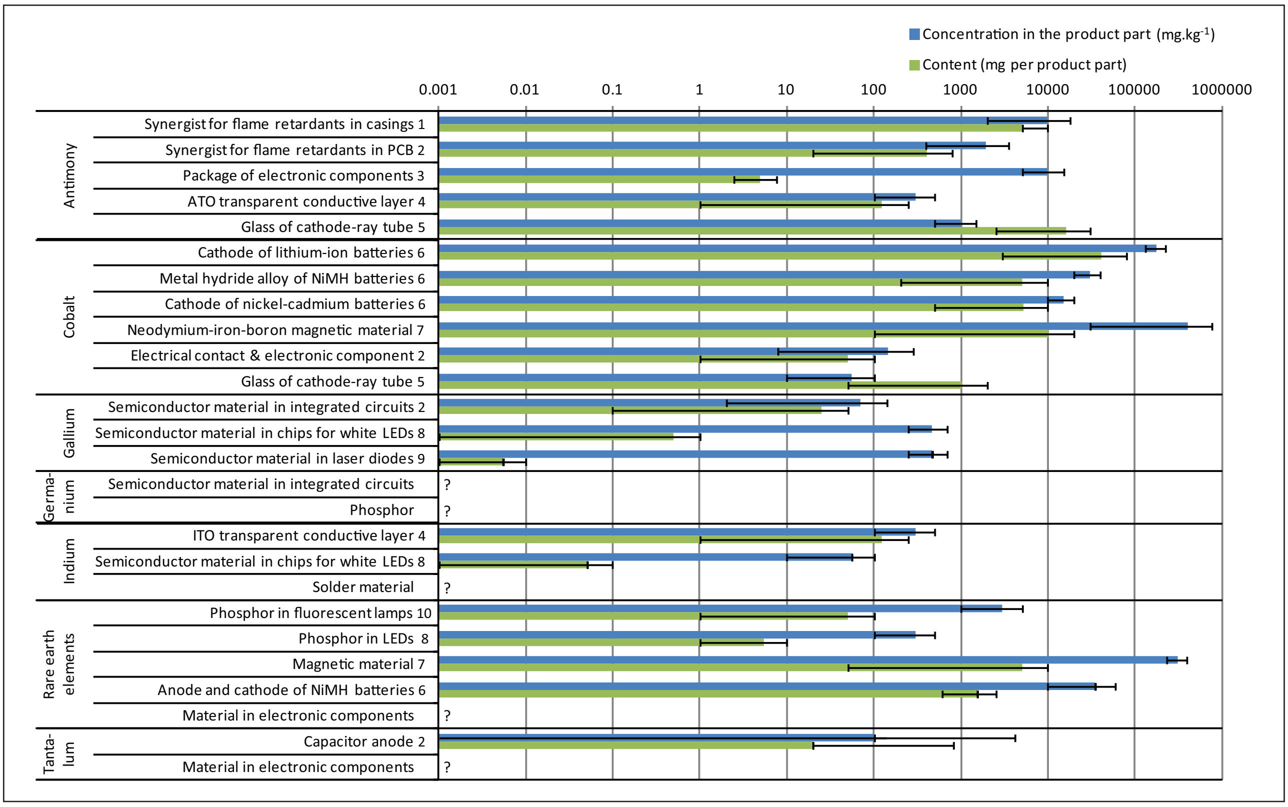

Figure 5. Rough estimate of metal concentration and content, based on the data presented.
Product parts considered to estimate concentration and content: 1—plastic casing; 2—printed circuit board; 3—electronic component; 4—liquid crystal display panel; 5—glass of cathode ray tube; 6—battery; 7—magnet; 8—light-emitting diode (LED); 9—diode; 10—cold-cathode fluorescent lamp. ATO: antimony–tin–oxide; NiMH: nickel–metal hydride; ITO: indium–tin–oxide.

Schaik (2008) and Van Schaik and Reuter (2007) presented the collection of this type of data. For critical metals, there is neither consistent methodology nor a knowledge base published yet. This shows that knowledge of downstream processes is required to define a solid data basis on critical metals in relevant waste stream like WEEE.

Table 4 summarizes the information demand for recycling-oriented product characterization in order to enhance critical metal recycling and allow multi-criteria optimization and decision making.

Conclusions

The research provides a methodological framework to deal with the question ‘What should be done to improve the recovery of critical metals from WEEE?’, as well as some answers through the listing of 26 applications using the selected metals, from which eight applications were identified as relevant for further research. These are the use of antimony as flame retardant; gallium and germanium in ICs; REE in phosphors of CCFLs and LEDs, as well as in permanent magnets; and cobalt in batteries, tantalum capacitors and ITO in flat displays. The findings will be used for further research in the frame of the UPgrade project (project duration 2012–2015), which aims at enhancing the recovery of trace metals along the value chain by developing new liberation and separation steps (mechanical, thermal, chemical), sufficiently considering the technological requirements of final recovery processes.

Next steps towards enhancing metal recovery are:

- developing a methodology of recycling-oriented characterization considering critical metals;
- increasing the knowledge base of metals concentrations and content in WEEE and their specification;
- holistic assessment of recycling processes regarding the overall recycling efficiency;
- development and implementation of managerial and technological innovations to improve in a holistic manner the metal-specific recovery rates.

Other research projects have started in parallel in European countries to investigate the potential of recycling to secure the supply of critical metals to the industry and develop technological and managerial recycling solutions. The Swiss project E-RECMET, the project RePro, commissioned by the German Federal Environment Agency, the European project RECLAIM and the Dutch Materials Innovation network can be named. A fruitful cooperation between these projects would bring synergistic effects and lead to innovative solutions for implementation into practice.

Table 2. Occurrence in Waste Electric and Electronic Equipment of the identified applications.

Metal	UNU key	301	302	303	304	305	306	307	308	309	401
	Application	Small IT and accessoires	Desktop PCs	Laptop PC’s	Printing and imaging	Telecom	Mobile phones	Professional IT	CRT monitors	Flat panel monit ors	Small CE and accessoires
Antimony	Synergist for flame retardants	Plastic casings, printed circuit boards (PCBs) and other plastic parts[a]									
	Packaging of electronic components	PCBs									
	ATO transparent conductive layer			LCD panel			LCD panel			LCD panel	
	Glass of CRT								Glass		
Cobalt	Cathode of lithium-ion batteries	Batteries		Batteries			Batteries				
	Metal hydride alloy of NiMH batteries	Batteries				Batteries					
	Cathode of nickel-cadmium batteries					Batteries					
	Magnetic material	Loud speakers	HDD	Loud speakers, HDD		Loud speakers	Loud speakers			Loud speakers	Loud speakers
	Electrical contacts and active part of electronic components	PCBs									
	Glass of CRT								Glass		
Gallium	Semiconductor material in integrated circuits	ICs		ICs		ICs	ICs				
	Semiconductor material in chips for white LEDs			Display backlighting	Scanner lamp		Display backlighting			Display backlighting	
	Semiconductor material in laser diodes		DVD, CD,Blu-ray-player	DVD, CD,Blu-ray-player							
Germanium	Semiconductor material in integrated circuits	ICs		ICs		ICs	ICs				
	Phosphor				Lamps?						
Indium	ITO transparent conductive layer			LCD panel			LCD panel			LCD panel	
	Semiconductor material in chips for white LEDs			Display backlighting	Scanner lamp		Display backlighting			Display backlighting	
	Solder material	Rarely used in PCBs									
Rare earth elements	Phosphor in fluorescent lamps[c]			Display backlighting						Display backlighting	
	Phosphor in LEDs			Display backlighting	Scanner lamp		Display backlighting			Display backlighting	
	Magnetic material	Loud speakers	HDD	Loud speakers, HDD		Loud speakers	Loud speakers			Loud speakers	Loud speakers
	Anode and cathode of NiMH batteries	Batteries				Batteries					
	Material in electronic components									PCBs?	
Tantalum	Capacitor anode			Capacitors			Capacitors				
	Material in electronic components									PCBs?	

Metal	UNU key	402	403	404	405	406	407	408	409
	Application	Portable audio and video	Radio and hi-fi	Video	Speakers	Cameras	CRT TVs	Flat display panel TVs	Professional CE equipment
Antimony	Synergist for flame retardants								
	Packaging of electronic components								
	ATO transparent conductive layer							LCD panel	
	Glass of CRT						Glass		
Cobalt	Cathode of lithium-ion batteries	Batteries				Batteries			
	Metal hydride alloy of NiMH batteries	Batteries				Batteries			
	Cathode of nickel-cadmium batteries	Batteries							
	Magnetic material	Loud speakers	Loud speakers	Loud speakers	Loud speakers	Loud speakers	Loud speakers	Loud speakers	
	Electrical contacts and active part of electronic components								
	Glass of CRT						Glass		
Cobalt	Semiconductor material in integrated circuits								
	Semiconductor material in chips for white LEDs	Display backlighting				Display backlighting		Display backlighting	
	Semiconductor material in laser diodes								
Germanium	Semiconductor material in integrated circuits								
	Phosphor								
Indium	ITO transparent conductive layer							LCD panel	
	Semiconductor material in chips for white LEDs	Display backlighting				Display backlighting		Display backlighting	
	Solder material								
Rare earth elements	Phosphor in fluorescent lamps[c]							Display backlighting	
	Phosphor in LEDs	Display backlighting				Display backlighting		Display backlighting	
	Magnetic material	Loud speakers	Loud speakers	Loud speakers	Loud speakers	Loud speakers	Loud speakers	Loud speakers	
	Anode and cathode of NiMH batteries	Batteries				Batteries			
	Material in electronic components								
Tantalum	Capacitor anode	Capacitors				Capacitors			
	Material in electronic components								

IT: information technology; PC: personal computer; CRT: cathode ray tube; CE: consumer electronics; ATO: antimony–tin–oxide; LCD: liquid crystal display; NiMH: nickel–metal hydride;
HDD: hard disk drives; IC: integrated circuits; LED: light-emitting diode; ITO: indium–tin–oxide.

[a] 22% of the casings of televisions and PC monitors contained at least 1% antimony (Schlummer et al., 2007), in general 4–5% of the high impact polystyrene and acrylonitrile butadiene styrene plastic contain antimony oxide (European Flame Retardants Association, 2011).

[b] In 2009 no alternative to ITO could be found in flat displays (Patel-Predd, 2009).

[c] The share of flat displays in WEEE is currently increasing (Chancerel et al., 2012), but the market share of devices with cold-cathode fluorescent lamp (CCFL)-backlighting decreased rapidly, so that the arising of displays with CCFL backlighting is expected to be temporary.

Table 3. Accuracy of the available data.

Metal	Application	Accuracy of the data already available
Antimony	Synergist for flame retardants	Indicative values based on measurements available for casings and PCBs
	Package of electronic components	Indicative values based on estimations and standards
	ATO transparent conductive layer	No data
	Glass of CRT	Indicative values based on measurements
Cobalt	Cathode of lithium-ion batteries	Indicative values based on estimations, measurements and producer information
	Metal hydride alloy of NiMH batteries	Indicative values based on estimations and measurements
	Cathode of nickel–cadmium batteries	Indicative values based on estimations
	Magnetic material	Indicative values based on industry standards specifications
	Electrical contact	Indicative values based on estimations and measurements of PCBs
	Active part of electronic components	Indicative values based on standards and measurements of PCBs
	Glass of CRT	Indicative values based on measurements
Gallium	Semiconductor material in integrated circuits	No data on chips, indicative values based on measurements of PCBs
	Semiconductor material in chips for white LEDs	Indicative values based on measurements and estimations
	Semiconductor material in laser diodes	No data
Germanium	Semiconductor material in integrated circuits	No data
	Phosphor	No data
Indium	ITO transparent conductive layer	Detailed data based on measurement for different equipment types
	Semiconductor material in chips for white LEDs	Indicative values based on measurements and estimations
	Solder material	No data
Rare earth elements	Phosphor in fluorescent lamps	Indicative values predominantly based on estimations
	Phosphor in LEDs	Indicative values predominantly based on estimations
	Magnetic material	Values based on measurements and standards
	Anode and cathode of NiMH batteries	Indicative values based on rough estimations
	Material in electronic components	No data
Tantalum	Capacitor anode	Indicative values based on measurements and out-dated standards
	Material in electronic components	No data

ATO: antimony–tin–oxide; CRT: cathode ray tube; PCBs: printed circuit boards; NiMH: nickel–metal hydride; LED: light-emitting diode; ITO: indium–tin–oxide.

Table 4. Theoretical framework of recycling-oriented product characterization for critical metals based on International Resource Panel/M. Reuter et al. (in press), Rotter et al. (2013, 2013b) and Van Schaik and Reuter (2010).

Aspect of information demand	Criteria
Liberation behavior	Perfect liberation, remains closely connected to other materials after mechanical liberation
Metal specification	Metallic, alloyed metal, intermetallic phases, metal oxide, other metal salt....
Material specification	Casted, sintered, powdery, coated, laminated ...
Metal association	'Sister' metals impurities

Conflict of interest

The author declares that there is no conflict of interest.

Funding

This work was funded by the German Federal Ministry of Education and Research (BMBF) (UPgrade - Project reference number 033R087A).

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