

WEEE in Serbia: Status of pre-treatment methods and its influence on the recovery of critical metals

Berežni I^{*}, Marinković T¹, Stanisavljević N¹, Batinić B¹

¹Department of Environmental Engineering, Faculty of Technical Sciences, University of Novi Sad, Novi Sad, 21 000, Serbia

*corresponding author email: isidoraberezni@uns.ac.rs
telephone: 066/90-46-956

Abstract

Waste electrical and electronic equipment (WEEE) represent one of the fastest growing waste streams in the world, with an annual growth rate of 3% to 5%. In addition to containing potentially hazardous substances, it also contains valuable secondary raw materials which can be recovered by adequate recycling and recovery treatment. Pre-treatment is a key step in e-waste management to ensure the efficiency of subsequent processes and the quality of output materials. Currently, conventional recycling methods are largely based on recovering ferrous and non-ferrous metals, plastic and glass, but majority of critical metals and rare earth elements are lost during the pre-treatment processes. In this paper, on the example of Serbia, and using the material flow analysis (MFA), the current status of WEEE and pre-treatment methods are presented. Further, an overview of the presence of certain critical metals (CM) in most relevant categories of electronic equipment is given and general recommendation for avoiding CMs losses within the WEEE treatment chain are discussed.

Keywords: electronic waste, pre-treatment, recycling, material flow analysis, critical metals

Introduction

During the past decades, rapid technological progress resulted in a huge consumption increase of electrical and electronic products. As a result of the expansion of the electronic market and the shorter lifespan of many electronic devices, a record of 53.6 million metric tonnes (Mt) of e-waste was generated and reported in 2019, with projection of growth up to 74.7 Mt by the year of 2030 [1]. In addition to growing quantities, its quite complex composition, both containing valuable materials and harmful substances makes WEEE management a challenge [2].

When discarded, electronic and electrical devices present considerable environmental and health challenges, much higher than other types of waste, e.g., around 40% of heavy metal contamination in landfills comes from e-waste [3]. Containing up to 60 elements in composition, both precious metals (gold, silver, and platinum group metals) and critical metals such as selenium, gallium, tellurium and indium, EEE constitutes a considerable fraction of the total demand for metal resources [4]. Precious and critical metals present essential constituents of many electronic products, in particular IT and communication equipment. From a tonne of discarded mobile phones (without batteries) can be obtained up to 300g of gold, representing a much higher yield than most efficient gold mines [5]. Further, the concentration of precious metals in polychlorinated biphenyls (PCBs) is usually much higher than in ores. If compare mind ores for the extraction of gold and palladium which contain less than 10g/t of precious metals with concentration in PCBs of personal computers of 250g/t of gold and of 110g/t of palladium it became obvious how important recovery of these metals from e-waste is [6]. Moreover, environmental impacts of the secondary production operations are much lower than primary production. The extraction of precious metals through mining have negative environmental impact which includes significant emissions of greenhouse gases and energy, water and land usage [7]. Thus, recycling chain for WEEE has to be analysed in order to access the recovery options for these valuable elements.

E-waste recycling chain consist of several steps (collection, pre-processing, end-processing) carried out by specialized operators. After the collection, pre-processing is the first and crucial step of e-waste treatment due to the fact that it determines to which recovery or disposal processes the material are fed [7]. Pre-treatment can be carried out manually, mechanically or combining those two technologies. To separate reusable parts and hazardous components WEEE is selectively dismantled during the manual and mechanical processes. Different technologies, such as shredding, magnetic separation, eddy current and density separation are used to separate metals from non-metals [8]. Mechanical properties of the liberated materials in the output as well as further

treatment steps depend on the used pre-treatment technology [9] and therefore it is necessary to develop specialized systematic approaches in certain waste streams that are composed by higher concentrations of critical metals.

The objective of this paper is to present the currently applied pre-treatment processing using Serbia as a case study. In addition, the aim is to present a concise overview on e-waste categories and products in terms of critical metals and their fate during the pre-processing phase.

Materials and methods

This paper focuses on the WEEE management in Serbia with an emphasis on the pre-treatment processes. The flows of WEEE were investigated on the country level for the selected reference year, 2020. Research studies, statistical data, reports, legislation and other public documents published by WEEE national organizations and Environmental Protection Agency were used to analyse current e-waste management system in Serbia. In addition, material flow analysis (MFA) was used for the further analysis and was carried out with the help of the software STAN. The results are given in two MFA diagrams, one of which shows the overall e-waste flows at the state level during the reference year, while the other represent e-waste flows within the pre-treatment plant focusing on output flows. In regard to this, the system boundary for the first diagram is determined by physical boundaries of Serbia and thereby only the flows and processes taking place within the borders of the country were included, while for the second diagram the boundary of the system is represent by the plant. Further, for the second goal of the paper, a literature review was performed to define high-grade e-waste products in terms of precious and critical metals and based on all, state the fate of these metals in pre-treatment processes and give the general recommendations for avoiding these losses.

Results and discussion

General WEEE management system in Serbia

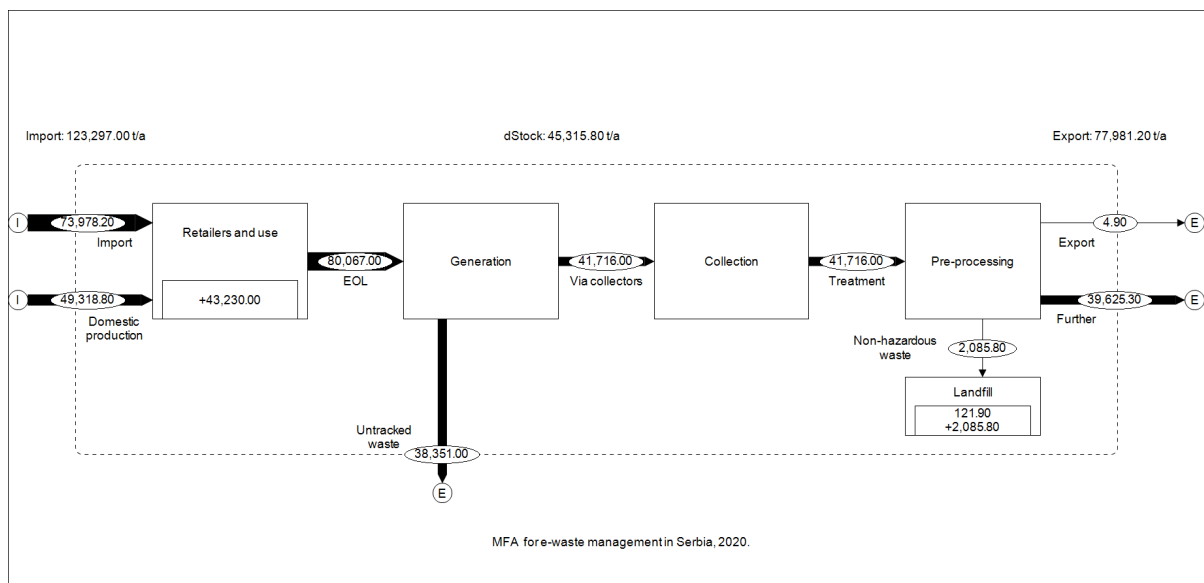
WEEE management system in Serbia consist of collection of this waste stream and its pre-treatment, while the final treatment technologies are not represented. Serbia is an EU candidate country and therefore has harmonised and adopted most of the requirements from WEEE Directive 2012/19 through national legislation. The Ministry of Environmental Protection (MEP) and Serbian Environmental Protection Agency (SEPA) are leading participants in monitoring and controlling the WEEE flows and collecting the required data. Beside these institutions, there are other participants in e-waste management like producers or importers of EEE products, which became waste after use and waste producers and operators who perform collection, transport and treatment of waste.

Producers and importers of products that after use became WEEE have the obligation to keep daily records on the quantity and type of produces and imported products. This is defined by the Law of Waste Management and by-laws and all the collected data should be submitted to SEPA in the form of reports. For that purpose, SEPA established the National Registry of Pollution sources information system (an online platform) where all the WEEE related information (e.g. daily records about the number, the mass and the type of EEE placed on the market) are collected and processed. All WEEE generators and operators must also record all annual quantities of treated/exported WEEE and send them to SEPA [10].

There are four main operators in Serbia that carry out organised collection and recycling of electronic waste. In business sector around 500 companies participates in collection of e-waste through formal sector, collecting mainly IT and telecommunication equipment. Beside this, there is significant informal network in Serbia for collection of e-waste. Those data are not represented in official statistics, but it is estimated that there are between 5,000 and 8,000 informal collectors of e-waste. Informal collectors are mainly interested in the recovery of all types of waste that contains metals. That is why some waste from electrical and electronic equipment is sold in existing markets scrap metal that often work in the “gray” zone. In addition, collectors often supply local workshops of used goods with spare parts removed from electrical waste and electronic equipment [11].

Concerning the quantities of generated WEEE in Serbia, there is no exact data, but only the estimations of around 80,000 t/year. According to the Report on the management of special waste streams in the Republic of Serbia for 2020 [12], the quantity of EEE placed on the national market amounted to 19,425 t. If compare this data with estimated generated amounts, it is clear that there is a gap in reporting due to excessive difference between the figures. Given the generated quantity, a more realistic picture of the products placed on the market should be much higher. Considering this, during the material flow input analysis, the amount of electrical and electronic equipment placed on the market between 2011 and 2020. was taken into account. Results of e-waste system management is presented in Figure 1, where low collection rates lead to scenario where almost half of generated e-waste is being untracked.

Figure 1 MFA for e-waste management in Serbia, 2020.



Last step in system management is pre-treatment from where some materials is exported for further treatment, while materials which can be used directly goes in facilities for recycling/recovery.

Applied pre-treatment processing

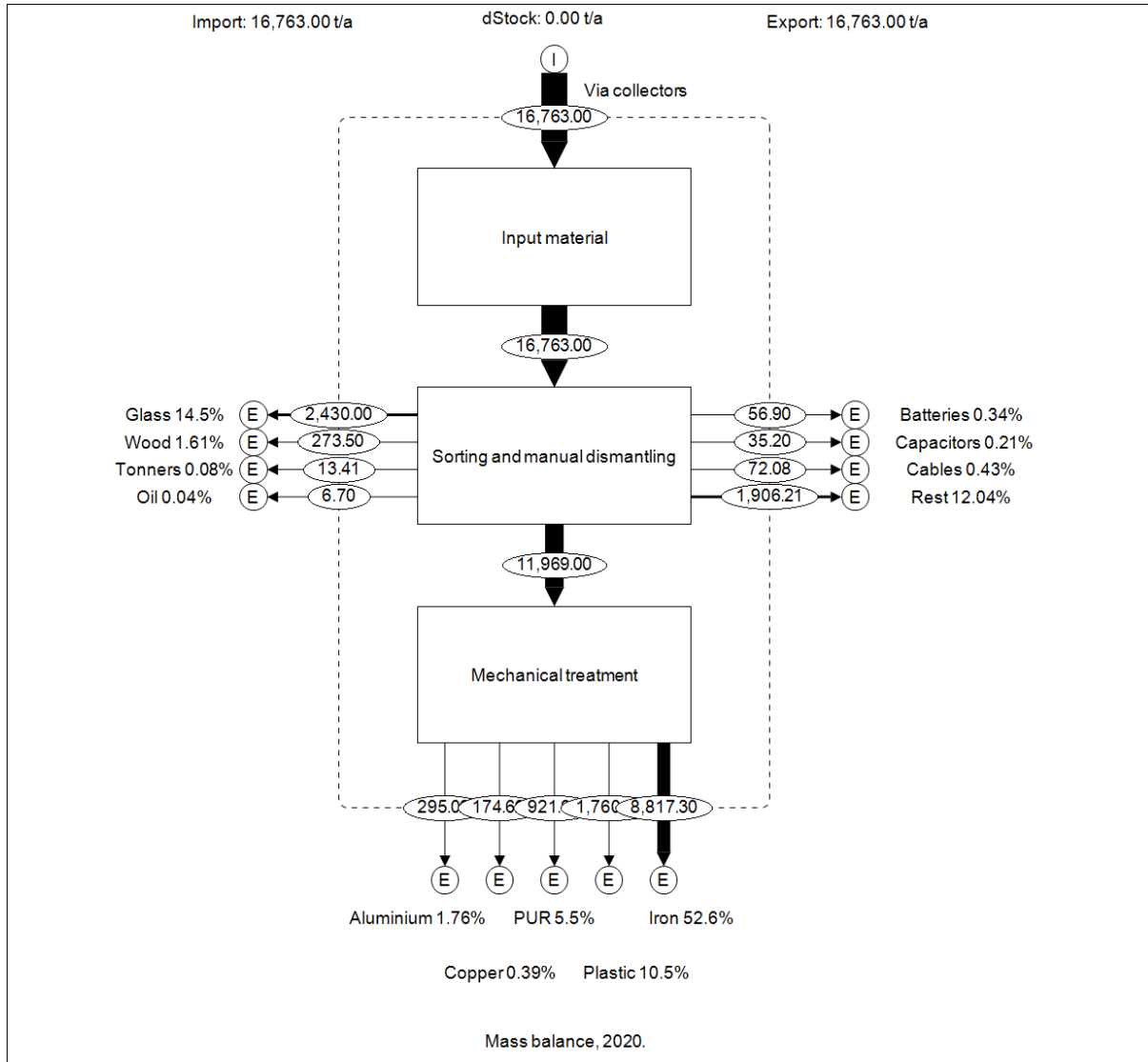
Once e-waste is collected, dismantling, pre-processing and end processing are inevitable stages for safe disposal or recycling. To ensure that liberated materials enter the appropriate recovery process pre-treatment phase is crucial.

In general, it includes manual dismantling, mechanical processes and combinations of manual and mechanical pre-processing. Manually extracted components (batteries, cables, cathode ray tubes etc.) are sent further to secondary treatment plants for further processing. Mechanical treatment consists of shredding, milling, magnetic and other types of separation, whereas refrigerator treatment plants usually have a line for Freon separation. Pre-treatment technologies that are currently in use are customized for efficient separation and recovery of “mass relevant” fraction in e-waste, mostly metals (Fe, steel, Ni, Al, Cu) and non-metallic fraction like plastic, rubber, glass and textile, while the majority of CMs are lost as they stay coupled with dominant metal output fractions, or end up in the dust from the process [10,13]. In order to more precisely present the pre-treatment technology, in this paper, a plant from Serbia was taken as a case study. Using a previously conducted study [10] and more recent data, for the selected representative year 2020, steps of pre-treatment phase are explained further in text.

As it can be seen in Figure 2 treatment involves manual separation and mechanical treatment. First step is dismantling of various components, hazardous and/or valuable such as cables, batteries, cathode ray tubes, glass etc. Some electronic and electrical devices, like TVs and CRTs that arrive in the plant are subjected only to manual separation, they are decomposed into components and then separated into hazardous and non-hazardous groups,

after which they are exported abroad for further treatment. Manual sorting and dismantling are followed by mechanical treatment of individual devices. Based on their specific physical characteristics such as weight, size, shape, density, and electrical and magnetic characteristics, materials and metals contained in WEEE are liberated.

Figure 2 Mass balance for treatment pre-treatment plant, 2020.



First phase of mechanical pre-treatment involves shredding and crushing processes in order to reduce size of input materials. This is followed by magnetic separation to separate group of ferrous metals (Fe, steel, Ni). For this purpose, low intensity magnetic drum separators are widely used. Further, non-ferrous metals (Al, Cu, etc.) are separated from other type of materials (plastic, glass, etc.), by using some of the electric conductivity-based separation technique. In this case, this step is done using the Eddy current separation, which is also one of the most commonly used in e-waste recycling processes. Finally, as 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. These techniques include water or airflow tables, heavy media floating, sifting, etc [13,14]. Beside the conventional mechanical processes, there are sensor and optical based techniques that can also be applied [15,16].

The major percentage of obtained raw materials is iron, followed by plastic, wood and aluminium. The size of the output materials leaving the plant ranges between 0.1 and 100 mm, and the raw materials are 99% pure and can be used directly.

Critical metals in WEEE and their fate in pre-treatment processes

Due to their unique physical and chemical properties, metals are an indispensable part of electronic equipment. By using an increasing number of metals from the periodic table, electronic equipment becoming more functional. Therefore, electronic equipment is considered as highly heterogeneous mixed materials comprising precious metals in printed circuit boards (PCBs), cobalt in rechargeable batteries, indium in LCD displays, etc. Demand for EEE products is increasing every year, causing the higher need for critical metals. To meet certain specifications in individual EEE products several typical critical metals are widely consumed in electronics: lithium and cobalt (rechargeable batteries) indium (LCD glasses, semiconductors/LED), REEs (permanent magnet, battery alloy), gold (PCBs, bonding wire, contacts), silver (switches, lead-free solders, conductors), platinum (hard disk, resistors, plasma display panels) etc. Beside the huge demand of critical metals, their deficiency in the Earth’s crust and existence in a few regions present serious depletion and supply risk. Thus, to address this problem, the most reasonable method is to increase the recycling efficiency of secondary resources [17].

Existing pre-treatment technology are efficient for separation and recovery of “mass relevant” fraction in e-waste, mostly metals (Al, Cu, Fe, steel) and non-metallic fractions such as plastic and glass. As a result, the majority of CMs are lost, as they stay coupled with dominant metal output fractions, or end up in the dust from the process. Therefore, it is important to know in advance the composition of the e-waste in terms of critical metals. The content of EEE in terms of precious and critical metals is given in the Table 1 [18].

Table 1 Content of EEE products in terms of precious and critical metals

Materials (g/units)	Sb	Co	Gd	Ga	Au	In	Pd	Pt	Ag	Ta	Tb	W	Y
Product													
LDC notebooks	0.77	0.065	<0.001		0.22	0.04	0.04	0.004	0.25	1.7	<0.001		0.002
LED notebooks	0.770	0.065	<0.001	0.0016	0.22	0.04	0.04	0.004	0.25	1.7			0.002
CRT TVs	14												
LCD TVs	0.71		<0.001		0.11	0.003	0.044		0.45		0.002		0.11
LED TVs	0.71		0.002	0.005	0.11	0.003	0.044		0.45				0.005
CRT monitors					0.31								1
LCD monitors			<0.001	0.003	0.2	0.079	0.04		0.52			0.663	<0.016
LED monitors			0.002	0.003	0.2	0.082	0.04		0.52			0.663	<0.001
Cell phones		3.8			0.024		0.009		1				
Smart phones	0.084	6.3			0.038		0.015	0.004	0.244				
PV panels			0.119		0.119								
Tablets	0.154	0.013	<0.001		0.044	0.008	0.008		0.05				<0.001

Products that include IT and telecommunication equipment, as well as lighting equipment are the richest of precious metals and rare earth elements. For example, indium in the form of ITO (In₂O₃ (90–95%) and SnO₂ (5–10%)) is widely used as transparent conductive films in LCDs due to their adequate electrical conductivity and optical transparency. LCDs have replaced cathode ray tubes (CRT) displays in most appliances because of the small volume, light weight and low power consumption and became leading technology in flat panel display production, which accounts for almost all cell phones, computers and 87.3% of TV. This lead to increased indium consumption, which confirms the fact that in period between 1994 to 2014 production of indium increased from 149 tons to 819 tons [17]. Further, presence of rare earth elements is ranging from smartphones and flat screen TVs to all sorts of electric motors, nickel-metal hydride (NiMH) batteries, high performance metal alloys and even automotive catalysts. Precious metals, that include gold, silver and platinum group metals have properties

such as good electrical conductivity, high melting point and corrosion resistance, and so they are ubiquitous in electronic devices, multilayer ceramic capacitors and hybridized integrated circuits. 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 [19].

As complete automation of e-waste disassembly is not currently technically feasible or cost-effective, technical optimization is needed to improve manual disassembly during pre-treatment. Precious metals and rare earth elements are confirmed to be present in WEEE input at concentrations of several mg/kg. Chancerel *et al.* [7] estimated that there is around 67.7g of Ag, 11.2g of Au and 4.4g of Pd per ton of WEEE classified in group of IT, telecommunications, and consumer equipment, while according to Oguchi *et al.* [20] around 5mg/kg of REEs are found in the input WEEE. However, as the mechanical treatments are primarily designed to separate and concentrate “base metals”, in the metallic output fractions higher concentrations of these metals were detected, mainly aluminium and copper. Relevant concentrations of some 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 is especially related to REEs, which were found in the dust stream in concentrations one order of magnitude higher than in input WEEE, clearly showing that critical materials are reasonably lost during the mechanical pre-treatment of WEEE [2]. According to Chancerel *et al.* [7] only a third of precious metals (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, while the rest ends up in either plastic (32%) or dust fractions (24%) leading to material losses. Losses of around 90% are also found resulting from the mechanical treatment of printed wired boards of liquid-crystal display (LCD) TVs [21]. As regards to REEs, less than 2% of these elements ends up in the potentially recyclable metallic fractions while more than 80% of all the REEs contained in WEEE was lost through the dust stream. Furthermore, study conducted in a conventional shredding-based WEEE treatment plant in Denmark, showed complete losses of REEs contained in hard disk drives in form of NdFeB magnets [22]. Oguchi *et al.* [20] estimated that pre-sorting of the small WEEE would reduce the percentage of Au that is landfilled in the small-grain fraction from 74% to 57%. Also, they obtained the following percentages for the amounts of metals that could be separated before shredding: Ta (48%), Ag (54%), Bi (11%), Pd (11%), and Ga (30%). Hence, separate collection and pre-sorting of small WEEE would be effective for the recovery of these metals. It is essential to remove the components that contains valuable metals, such as PCB and batteries and feed these components separately to recycling facilities. This also has been confirmed by a study of Ueberschaar *et al.* [23], where 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. In the case of gallium, which is found in smaller quantities in PCBs and LEDs, if it is subject to conventional recycling technologies, it can be diluted with other materials. Gallium rich components must be separated prior to any mechanical processing with other materials to prevent it from being transformed into slag. 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 [13, 24].

Conclusion and recommendations

Rapid technological development lead to use of large amounts of critical metals in electronic equipment. The higher demands in the future will burden the supply risk of these critical metals. As WEEE represent an important secondary resource of these critical metals, their recycling can potentially relieve the supply risk.

The efficiency of pre-treatment was found to be crucial for precious and critical metals. With appropriate handling and use of technology, losses of critical metals can be prevented. Currently applied technologies work effectively for separation of mass-relevant metals such as iron, aluminium and copper, while CMs and REEs are almost completely lost as they end up in a form of the output stream from which cannot be recovered. Thus, improvements in e-waste system management and adjustments in pre-treatment technology is needed in order to maximise the overall recovery of critical metals.

As regards to e-waste management system, more reliable and transparent information about the content of critical metals in the different equipment groups and their component is needed and within that increase the collection rates for all of these product groups. Further studies and surveys on recycling behaviour of consumers can provide insight on how collection program can be improved. By raising awareness on the importance of recycling and developing incentives for consumers to bring back their items that are no longer in use can improve collection

rates. More efforts are also needed to control implementation of the policies. Beside this, optimized structure and design of electronic products would contribute to easier manual disassembly and recycling processes.

When it comes to pre-treatment, it is essential to focus activities on improvement of manual disassembly and separation of target components in e-waste that contain high concentrations of critical metals. Outputs from pre-treatment stage must be fractions with appropriate characteristic for end-processing facilities. In order to achieve this, focus on future studies should be on developing technologies for automatic recognition, sorting, and dismantling of WEEE, in order to make recovery of CM from heterogenous WEEE flows more efficient [13]. Finally, revision of WEEE Directive and defining recycling targets based on specific material or components, is needed to avoid further losses of CMs from WEEE.

Usually, WEEE recycling is not only technological problem, but also economic and so, for the successful implementation of the above measures, economic viability of technological improvements is one of the main preconditions.

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