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Separation efficiency of valuable and critical metals in WEEE mechanical treatments

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Highlights

•The fate of critical metals during WEEE mechanical treatments was investigated.
•The dust fraction could turn from waste stream to alternative source of REEs.
•Technical aspects related to potential recovery of critical metals were discussed.

Abstract

The high demand of rare earth elements (REEs) in electronics industry and their high supply risk owing to the dependence on limited source countries have increased the interest towards Waste Electrical and Electronic Equipment (WEEE) as a potential secondary source of these elements, identified as critical metals. Although REEs are present in a wide range of high-tech products in relative low concentrations, the recycling of critical metals from WEEE is regarded as an important opportunity for promoting the conservation of primary resources and preventing waste production. However, the existing systems for WEEE collection and treatment mainly focus on the recovery of base and precious metals while the fate of REEs has not been addressed yet. To this end, the present study aims at evaluating the substance flows of critical metals in WEEE mechanical pre-treatments as these processes, preceding the metallurgical treatments of refining, determine the amount of metals entering the further recovery chain. The separation efficiency of a full scale mechanical process, including a sequential shredding and selection treatment of small WEEE, was investigated assessing the mass balance of both base metals and special metals as well as the quality of the output products. The mass flow analysis revealed that after pre-treatments only a third of precious metals entering the treatment process was conveyed to the target output destined to further recovery and less than 2% of REEs was concentrated in the potentially recyclable metallic fractions, while approximately 80% of these elements was distributed in the dust stream actually destined to landfill. Furthermore, the study pointed out that the fine fraction of the metallic outputs from the sorting process is characterized by a major degree of purity, indicating that both the dust stream and the fine grain fraction could be regarded as secondary sources for the recovery of valuable and critical metals from WEEE.

Keywords

Waste electrical and electronic equipment
Flow analysis
Precious metals
Rare earth elements
Secondary source
Dust stream

1. Introduction

Due to the increasing exploitation of resources and the scarcity of native raw materials, the strategies of waste management have been turned from the linear model of “take-use-waste” to a circular approach based on the prevention of waste as well as the re-introduction of the materials into the economic “loop”. In place of virgin materials, compounds and elements are desirably reclaimed from anthropogenic stock resources, such as waste which acts as “urban mines” (Cossu and Williams, 2015). In this regard, great attention has been focused on Waste Electrical and Electronic Equipment (WEEE) as this waste stream is characterized by the highest growth rate per year and by the most wide-ranging source of materials (Tanskanen, 2013, Widmer et al., 2005).

As a result of the continuous expansion of the electronic market and the reduction of the lifespan of many electronic devices, around 20–50 million tons of WEEE are annually generated worldwide (UNEP, 2013) and this trend was expected to reach 65.4 million tons in 2017 (Environment OREP, 2014). Such large volume of produced waste contributes to make WEEE management a challenge, especially when considering its extremely variable and complex composition, coupling valuable materials with harmful substances (Baldé et al., 2015, Cui and Zhang, 2008). If the presence of hazardous components in WEEE can deal with the potential risk for both human health and environment as a result of an improper handling of this waste (Kiddee et al., 2013, Tsydenova and Bengtsson, 2011), the relatively high concentrations of metals can provide a promising reserve of these materials (Jadhav and Hocheng, 2012, Lee and Pandey, 2012, Oguchi et al., 2011, Tuncuk et al., 2012).

Both base and precious metals are largely used in the industry of electrical and electronic devices. Their economic value, along with the limited reserves and the environmental impacts related to their primary production (Behrendt et al., 2007), represents a relevant incentive for resource recovery from waste materials. This aspect is especially evident for rare earth elements (REEs), since this group of metals, which are widely used in many field of high-tech applications (Schüler et al., 2011), has been identified by the European Commission as the most critical: its scarce worldwide production, mainly limited to China, and the current low recycling rate entail its high supply risk (Binnemans et al., 2013, European Commission, 2014).

Due to their magnetic, luminescent and chemical properties, REEs are highly demanded in electronics industry as they make possible the miniaturization of information technology (IT) components and batteries. Rare earths are used in several parts or components of electronic devices, such as fluorescent lamps, magnets, accumulators, semi-conductors, capacitors and batteries. The higher contents of these critical elements can be found in products containing phosphors (fluorescent lamps, cathode ray tubes, LED and plasma display panels), neodymium-iron-boron (NdFeB) magnets (hard disk drives, speakers, headphones, mobile phones) and nickel-metal hybrid batteries (NiMH) (Binnemans et al., 2013, Tunsu et al., 2015). Focusing on these keys applications, Binnemans et al. (2013) roughly estimated a REE potential recycling in 2020 ranging from 5600 to 10700 tons, which could significantly contribute to the overall supply of rare earth elements. In this regard, the recovery of REEs from end-of-life products is beneficial in both economic and environmental terms: the extraction of rare earths from native minerals is indeed extremely challenging as they are often found dispersed and mixed with radioactive elements (Jha et al., 2016).

In European countries the recycling of WEEE is a legal obligation and the recovery of metals from WEEE is currently achieved through mechanical and metallurgical treatments: the formers are mainly used as pre-processing in order to physically upgrade the material contents, while the latter ones, relying on techniques coming from the metallurgical sector namely pyro- and hydrometallurgy, act as refining processes (Cui and Zhang, 2008, Khaliq et al., 2014). However, the existing technologies for the recovery of metals from WEEE primarily focus on precious and base metals. In the last years several research efforts have addressed the recycling of REEs from batteries, phosphors and magnets through hydrometallurgical treatments (Binnemans et al., 2013, Tunsu et al., 2015). Moreover, a number of European research projects related to the recovery of REEs from end-of-life products are currently ongoing (Tunsu et al., 2015) and an increasing patenting trend in this area has been reported as well (Tsamis and Coyne, 2015). Nevertheless, the industrial scale applications are still limited (Binnemans et al., 2013, Tsamis and Coyne, 2015, Tunsu et al., 2015).

Over the recycling chain, the mechanical treatments play a key role as their separation efficiency ensures the material concentration in the output streams destined to further recovery processes (Chancerel et al., 2009, Meskers and Hagelüken, 2009, Meskers et al., 2009). During mechanical processes WEEE is selectively dismantled in order to separate re-usable parts and hazardous components. Then, well-established selection technologies, such as shredding, magnetic separation, eddy current and density separation, are used with the main aim of separating metals from non-metals (Khaliq et al., 2014). The effectiveness of the sorting process is affected by the type and the combination of the adopted selection techniques as well as by the physical characteristics of the waste material (Cui and Forssberg, 2003, Meskers and Hagelüken, 2009, Sun et al., 2015, Veit et al., 2002). Mechanical treatments are recognized to efficiently recover common metals such as iron and copper while precious metals are often lost (Bachér et al., 2015, Chancerel et al., 2009, Cui and Zhang, 2008, Lu and Xu, 2016, Oguchi et al., 2012, Veit et al., 2002). Although the effectiveness of the mechanical processes in separating metal fractions from polymers as well as in obtaining metal-concentrated fractions has been widely investigated, the fate of rare earth metals in WEEE pre-treatment stages has not been addressed yet. The comprehensive understanding of this aspect is, however, essential to pursue the effective recovery of these elements from WEEE, as the novelty that marks the concept of critical raw materials has not yet allowed the development of established methods (Van Eygen et al., 2016).

This study aims at evaluating the separation efficiency of valuable and critical metals in WEEE mechanical treatments. The distribution and the concentration of both base and special metals in the output fractions of mechanical pre-processing are discussed and the quality of the obtained materials is pointed out in the view of their further refining. Wider considerations on technical implications are finally underlined.

2. Materials and methods

2.1. Sampled materials

For the purposes of this study a full-scale WEEE treatment plant operating in the South of Italy was considered. The plant is designed to treat 2 tons per hour of waste coming from small electronic equipment, IT and consumer appliances. The input WEEE is processed through a mechanical treatment line that mainly enables the separation of recyclable metals from plastic fractions.

The process line consists of a two-stage shredding pre-treatment in order to reduce the particle size of the incoming WEEE below 20 mm. The shredders are followed by magnetic separators that sort out iron scraps from the waste stream before entering the subsequent section. This section includes a selection device, covered by patent, which basically forces the shredded waste to high speed, so that the composite scraps bump into the device wall and plastics flake off, while metals form small grains as a consequence of these impacts. Plastics and metal grains are then further separated via sieving, density-based fluid bed separation and electrostatic separation. Due to their different size and density properties, the combination of these selection techniques provides, moreover, the separation of the metallic grains into two fractions, one mainly composed of copper and the other one of aluminium. Through the entire line, a process air filtration is provided using bag filters and dust particles are collected as well. Five output fractions are obtained at the end of the process:

i. ferrous scraps, accounting for about 50% by weight of the incoming waste. This fraction is destined to material recovery;
ii. dust fraction, around 5–8% of the processed WEEE. This fraction is disposed of in landfill;
iii. plastic material, representing almost the 30–35% of the input WEEE. It is sent to incineration as its great heterogeneity limits its effective recycling;
iv. copper grains, accounting for 3.5–6% of the treated waste and destined to further refining processes;
v. aluminium grains, corresponding to 2.5–4% of the incoming waste and sent to recycling processes.

Dust, plastic material as well as copper and aluminium grains were sampled for the experimental activity, according to the Italian regulation for waste sampling UNI 10802:2013. The sampling campaigns covered one year, in which 8500 tons of mixed small WEEE were processed. The grab samples consisted of granules and dusts. As the regulation UNI 10802:2013 defines these fractions as solids for which the number of increments is not influenced by the material size, the sample quantities were based on the experimental purposes. However, not less than 1 kg of each fraction was collected from the plant stockpiles using a composite sampling method and sent for material characterization and analysis. Large iron scraps were not included among the sampled fractions as the visual inspection provided their classification as ferrous materials. Sampling points are indicated in Fig. 1, plotting the general flowchart of the mechanical process under investigation.

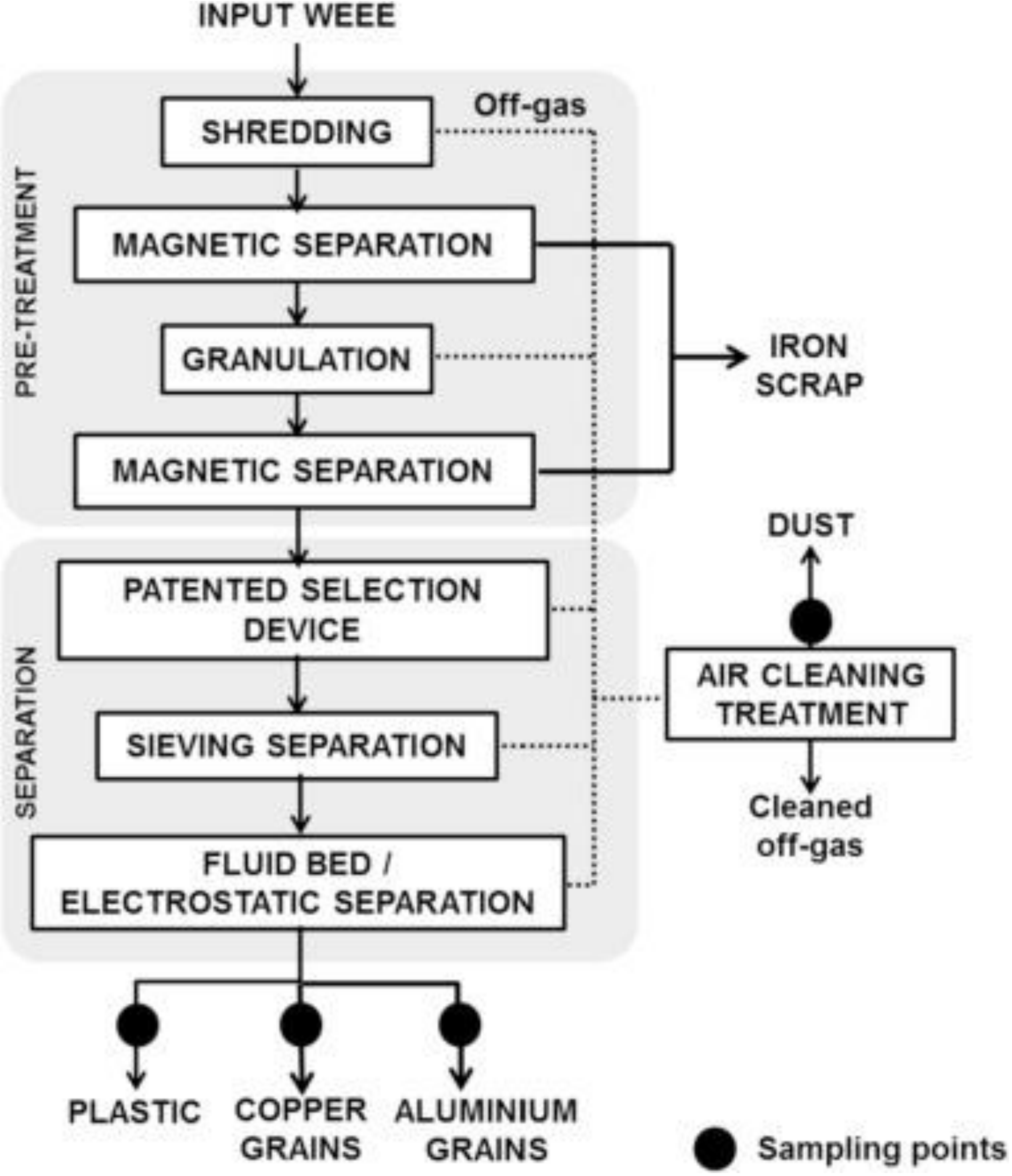

Fig. 1. General flowchart of the mechanical process under investigation.

2.2. Material characterization

Representative samples for laboratory analysis were obtained from the primary samples (1 kg) by means of a quartering sample size reduction. For each output fraction under investigation, test portions of 3 g were analysed for their metal content, using the aqua regia extraction standard procedure ISO 11466:1995 as aqua regia has been proved the best reagent for rare earths and other trace metal determinations (Hobohm et al., 2016). The concentration of selected base metals (aluminium, cadmium, copper, iron, nickel, lead and zinc), precious metals (gold, silver, palladium, platinum) and rare earth elements(cerium, europium, lanthanum, neodymium and yttrium) was determined by means of using inductively coupled plasma-optical emission spectroscopy (ICP-OES, Thermo iCap 6000 series).

Metallic granules were further characterized in terms of size distribution by employing ASTM Retsch testing sieve series. According to previous literature studies (Veit et al., 2002, Zhang and Forssberg, 1997), the metallic granules were then separated into three different size fractions: F1 (smaller than 1.18 mm), F2 (between 1.18 and 2 mm) and F3 (larger than 2 mm). The metal content was thus analysed in each fraction using the aqua regia digestion procedure in order to point out the quality of the output products by means of the metal concentration. The distribution of the selected metals into the three size fractions was thus obtained based on the metal concentration and the mass of each fraction.

Each analytical determination was done at least in triplicate.

2.3. Mass flow analysis and recovery yields

A mass flow analysis was conducted to evaluate the performance of the mechanical process (Chancerel et al., 2009, Oguchi et al., 2013, Ueberschaar et al., 2017) with regard to the selected metals.

The following assumptions were considered (Cesaro et al., 2017):

- dust particles originated from the different treatment units end up in a single flow, whose composition was considered as representative of the entire line;
- large iron scraps were entirely classified as ferrous materials by visual inspection and the concentrations of other metals in this fraction were considered to be negligible.

The mass flow of the metal *i* concentrated in the output flow *j* was obtained by multiplying the concentration of the metal *i* detected in output flow *j* with the corresponding mass of the flow *j* (Chancerel et al., 2009):

$M_{ij}= C_i \cdot M_j$ [1]

with: M_{ij} = mass flow of metal *i* concentrated in flow *j*, mg;
C_i = concentration of metal *i* in flow *j*, mg/kg;
M_j = mass of flow *j*, kg.

The mass flow analysis provided the distribution of the input elements into the output processed fractions (Ueberschaar et al., 2017).

The separation efficiency was thus evaluated determining the potential recovery yield (Christensen, 2011) of the metal *i* in the flow *j* through the following equation:

$R_{ij} = M_{ij} / M_{i\text{-input}} \cdot 100$ [2]

with: R_{ij} = recovery yield of metal *i* in the flow *j*, %;

M_{ij} = mass flow of metal *i* contained in the flow *j*, mg;

$M_{i\text{-input}}$ = mass of metal *i* in the input, mg.

The mass of the metals in the input WEEE was determined as sum of the metal contents in the outputs (Oguchi et al., 2012).

3. Results and discussion

3.1. Material characterization

The average metal content detected in the investigated fractions is summarised in Table 1.

Table 1. Average metal content of the investigated fractions expressed as mg/kg.

Element		**Input WEEE[a](100%*w/w*)**	**Output fractions**			
			Aluminium grains (2.5–4% *w/w*)	**Copper grains (3.5–6% *w/w*)**	**Plastic (30–35% *w/w*)**	**Dust (5–8% *w/w*)**
Common metals	Al	31981.7	332172.9 ± 32812.5	4296.6 ± 624.7	48756.4 ± 1805.3	35436.3 ± 7454.4
	Cd	40.1	65.7 ± 14.7	35.1 ± 19.3	61.5 ± 22.4	196.4 ± 4.4
	Cu	60289.6	192350.2 ± 4626.6	761106.4 ± 88513.0	14997.7 ± 1711.4	26624.2 ± 3496.2
	Fe	504424.0	24089.6 ± 1804.2	31353.8 ± 1435.9	1829.2 ± 116.2	12423.1 ± 4086.6
	Ni	712.4	5257.4 ± 392.1	5956.1 ± 2198.6	195.3 ± 13.4	1028.4 ± 97.8
	Pb	3742.8	20449.2 ± 1458.3	2273.0 ± 223.1	2537.7 ± 32.9	24705.1 ± 5953.8
	Zn	8463.4	73216.7 ± 5033.2	71944.2 ± 2018.1	1824.9 ± 241.7	7926.4 ± 3110.0
Precious metals	Au	5.1	35.0 ± 11.7	21.6 ± 13	4.6 ± 1.0	11.0 ± 3.7
	Ag	32.4	102.6 ± 28	204.4 ± 29	23.8 ± 13.5	105.5 ± 13.0
	Pd	3.0	22.3 ± 12.3	6.8 ± 1.9	2.3 ± 0.5	11.7 ± 9.5
	Pt	4.1	<1.3	2.8 ± 0.8	12.1 ± 1.2	<1.3
Rare earth elements	Ce	4.4	1.7 ± 0.7	<0.1	3.0 ± 0.25	42.5 ± 5.9
	Eu	0.5	<0.1	0.1 ± 0.0	0.2 ± 0.1	5.1 ± 0.7
	La	4.5	1.4 ± 0.4	1.4 ± 0.1	2.2 ± 0.1	45.8 ± 5.7
	Nd	5.6	1.9 ± 0.9	0.4 ± 0.1	3.0 ± 0.3	56.6 ± 5.4
	Y	6.2	0.2 ± 0.1	0.1 ± 0.0	1.9 ± 0.1	69.6 ± 6.2

[a] Calculated values.

Metals are recognized to be the dominant fraction by weight in WEEE (Widmer et al., 2005). Results of the present study confirmed that iron (Fe), copper (Cu) and aluminium (Al) are the prevalent metals contained in WEEE as extensively reported in other studies (Meskers and Hagelüken, 2009, Widmer et al., 2005). The average content of these metals determined in the input WEEE (around 504 g/kg Fe, 60 g/kg Cu and 32 g/kg Al) was found to be roughly consistent with the values shown by similar investigations (Morf et al., 2007, Oguchi et al., 2012). However, high standard deviations were recorded among the sampling campaigns as the wide heterogeneity which characterizes the electronic equipment generally results in a varied metal composition. Besides the extreme variability of the input materials, the standard deviation values are also linked to the uncertainties of the measurements which can be referred to the sampling method and the chemical analysis (Chancerel et al., 2009). In this regard, no standard methodologies are currently available for sampling, sample preparation, chemical analysis and statistical evaluation of heterogeneous matrices such as WEEE. Thus, further research efforts should focus on harmonizing procedures for improving the quality and the accuracy of data (Morf et al., 2013, Ueberschaar et al., 2017).

Precious metals and rare earth elements were found in input WEEE at concentrations of several mg/kg, confirming their presence as trace elements. Although used in a variety of electronic components, gold (Au), silver (Ag), palladium (Pd) and platinum (Pt) are reported to be relevant in printed circuit boards (PCBs), where can be found in concentrations much higher than that in ores (Ghosh et al., 2015). The concentration of these precious metals has thus been mostly quantified in PCBs, whereas few quantitative data are referred in literature to the precious metal content in other components, as rather reported in the present study. Nevertheless, the average concentration values of precious metals determined in the incoming flow and reported in Table 1 (32 mg/kg Ag, 5 mg/kg Au, 3 mg/kg Pd) were consistent with the study of Chancerel et al. (2009), who estimated around 67.7 g of Ag, 11.2 g of Au and 4.4 g of Pd per ton of WEEE classified in the collection group of IT, telecommunications, and consumer equipment.

As shown in Table 1, around 5 mg/kg of REEs were estimated in the input WEEE, in agreement with the investigation of Oguchi et al. (2012). However, it is worthy pointing out that data on REE content in WEEE are rather fragmented. Although these critical metals are widely used in electronics sector due to their specific physical or electro-chemical characteristics (Bakas et al., 2014), trade secrets covering specific devices still hinder the information retrieval on their content in electronic appliances (Buchert et al., 2012).

As can be seen in Table 1, the higher concentrations of base metals were detected in the metallic output fractions, namely aluminium and copper grains, as the mechanical treatments are primary designed to separate and concentrate these metals. Compared to the input WEEE, the average content of Cu and the average concentration of Al were one order of magnitude higher in the output fractions of copper grains (760 g/kg) and aluminium grains (330 g/kg), respectively. Relevant concentrations of metals were also found in the plastic output flow as well as in the dust one, which are both not involved in the metallurgical process of recovery. This evidence was particularly relevant for REEs which were found in the dust stream in concentrations one order of magnitude higher than in input WEEE. Such condition entails that critical materials are reasonably lost during the mechanical pre-treatment of WEEE.

3.2. Separation efficiency of mechanical treatments

3.2.1. Metal distribution in output fractions

A mass flow analysis was carried out in order to assess the performances of the mechanical treatment under investigation in terms of metal separation efficiency. The share of the analysed metals in the output fractions, expressed as percentage, was plotted in Fig. 2.

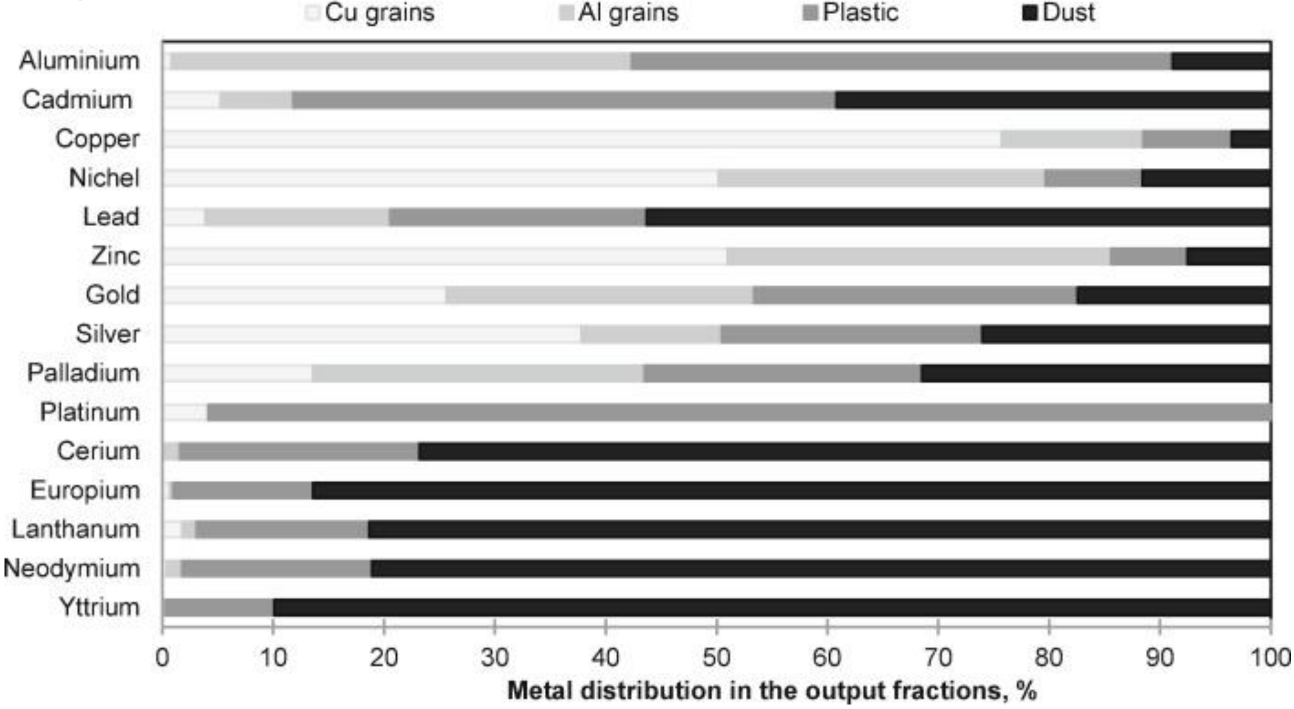

Fig. 2. Distribution of selected metals in the output fractions.

Experimental results showed that Cu and Al were differently concentrated in the metallic output grains, which are typically destined to further metal recovery. Approximately 40% of Al present in the input WEEE was distributed in the aluminium grain fraction.

However, about half of the input content of this metal was found in the plastic stream. This outcome suggested a poor separation of this metal, which ended up into a fraction not involved in the metallurgical recovery process. On the other hand, as highlighted in Fig. 2, more than 70% of the total Cu was sent to the corresponding output fraction, so as to ensure a relevant recovery of this metal. The different degree of separation observed in the present study for both Al and Cu may be related to the form in which each metal is present in the input material. Most metals are mainly present in WEEE as alloys or encapsulated in multi-material agglomerates. As this condition seems to occur more for aluminium than for copper, as reported by the investigation of Sun et al. (2015), it can result in a high liberation degree for Cu and a medium one for Al after the crushing process, which in turn affects the subsequent separation efficiency (Quan et al., 2012).

Approximatively half of gold, silver and palladium contained in the input WEEE was found to be distributed in the metallic output fractions, while relevant losses were recorded for platinum, which is part of the platinum-group metals (PGM), even clustered as critical raw materials (European Commission, 2014). As can be observed in Fig. 2, around 95% of platinum was gathered in the plastic stream not destined to metal recovery. Gold and palladium are usually present in WEEE as small contactors or multi-layered ceramic compounds whereas platinum is mainly used in switching contacts (relays, switches) or as sensors (Cui and Zhang, 2008). These elements can be either easily broken off by shredding processes or remain attached to other components (Meskers and Hagelüken, 2009, Meskers et al., 2009). Precious metals would, thus, end up in dusts as it happened for gold and palladium or would turn out in wrong material streams as for platinum. In the latter case, the relevant presence of Pt in the plastic stream demonstrated that it was not effectively liberated after the crushing processes but it was likely prone to stick to plastic residues. The mechanical action of pre-treatments resulted, therefore, unfavourable for precious metals.

As shown in Fig. 2, REEs were similarly found to be almost distributed in the collected dust particles actually destined to landfill.

The potential recovery yields for both precious and critical metals are outlined in Fig. 3 as aggregate flow data. Only a third of precious metals, including gold, silver, platinum and palladium, was sent to the copper metallic fraction from which they can be effectively recovered as the existing metallurgical processes for the recovery of copper are designed to recover precious metals (Chancerel et al., 2009). Instead, a considerable amount of precious metals ended up in either plastic (32%) or dust fractions (24%), leading to material losses roughly comparable to that reported by other studies (Bachér et al., 2015, Chancerel et al., 2009). Peeters et al. (2013) found even losses of around 90% resulting from the mechanical treatment of printed wired boards of liquid-crystal display (LCD) TVs.

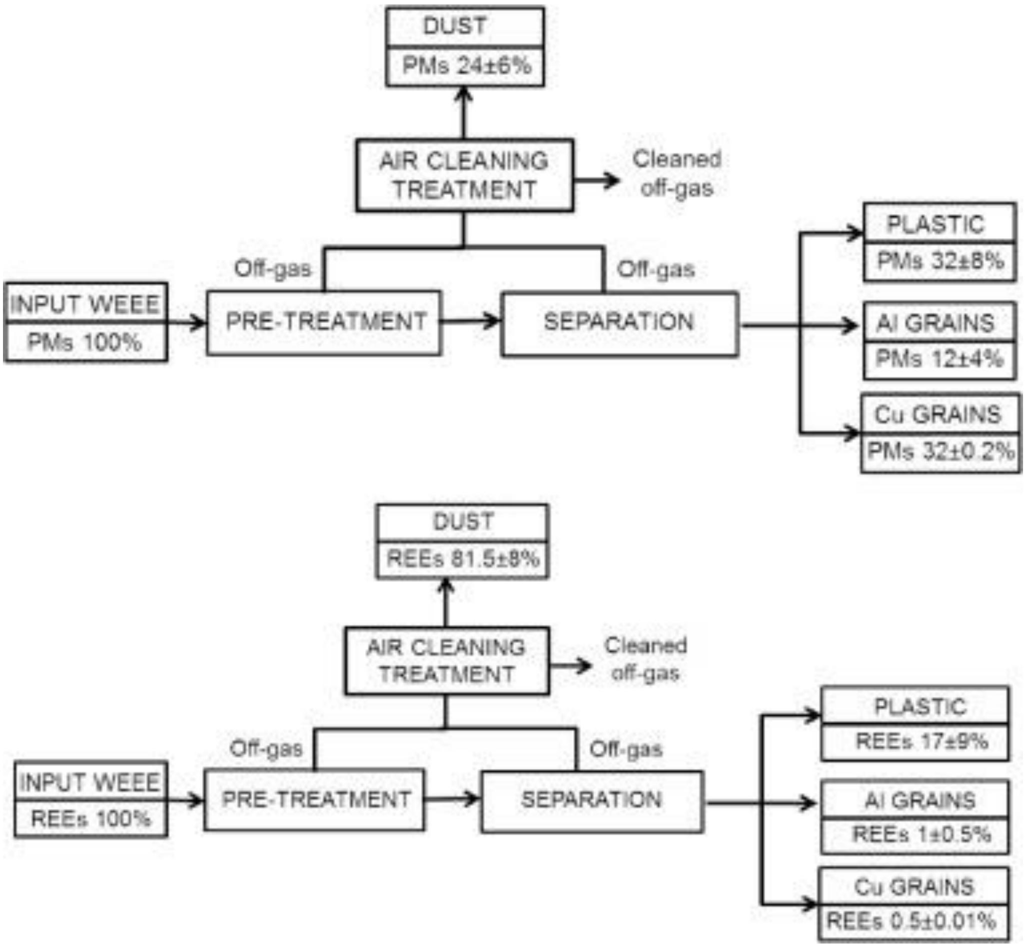

Fig. 3. Flows of precious metals (PMs) and rare earth elements (REEs) throughout the WEEE mechanical treatment under investigation.

Regarding REEs, less than 2% of these elements was concentrated in the potentially recyclable metallic fractions while more than 80% of all the REEs contained in WEEE was lost through the dust stream. In this regard, Habib et al. (2015) demonstrated the complete losses of REEs contained in hard disk drives (HDDs) in form of NdFeB magnets, tracking their flow in a conventional shredding-based WEEE treatment plant in Denmark. Indeed, REEs have a dissipation rate of over 90% due to the material losses that can occur across their life-cycle stages, ranging from their extraction/manufacturing to their final disposal, either into the environment or in other material flows or in landfills (Zimmermann and Gößling-Reisemann, 2013).

This analysis is, however, challenged by relative high uncertainties ascribed to large differences in composition of the input appliances and the sampling methods performed for the study (Chancerel et al., 2013). In mass flow analysis the uncertainties are often ignored or restricted to a qualitative evaluation although systematic approaches need to be developed for improving the reliability of results. These methodologies are still missing (Laner et al., 2014) and thus the data obtained from the conducted analysis should be considered as reference values (Chancerel et al., 2013), addressing the fate of critical metals in WEEE pre-processing. For instance, this condition is particularly relevant for Neodymium which is largely used for NdFeB magnets present in HDDs (München and Veit, 2017). As these components have a brittle nature, after shredding processes they are easily converted into dust particles that retain their magnetic properties due to the presence of iron, thus remaining attached to ferrous surfaces (Habib et al., 2015). However, the magnets account for about less than 3% of the HDD total weight (München and Veit, 2017) and computers are not the predominant fraction in the mixed WEEE entering the process under investigation.

As highlighted by the curves reporting the cumulative weight of either copper or aluminium grains against the particle diameters, the aluminium fraction showed a coarser granular composition compared to the copper one: around 90% of the aluminium grains was smaller than 8 mm, while 90% of the total weight of copper granules had a size smaller than 2 mm. This evidence endorses the separation by size of the two output metallic fractions, as the finest one (<1 mm) is mainly composed of copper, whereas the remaining portion (up to 15 mm) mostly consists of aluminium.

As previous studies have reported that different kind of base metals can be liberated at different size fractions (Cui and Forssberg, 2003, Ghosh et al., 2015, Guo et al., 2011, Menad et al., 2013, Veit et al., 2002, Zhang and Forssberg, 1997), the metallic granules were separated by size into three fractions, which were further chemically analysed for their metal content, in order to point out the quality of the output fraction with particular regard to the effect of particle size to the process performances.

For each metallic granular output, the detected concentrations were used to calculate the distribution of the investigated metals into the different size fractions (Fig. 5).

As can be observed in Fig. 5, the fine fraction of aluminium grains (<1.18 mm) showed the higher concentrations of Al and precious metals, while Cu was mostly concentrated in the coarse fraction (>2 mm). Similarly, Cu and precious metals were mostly concentrated in the fine fraction of copper granules, which even presented the higher concentration of Al in comparison with the coarse fraction. The small portion of REEs which ended up in the metallic granules, avoiding to be lost through dusts, was distributed in the finest fraction as well (Fig. 5). Moreover, as also highlighted by the metal characterization of the metallic granules shown in Table 1, these fractions were mostly composed of aluminium and copper but also other metals were present. These impurities consisted mainly of iron and nickel: iron was primarily concentrated in the coarse fraction of both metallic granules while nickel was mostly found in the coarse fraction of aluminium grains and in the medium one of copper granules.

The physical characteristics of the waste material, such as particle size and shape, are recognized to strongly affect the selectivity and the separation efficiency of mechanical processes (Cui and Forssberg, 2003, Sun et al., 2015, Veit et al., 2002). A comminution below 2 mm has been reported to be sufficient to achieve a complete liberation of copper particles (Zhang and Forssberg, 1997). Although an intensive size reduction is generally related to an increasing degree of the material liberation (Guo et al., 2011), it is also responsible for a greater production of dusts, and thus for major losses in terms of valuable materials. Moreover, the size of particles can itself limit the effectiveness of the separation techniques as they are characterized by a workable size particle ranges which can decrease the process selectivity (Zhang and Forssberg, 1997). However, besides particle size, the particle shape has also an influence on the separation process (Veit et al., 2002) as well as the design and the type of the electronic device and the separation technique performed (Bachér and Kaartinen, 2017).

In this study, the influence of particle size on process performances and, thus, the quality of the output fractions is toughly related to the adopted treatment technologies. The high concentration of Al in the coarse fraction of copper grains, as well as the high concentration of Cu in the coarse fraction of aluminium grains, underlined that the fraction with a diameter greater than 2 mm, namely the coarse fraction, is characterized by larger impurities. As hypothesized in our previous study (Cesaro et al., 2017), this condition may be mainly associated with the process that occurs in the patented device, in which the detachment of plastics from metals is achieved. During this process, plastics flake impacting the device walls, while the metals crumple forming grains which can incorporate quantities of impurities proportionally to the granule size, lowering the quality of the relative output fraction. Thus, the coarse fraction turns to be affected by relevant concentrations of other metals and this is more evident for the aluminium stream since it is characterized by grains with a larger size, as showed by the particle size distribution (Fig. 4). As a consequence, the finest fractions are characterized by a major degree of purity. Moreover, the distribution of both precious metals and REEs mostly in the fine portions of the metallic output stream, although in low concentrations, indicates that refining treatments could focus on the fraction with a particle diameter lower than 2 mm.

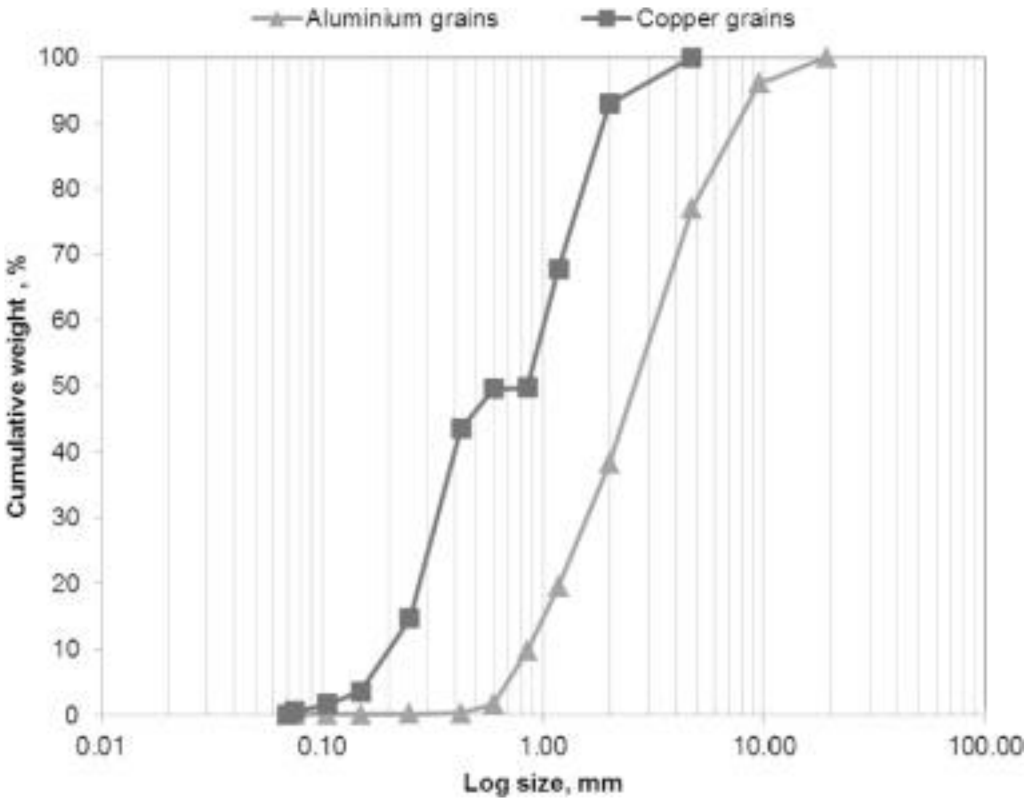

Fig. 4. Particle size distribution of the metallic output fractions.

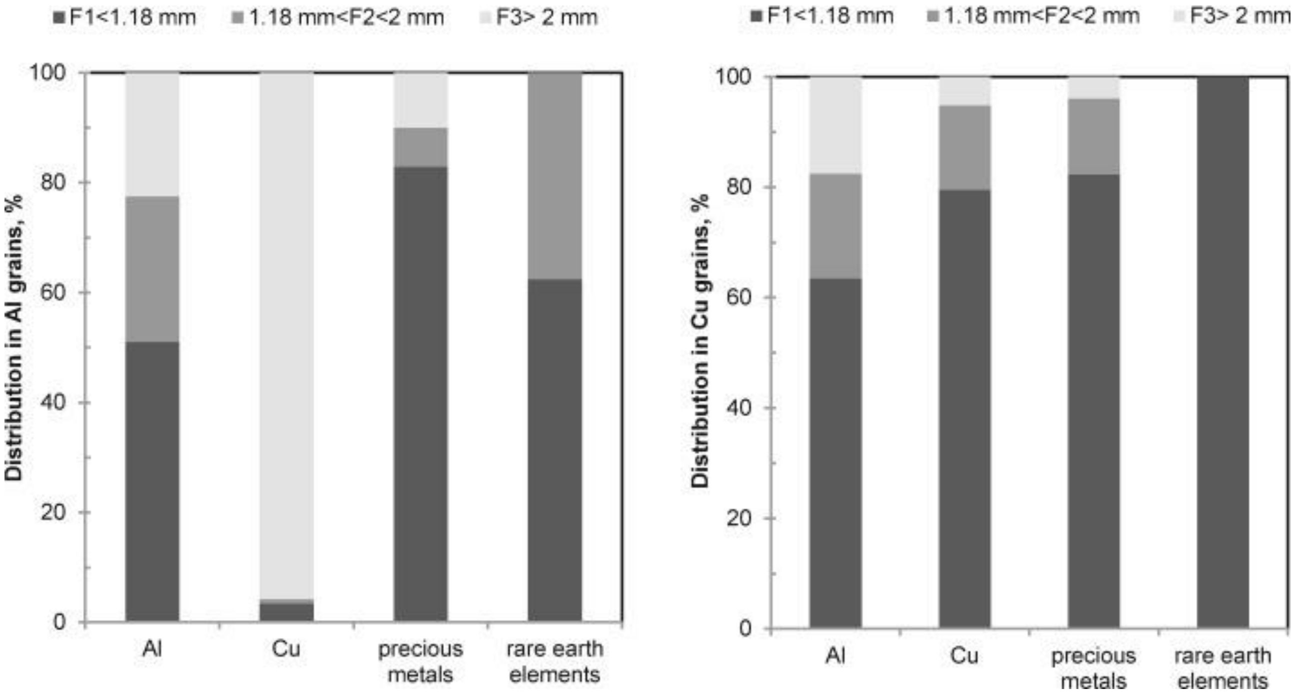

Fig. 5. Distribution of selected metals as function of size range in Al grains (left graph) and Cu grain (right graph).

4. Technical aspects

This study confirmed that conventional mechanical treatments enabled an efficient separation of base metals, whereas lower recovery yields were provided in terms of both precious metals and especially REEs. The latter were observed to be subjected to significant losses as they were mainly conveyed in the dust stream originating from mechanical units. These losses can be mainly related to the shredding technology that gathered the liberated materials into the small particles (Bachér et al., 2015, Chancerel et al., 2009). Although a comminution is necessary to achieve a liberation of the metals, mechanical processes, such as shredding and granulation, affect negatively the recovery of precious metals and REEs from WEEE, as losses can occur in the form of dusts. In this regard, the selective manual disassembly of valuable components appears to be desirable if compared to mechanical automated processes, but the benefits in terms of output quality have to be weighed up against the increasing cost of the process, as more time is spent and a lower treatment capacity is persuaded (Meskers and Hagelüken, 2009). Besides the selective dismantling, the recovery of valuable materials could be clearly improved by eco-design strategies which address the design of the electronic device for its easier recycling (Ardente et al., 2014). In this view, both selective disassembly and eco-design can be regarded as upstream-oriented strategies which set the beginning of pipe for the effective recovery of the valuable materials contained in WEEE. Otherwise, the losses of critical and valuable materials could be handled via downstream or “end of pipe” strategies, implemented at the end of material processing. In this regard, the treatment of the dust originated from mechanical processes could be addressed in order to recover the strategic materials which end up in this waste stream: the detected concentration of valuable metals as well as the relatively lower variability in terms of metal concentrations of the dust fraction in comparison with the input material could reasonably lead to efficient and sustainable recovery operations. Although the dust collected from a mechanical treatment line accounts for low mass percentage, it represents an operating cost for the plant as this fraction is actually destined to landfill disposal (Bachér and Kaartinen, 2017, Wang et al., 2015). Nevertheless, dust fraction appears worthy of reuse (Wang et al., 2015) since the recovery of critical metals, which are mainly conveyed in this waste stream, is particularly attracting due to their price, low availability and high demand. In this respect, because of the relative low concentrations of critical metals in dust materials, a biometallurgical treatment for the recovery of critical materials would be desirable as the low cost of the process makes its application of relevant interest for low grade materials (Beolchini et al., 2012). However, the benefits related to the recycling system cannot be limited to the recovered materials but additional aspects should be considered, such as environmental impacts and toxicity control associated to recycling practises (Cesaro et al., 2017, Nelen et al., 2014).

5. Conclusions

Nowadays the recycling of WEEE is regarded as a challenge due to its heterogeneity in terms of components as well as the presence of both hazardous substances and critical metals, whose recovery represents a charming driver for WEEE recycling. Mechanical treatments cover the first step of the WEEE recycling chain and therefore their effectiveness has a key role for further refining processes.

The present study addressed the recovery of rare earth elements from WEEE during the mechanical treatments which are conventionally applied in Europe.
The separation efficiencies of WEEE mechanical treatments were investigated assessing the metal mass balances thought the treatment line with particular emphasis on critical metals.

The metal characterization of samples collected at a full-scale treatment plant confirmed the presence in WEEE of both precious metals and REEs at trace level concentrations (mg/kg), while iron, copper and aluminium were found as the prevalent metals. A mass flow analysis proved that considerable amount of precious metals (68%) ended up in output fractions not involved in the recovery process and showed that around 80% of the REEs contained in the incoming WEEE was concentrated in the dust fraction originating from process air cleaning. In the metallic output flows both precious metals and REEs were mainly associated to the fine fraction (<2 mm), which revealed less impurities.

The experimental activity pointed out that the components containing precious metals and REEs can be easily pulverized using crushing processes. In this respect, the reduction in critical metal losses resulting from mechanical treatments could be pursued by setting up proper upstream or “tail-end” strategies. In this view, both the dust stream and the fine metallic fraction derived from WEEE mechanical treatments appeared as potential target matrices to be further processed via refining treatment, such as hydro or biometallurgical techniques, for valuable and critical metal recovery from WEEE.

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