

Applied WEEE pre-treatment methods: Opportunities to maximizing the recovery of critical metals

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Abstract

WEEE is a fast-growing waste stream that includes potentially hazardous substances, but also valuable secondary raw materials, which can be recovered by adequate recycling and recovery treatment. In the last years, the research interest has moved from the conventional recycling (recovery of ferrous and non-ferrous metals, plastic, glass and other "mass relevant" fractions presented in WEEE), to the innovational recycling, aimed to recover trace elements, such as critical metals (CMs) and rare earth elements (REEs). Currently, the majority of CMs and REEs are lost during the pre-treatment processes. In this paper, an overview of the most relevant e-waste categories and products in terms of CMs and REEs presence, a description of currently applied pre-treatment methods and fate of the observed group of metals during pre-processing phase, as well as general recommendation in order to avoid losses of CMs and REEs within the WEEE treatment chain, are elaborated.

Keywords: Electronic waste, physical separation, secondary resources, rare earth elements.

1. Introduction

According to the European Commission Report (EC, 2017), 27 raw materials, which have strategic importance for EU manufacturing industry, high economic importance and high supply risk, were identified as critical raw materials (CRMs) at EU level. EU Raw Materials Initiative started in 2008 to define their secure, sustainable and affordable supply. The CRMs list, updated every 3 years, is a crucial element of this Initiative. EU interest on CRMs is focused on the support of technical solutions that could alleviate EU dependence on import from developing countries. Within Report (EC, 2017), *"Import reliance rate"* calculated as a ratio between net imports of specific CRM in EU countries and its production on the EU level, shows that for 14 materials of 27, its value is equal, or nearly 100%. On the other hand, *"Recycling input rate"* which indicates the ratio of recycling and EU demand of a given critical raw material, is only up to 1% for 17 observed CRMs (Figure 1).

Moreover, EU Raw Materials Initiative is fully coherent with EU strategy about transitioning towards a low-carbon, resource efficient and more circular economy. In line with that, WEEE is recognized as a very important secondary resource of CRMs (Chancerel *et al.*, 2013; Marra *et al.*, 2015; Diaz *et al.*, 2015). In particular, metals from the list of critical raw materials, such as gallium, tantalum, indium, cobalt, beryllium, tungsten, etc., which can be found in WEEE are therefore defined as a critical metals (CMs). Besides, a specific group of critical metals like europium, terbium, yttrium, cerium, gadolinium, neodymium, and others, also contained in WEEE, are designated as a rare earth elements (REEs).

In addition, base metals (iron, steel, copper, aluminum) and precious metals (gold, silver, palladium, platinum) as the most dominant metals in WEEE, even if not CMs, are very important for EU economy as they are needed in large amounts as starting point of manufacturing chains. Therefore, it is evidently that the contribute that WEEE may give within a circular economy approach plays a primary role.

In order to separate components that contain valuable materials or to upgrade relevant fractions prior to recycling process, it is crucial to focus on increasing the collection rates and mainly on optimizing the operating conditions of pre-processing stage in WEEE recycling chain. In fact, the recovery of a specific material from end-of-life e-waste input stream increases with decreasing of impurities in the final material (Chancerel *et al.*, 2009). Even the key role is the determination of the obsolete product characteristics, the main defect still

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occurs at the pre-treatment stage (Meskers *et al.*, 2009), which includes liberation through manual dismantling, and/or shredding and manual and/or mechanical separation (Bigum, 2012), and result overall the reduction of the recycling efficiency. In order to enhance critical metals recovery, it is necessary to develop specialized systematic approaches in those certain e-waste streams that are composed by higher concentrations of CMs.



Figure 1. Reliance on import and recycling input rates for selected CRMs on EU level (data used from EC, 2017)

Therefore, a compromise between the quality and quantity of grade-recovery fraction has to be carried out to minimize the losses of valuable metals and the distribution of critical metals over the outputs of the WEEE preprocessing stage. The objective of this paper is to present a succinct overview on the e-waste categories and products in terms of CMs and REEs, the currently applied pre-treatment processes, and the fate of the CMs/REEs during pre-treatment phase.

2. Relevant e-waste categories and products in terms of CMs/REEs

The environmental impact, the exploitation of resources and the global regulations, but mainly the values contained in e-waste become the main driving forces for the recovery of critical and rare earth metals from e-waste. Not only the recycling of e-waste leads to conservation of primary ores, saving energy and reducing carbon footprint, but it should be also noticed that the amount of CMs in WEEE is comparable, and in some cases even significantly higher to the content of precious and critical metals in mineral ores (Işıldar *et al.*, 2017), which leads to the fact that WEEE becomes important resource within "urban mining" concept.

Since one of the most important parameter for pretreating e-waste without losing CMs is to know in advance their initial concentrations, the content of WEEE in terms of CMs is presented in Table 1. This is essential in order to classify the most important or most "mass relevant to CM" categories of WEEE included in Directive's catalogue.

Products that include IT and telecommunication equipment, consumer equipment (i.e., category 3 and 4 according to WEEE Directive 2012/19/EU), as well as lighting equipment (category 5), are the richest of precious metals and rare earth elements (Bigum, 2012;

Tsamis and Coyne, 2015). High-grade electronic scrap such as automotive catalysts, batteries, circuit boards, mobile phones and MP3 players need to be separated prior to mechanical pre-processing to prevent irrecoverable losses (Gunn, 2014). For low-grade electronic scrap such as small domestic appliances and audio-visual equipment, a coarse size reduction is needed prior the removal of circuit board fractions. In addition, high-strength magnets used in motors and hard disk drivers may represent an important source in terms of REEs because their concentration within them is significant high (Gunn, 2014).

Focusing on printed circuit boards (PCBs), these contain polymers, ceramics as well as metals. Specifically, the metal content is around 28% (consisting of copper: 10-20%, lead: 1-5%, nickel: 1-3%). Some precious metals are also present (in decreasing order: silver, platinum metals like palladium, gold, to a total of \approx 0.3-0.4%) (Veit *et al.*, 2005). Metals such as gallium, indium, titanium, germanium, arsenic, selenium and tellurium can be found in lower content values in integrated circuits and semiconductors (Işıldar *et al.*, 2017).

Similarly, regarding batteries, mercury is located in the dry batteries and cadmium in the Ni-Cd batteries (Yue-qing and Guo-jian, 2004), but also cobalt is found in them (Sommer *et al.*, 2015).

According to the scientific community, REEs are significantly located into permanent magnets (particularly neodymium and praseodymium), nickel-hydride batteries and fluorescent lamps (Binnemans *et al.*, 2013; Hobohm *et al.*, 2016; Işıldar *et al.*, 2017). However, the low concentrations of rare elements, the high market price, the complex components and the heterogeneous combination of materials (Charles *et al.*, 2017; Marra *et al.*, 2015; Savvilotidou *et al.*, 2014) make their recovery difficult, but also challenging (Dias *et al.*, 2016).

3. Description of currently applied pre-treatment processes

For efficient recycling of e-waste, pre-treatment as a combination of manual (dismantling and separation) and physical processes (size reduction and different physical separation) represent a first and indispensable step. The main task of WEEE pre-processing is to ensure that liberated materials enter the appropriate recovery processes (Ueberschaar *et al.*, 2017a). Before final metal recovery in hydro and/or pyro metallurgical processing, appropriate pre-treatment steps, are crucial.

Currently applied pre-treatment methods are customized for efficient separation and recovery of "mass relevant" fraction in e-waste, mostly metals (Fe, steel, Ni, Al, Cu, brass), but also non-metallic fractions, like plastic, glass, rubber, wood, textile etc. Thus, the majority of CMs and REEs are lost, as they stay coupled with dominant metal output fractions, or end up in the dust from the process. This disables possibility for their recovery in hydrometallurgical or pyrometallurgical process, as end-processing steps (Chancerel *et al.*, 2009). In general, pre-treatment phase includes:

- Manually disassembly/dismantling
- Mechanical treatment processes
- Combination of manual and mechanical preprocessing

Manually dismantling phase is crucial to ensure removal of all relevant hazardous and/or valuable components such as: casings, external cables, cathode ray tubes (CRTs), PCBs, batteries, etc., defined in Annex VII of the WEEE Directive 2012/19, while this step becomes labor intensive (Zhang and Forssberg, 1998).

As shown in Figure 2, manual sorting and dismantling are usually followed by a traditional recycling processes, where metals and materials contained in WEEE are liberated and separated based on their specific physical characteristics such as weight, size, shape, density, and electrical and magnetic characteristics (Oliveira *et al.*, 2012).

Table 1. Precious, critical and rare earth metals content in high-grade e-waste products

Category/type of WEEE	Element	Element	Average	References
1 LARGE HOUSEHOLD APPLIANCES	category		content (wt/oj	
PCRs		Δσ	0 1F-01	D'Adamo et al. 2016
1000	Precious metals	<u> </u>	0.3E-02	D / ddino ct ul., 2010
		Pd	0.3E-02	
	Critical metals	Sh	0.8E-01	D'Adamo et al 2016
2. SMALL HOUSEHOLD APPLIANCES	Childen Hierdis	55	0.02 01	B Haumo et any 2010
		Ag	2.2E-04	Huismann et al., 2007
	Precious metals	Au	2.0E-05	
		Pd	1.2E-05	<u>.</u>
3. IT & TELECOMMUNICATION EQUIPMENT				
Personal computers	Precious metals	Ag	6.4E-02	Cui and Forssberg, 2007
Liquid-Crystal Display monitors	Precious metals	Au	2.3E-03	Bakas <i>et al.,</i> 2016
		Ag	7.4E-03	
		Pd	6.1E-04	
		ln	5.3E-02	Savvilotidou et al., 2015
Mobile phones		Pd	0.4E-02	
	Precious metals	Au	0.9E-02	Bakas <i>et al.,</i> 2016
		Ag	0.9E-02	
	Critical metals	Со	0.2E+01	
		Ве	0.1E-02	
PCBs (from laptops and mobile phones)		Ag	1.3E-01	- - Wang and Gaustad, 2012 -
	Precious metals	Au	3.6E-02	
		Pd	1.2E-02	
		Pt	2.2E-03	
	Critical metals	Ga	3.5E-03	
		Та	1.7E-02	 Wang and Gaustad, 2012
4. CONSUMER EQUIPMENT AND PHOTOVOLTAIC PANELS				
	Precious metals	Ag	0.6E-01	Dias <i>et al.,</i> 2016
Photovoltaic modules	Critical metals	In	7.9E-03	- Savvilotidou <i>et al.,</i> 2017
		Ga	1.5E-02	
TV sets	Precious metals	Ag	0.2E-02	- Cui and Forssberg, 2007
		Au	0.7E-02	
	Critical metals	W	9.1E-03	Bakas <i>et al.,</i> 2016
PCB from cathode ray tubes (CRTs) TV monitors	Precious metals	Ag	2.8E-02	- Cui and Forssberg, 2007
		Au	1.1E-02	
5. LIGHTING EQUIPMENT				
Fluorescent lamps		La	6.0E-01	Sun <i>et al.,</i> 2016
		Ce	6.0E-01	
		Y	0.8E+01	
Fluorescent powders (CRTs)		Y	7.8E-04	Innocenzi <i>et al.,</i> 2013
Phosphors		La	0.8E-01	Binnemans <i>et al.,</i> 2013
	Rare earth metals	Ce	1.1E-01	
		Eu	0.5E-01	
		Gd	0.2E-01	
		Tb	0.5E-01	
		Y	6.9E-01	



Figure 2. General scheme of applied processes in WEEE pretreatment stage

Typically, mechanical pre-treatment starts with size reduction, using shredding or crushing process. Depending on the needed size of output material, several shredding/grinding processes are used during the whole mechanical pre-treatment stage. After size reduction, separation of a group of ferrous metals (Fe, steel, Ni, etc.) is performed by magnetic separation, where low intensity magnetic drum separators are widely used for this purpose. This step is usually followed by separation of non-ferrous metals flow (Al, Cu, etc.) from other materials (mainly plastic, glass, etc.), by using some of the electric conductivity-based separation techniques. Eddy current separation and corona electrostatic are the most commonly used in e-waste recycling processes, especially after strong permanent magnets, such as iron-boron-neodvmium magnets, became available (Zhang and Forssberg, 1998).

Finally, based on the fact that every material particle in WEEE has a specific density, gravity concentration methods separate materials of different specific gravity by their relative movement in response to gravity. Gravity separation techniques include water or airflow tables, heavy media floating, sifting, etc. (Oliveira *et al.*, 2012).

As an alternative or in combination with some of the described conventional mechanical pre-treatment processes, sensor and/or optical based sorting techniques can be also applied (Hageluken, 2006). Usually, sensor sorting can be used in the advanced stage of pretreatment process, in order to further separate different types of plastics and possibly glass from the output stream. Menad et al. (2013) show that plastic can be efficiently separated from non-metallic fraction flow using sorting devices. Through the performed sensor characterization of samples from different recycling industries, it was concluded that recognition of plastic with IR (infrared) sensors is not applicable for black and coloured plastic fragments. However, sorting of coloured plastics in different polymer types can be efficiently done by fast NIR (Near Infra-Red) spectroscopy. Furthermore, flame retardants and other additives in plastic from WEEE (bromine, phosphor, antimony) can be identified with XRF device.

4. The fate of CMs in pre-treatment processes

Fully automated disassembly is currently not technically feasible and not expected to become economically viable in coming years (Duflou et al., 2008). Currently, the greatest need for the technical optimization of recycling is to ameliorate manual disassembly during the pre-treatment stage. Besides, Buchert et al. (2012) show that the general standard practice of shredding whole devices, in Germany, leads to considerable losses of CMs in particular precious metals - which cannot be compensated by the downstream sorting and refining processes. To improve the recovery of precious and other critical metals, it is essential to manually remove the components containing important raw materials such as PCBs and batteries and feed these components separately to recycling facilities. Based on elemental material flow analysis Ueberschaar et al. (2017a), concluded that in pre-sorting step due to battery removal, about 46% of cobalt and 38% of a group of REEs are separated from the input WEEE flow. High concentrations of cobalt and REEs (about 30% and 60%, respectively) were also found within ferrous metals fraction after use of magnet separator (Figure 3).



Figure 3. Mass balance of the cobalt, gallium, indium, tantalum and group of REEs during WEEE pre-processing stage (redrawn from Ueberschaar *et al.*, 2017a)

In order to examine the losses of precious metals due to shredding of PCBs, Chancerel *et al.* (2009), through an industrial test, determined the difference in concentration between unshredded and preshredded PCBs. The results showed 7% less precious metals in the preshredded PCBs compared with the un-shredded PCBs. Furthermore, the difference between preshredded PCBs (size < 8 mm) and shredded PCBs (size < 2.5 mm) indicates 62% less precious metals in the shredded PCBs.

Ueberschaar *et al.* (2017b) show that the relatively small shares of gallium bearing components on PCBs or in light emitting diodes (LEDs) lead to a dilution with other materials in recycling processes used conventionally. Ending in the pyrometallurgical process for copper and precious metals refining, gallium is transferred as oxidized

form to the slag. Thus, gallium rich components must be separated prior to any mechanical processing with other material. In particular, mobile phones and newly tablets are important gallium sources, bearing more than 40% of the total gallium loads in the IT and entertainment equipment.

On example of neodymium, Marra *et al.* (2015), demonstrated that about 90% of Nd is lost throughout conventional pre-treatment techniques, mainly ended up in the process air dust.

5. Recommendation to avoid losses of CMs/REEs within the WEEE treatment chain

In order to recover CMs from WEEE, some of the crucial improvements in steps that precede pre-treatment phase in overall recycling chain, includes:

- More reliable and transparent information about the content of CMs in the different equipment groups and their components
- Increase of the collection rates for all product groups that contain CM
- Optimized structure and design of EEE products that facilitates manual disassembly and recycling processes.

With regard to WEEE pre-treatment phase, more particular recommendation and measures for avoiding losses of CMs and REEs are:

- Optimization of pre-treatment processes based on further improvement of manual disassembly and separation of target components in WEEE with high concentration of CMs. Besides removal of the WEEE components that are legally stipulated, parts of equipment such as batteries containing cobalt, neodymium hard disk magnets, small PCBs, etc. (Buchert *et al.*, 2012), should be also removed and fed into a suitable recycling process.
- ii Outputs from pre-treatment phase must be fractions with form and characteristics appropriate for end-processing facilities. More investments and further research should be focused on promising technologies for automatic recognition, sorting, and dismantling of WEEE, in order to recover CM from heterogenous WEEE flows, more efficiently (Buchert *et al.*, 2012; Chancerel *et al.*, 2010).
- iii Quantitative targets for the recycling of WEEE are not formulated specifically in terms of material or components, but relate to the weight percent of the complete devices, which leads to negative incentives for the recovery of CM. Therefore, revision of the WEEE Directive in terms of setting targets for the recycling rates for specific critical metals and/or product groups, is recommended (Buchert *et al.*, 2012; Chancerel *et al.*, 2010).

6. Conclusion

Currently, the worldwide recycling rate of critical metals from WEEE is negligible. Although pre-treatment methods

can effectively separate mass-relevant metals like iron, aluminum and copper, CMs and REEs as a trace metals are practically completely lost throughout conventional techniques, mainly due to fact that these substances end up in a form of the output stream from which they cannot be recovered in final processing steps. Thus, it is obvious that adjustment of applied pre-treatment processes, in order to achieve adequate CM output stream for recovery, is needed. Measures such as enhancement of manual dismantling phase and separation of components in WEEE with high concentration of CMs, adjustment of pre-treatment process that enable appropriate output materials suitable for end-processing facilities, as well as amendment of the WEEE Directive and defining recycling targets based on specific material or components, represent the essential steps to avoid further losses of CMs from WEEE. However, providing the economic viability of the pre-treatment improvements, is one the main prerequisite for successful of implementation of aforementioned measures.

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