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 using of adequate recycling and recovery treatment solutions. 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 metals (REMs). Currently, the majority of CMs and REMs 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 REMs 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 REMs within the WEEE treatment chain, are elaborated.

Keywords: WEEE, Critical Metals (CMs), Rare Earth Metals (REMs), Waste Pre-treatment

1. Introduction

According to the European Commission Report **(EC, 2014)**, twenty raw materials which have both, high economic importance and high supply risk, were identified as critical raw materials at EU level. Beside base metals (iron, steel, copper, aluminum, etc.) and precious metals (gold, silver, palladium, platinum, etc.), WEEE is also very important secondary resource of critical metals (CMs) (Chancerel *et al.*, 2013; Marra, *et al.*, 2015; Diaz *et al.*, 2015). 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 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/REMs. 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 precious metals over the outputs of the operating conditions for WEEE preprocessing. The objective of this preliminary work is to present an overview on the e-waste categories and products in terms of CMs and REMs, the currently applied pre-treatment processes, and the fate of the CMs/REMs during pre-treatment phase.

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

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. 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 richest on precious metals and rare earth elements (Bigum, 2012; Tsamis, & 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).

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

No. of category (type) of WEEE according to Directive	Element category	Element	Mean wt.%	Reference
1. LARGE HOUSEHOLD APPLIANCES				
PCBs	Precious metals	Silver	0.1304	
		Gold	0.0359	
		Palladium	0.0117	**Wang and Gaustad, 2012**
		Platinum	0.0022	
	Critical metals	Gallium	0.0035	
		Tantalum	0.0172	
3.IT & TELECOMMUNICATION EQUIPMENT				
PCs	Precious metals	Silver	0.639	
Liquid-Crystal Display monitors	Precious metals	Gold	0.00233	
		Silver	0.0074 – 0.020	**Cui and Forssberg, 2007**
		Palladium	0.00061	
	Critical metals	Indium	0.530	**Bakas *et al.*, 2016**
		Gallium	0.00006	
PCB from an LCD monitor/TVs	Precious metals	Silver	0.025 – 0.13	**Savvilotidou *et al.*, 2015**
		Gold	0.006 – 0.49	
		Palladium	0.0019 – 0.0099	**Huismann *et al.* 2007**
Cellular phones	Precious metals	Palladium	0.004	
		Gold	0.009	
		Silver	0.090	
Hard disk drive PCB	Critical metals	Beryllium	0.001	
4. CONSUMER EQUIPMENT AND PHOTOVOLTAIC PANELS				**Dias *et al.*, 2016**
Photovoltaic modules	Precious metals	Silver	0.600	**Savvilotidou *et al.*, 2017**
	Critical metals	Indium	0.007870	
Television sets	Precious metals	Gallium	0.154	**Cui and Forssberg, 2007**
		Silver	0.020	
		Gold	0.007	**Bakas *et al.*, 2016**
	Critical metals	Tungsten	0.00915	
5. LIGHTING EQUIPMENT		Europium	0.49-0.88	
Cathode-ray tubes (CRTs)		Terbium	0.01-0.26	**Sun *et al.*, 2016**
Fluorescent lamps (CFLs)		Yttrium	4.57 - 14	
LEDs	Rare earth metals	Cerium	0.6	
Phosphors		Gadolinium	0.46	**Innocenzi *et al.*, 2013**
		Lanthanum	0.6 – 8	
Batteries (types)				
Li-ion LCO	Critical metals	Cobalt	14.0	
Li-ion NCA			2.5	
Li-ion NMC			4.0	
Li-ion other types			0.0	**Sommer *et al.*, 2015**
NiCd			1.0	
NiMH AB5			3.0	
NiMH other types			3.0	
Magnets (types)				**Nixon *et al.*, 2016**

RE-cobalt alloys	Rare earth metals		34 – 36	
Loudspeaker magnets			31	**Buchert *et al*., 2012**
Magnets for voice coil accelerator			29	
RE-transition element alloys	Critical metals	Cobalt	23 – 28	
RE-iron alloys	Rare earth metals	Neodymium	30 - 35	

For low grade electronic scrap such as small domestic appliances and audio-visual equipment, it is needed a coarse size reduction prior the removal of circuit board fractions. In addition, high-strength magnets used in motors and hard disk drivers are a potentially more important source in terms of REMs because their concentration within them is significant higher (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%) (Veit *et al*., 2005). Some precious metals are also presented (in decreasing order: silver, platinum metals like palladium, gold, to a total of ≈0.3–0.4%) (Veit *et al*., 2005). Similarly, regarding batteries, mercury (Hg) is located in the dry batteries and cadmium (Cd) 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, REMs are significantly located into magnets, nickel-hydride batteries and fluorescent lamps (Binnemans *et al*., 2013; Hobohm *et al*., 2016). 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

The main task of WEEE pre-processing is to ensure that liberated materials enter the appropriate recovery processes. Before final metal recovery in hydro and/or pyro metallurgical processing, appropriate pre-treatment steps, are crucial. Loss of CM and REM in overall recycling chain is mainly caused by fact that these substances in pre-treatment phase end up in output streams from which they can't be recovered in end-processing steps (Chancerel *et al*., 2009). In general, pre-treatment phase is consisted of:

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

Through the manually dismantling phase, hazardous or valuable components, such as: casings, external cables, CRTs, PCBs, batteries, etc., are manually separated, making this step labor intensive (Zhang and Forssberg, 1998). Manual sorting and dismantling is 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). Typically, mechanical pre-treatment start with size reduction, using shredding or crushing process. Depending on needed size of output material, several shredding/grinding processes are used during the whole mechanical pre-treatment stage. After size reduction, separation of 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 electric conductivity-based separation techniques. Eddy current separation and corona electrostatic are the most common used in e-waste recycling processes, especially after strong permanent magnets, such as iron-boron-neodymium 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 technics include water or airflow tables, heavy media floating, sifting, etc. **(Oliveira *et al*., 2012).**

4. Fate of CMs in pre – treatment processes

Currently the greatest need for the technical optimization of recycling is to ameliorate manual disassembly during the pre-treatment stage. Buchert *et al*. (2012), show that the generally 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 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. Fully automated disassembly is currently not technically feasible and not expected to become economically viable in coming years (Duflou *et al*., 2008). 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 and shredded PCBs indicates 62% less precious metals in the shredded PCBs compared with the preshredded PCBs. Ueberschaar *et al*. (2017), show that the relatively small shares of gallium bearing components on PCBs or in 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 CM/ REM 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 REMs 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.
- 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).
- 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 REMs 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 can not 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 are 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 of the main prerequisite for successful implementation of aforementioned measures.

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