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Multi-Layer Ceramic Capacitors in Lighting Equipment: Presence and Characterisation of Rare Earth Elements and Precious Metals

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Multi-Layer Ceramic Capacitors in Lighting Equipment: Presence and Characterisation of Rare Earth Elements and Precious Metals

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Abstract: In this study, multi-layer ceramic capacitors (MLCCs) detached from lighting sector's waste electrical-electronic equipment (WEEE) are characterised for the presence of rare earth elements (REEs) and precious metals (PMs). Their digestion was carried out with HNO_3 & aqua regia on a heating plate and characterised with the use of inductively coupled plasma optical emission spectroscopy (ICP-OES) & scanning electron microscopy with energy dispersive X-ray spectroscopy (SEM-EDX). The contents of REEs and PMs are 0.84 wt% and 0.60 wt%, respectively, and create an economic stored value in MLCCs that is essentially defined by PMs of 98.67% and by Pd 78.37%. The content of the main elements is: Nd 0.366 wt%, Y 0.220 wt%, Dy 0.131 wt%, Ag 0.467 wt%, and Pd 0.105 wt%. The results confirm the need of selective removal and separate recycling process of MLCCs from WEEE drivers.

Keywords: Measurements and characterization; lighting equipment; multi-layer ceramic capacitors; rare earth elements; precious metals; ICP-OES analysis

1. Introduction

Electrical-electronic equipment (EEE) at the end of its life is characterised as waste electrical-electronic equipment (WEEE) or simpler electronic waste (e-waste) and classified into one of its categories according to the European Union (EU) Directive 2012/19. Environmental protection and natural resources preservation require equipment recycling. E-waste includes at least 57 elements of the periodic table, some of which have significant economic value. Therefore, e-waste can be used as an important secondary source of base metals (BMs), special metals, rare earth elements (REEs), precious metals (PMs), glass, plastic and functional structures-components, which can be reused in equipment production or repair [1–4].

REEs include Lanthanides, scandium (Sc), and yttrium (Y), while PMs include platinum group metals, gold (Au), silver (Ag), and rhenium (Re). Generally, Lanthanides include lanthanum (La), cerium (Ce), praseodymium (Pr), neodymium (Nd), promethium (Pm), samarium (Sm), europium (Eu), gadolinium (Gd), terbium (Tb), dysprosium (Dy), holmium (Ho), erbium (Er), thulium (Tm), ytterbium (Yb), and lutetium (Lu). PGMs include the elements ruthenium (Ru), rhodium (Rh), palladium (Pd), osmium (Os), iridium (Ir), and platinum (Pt). Rare earth elements and precious metals are characterised by the EU (2023) as critical raw materials and are accompanied by the indicators "supply risk" (SR) and "economic importance" (EI). The vast number of these elements makes their recovery from e-waste a significant recycling challenge [5–10]. Presence of the abovementioned elements has different impact in EEE versus WEEE. More specifically, in the first case they improve energy efficiency and enhance its protection from environmental effects, while in the second due to higher and cleaner concentrations, boost the idea of "urban mining" through recycling-recovery process, and simultaneously counteract natural reserves exhaustion (balance problem). The presence of specific valuable elements in e-waste for 2019 was: Ag 1200 t, Au 200 t, Ir 1 t, Ru 0.3 t, Pd 100 t [2,11–20]

1.1. Recycling of specific electrical-electronic components from e-waste

During the recycling process of e-waste, the pre-treatment stage aims at a) their separation and dismantling, b) the removal of "hazardous" parts which need special handling, c) separation of structures-components based on their functionality, d) selective removal of components "look and pick", based on its concentrations of specific valuable metals or critical raw materials, e) the creation of recycling flows with simple composition and high concentrations, f) minimizing of critical raw materials losses and the improvement of recovery, g) the sustainability of recycling plants, and h) the ideal balance between the economic benefit of recycling and the reduction of its environmental footprint [3,7,8,12,21–25].

Characterisation of e-waste (structures-components), regarding the presence and quantity of specific elements, is using destructive or non-destructive methods, such as **s**canning **e**lectron **m**icroscopy with **e**nergy **d**ispersive **X**-ray spectroscopy (SEM-EDX), **s**canning **e**lectron **m**icroscopy **e**nergy **d**ispersive **s**pectroscopy (SEM-EDS), **t**ransmission **e**lectron **m**icroscopy **e**nergy-**d**ispersive **X**-ray **s**pectroscopy (TEM-EDXS), **X**-**r**ay **f**luorescence (XRF), **m**icrowave **p**lasma **a**tomic **e**mission **s**pectroscopy (MP-AES), **t**hermo**g**ravimetric **a**nalysis (TGA), **i**nductively **c**oupled **p**lasma **o**ptical **e**mission **s**pectroscopy (ICP-OES), **i**nductively **c**oupled **p**lasma **a**tomic **e**mission **s**pectroscopy (ICP-AES), and **i**nductively **c**oupled **p**lasma **m**ass **s**pectrometry (ICP-MS). Method choice is a crucial parameter that affects results. The choice considers essential parameters related to the sample, such as (a) type, (b) structure, c) construction materials, and (d) concentrations. For low concentrations, plasma-related methods, e.g., ICP-OES and ICP-MS, are selected that can detect extremely low concentrations [7,22,26–32]. Electrical-electronic equipment usually include or are accompanied by drivers (Figure 1a-b) responsible for its proper operation. Not only from a functional point of view but from a recycling point of view, drivers constitute an important or possibly EEE's most important structure [3,33]. Depending on the type of equipment, their percentage weight differs significantly (~2-30%) without this being an indication of its stored economic value. Driver consists of printed circuit board (PCB) and electrical-electronic components (EECs), which are divided into through-hole components (THCs) and surface mount devices (SMDs). Drivers may include multi-layer ceramic capacitors (MLCCs), usually in SMD form. Their number and total mass are a function of the circuit's electronic design and PCB's layout. Using capacitors of this type in the design and construction of drivers offers numerous advantages regarding minimal dimensions, reliability, and lack of polarity [13,15,16,29,31,34–37]

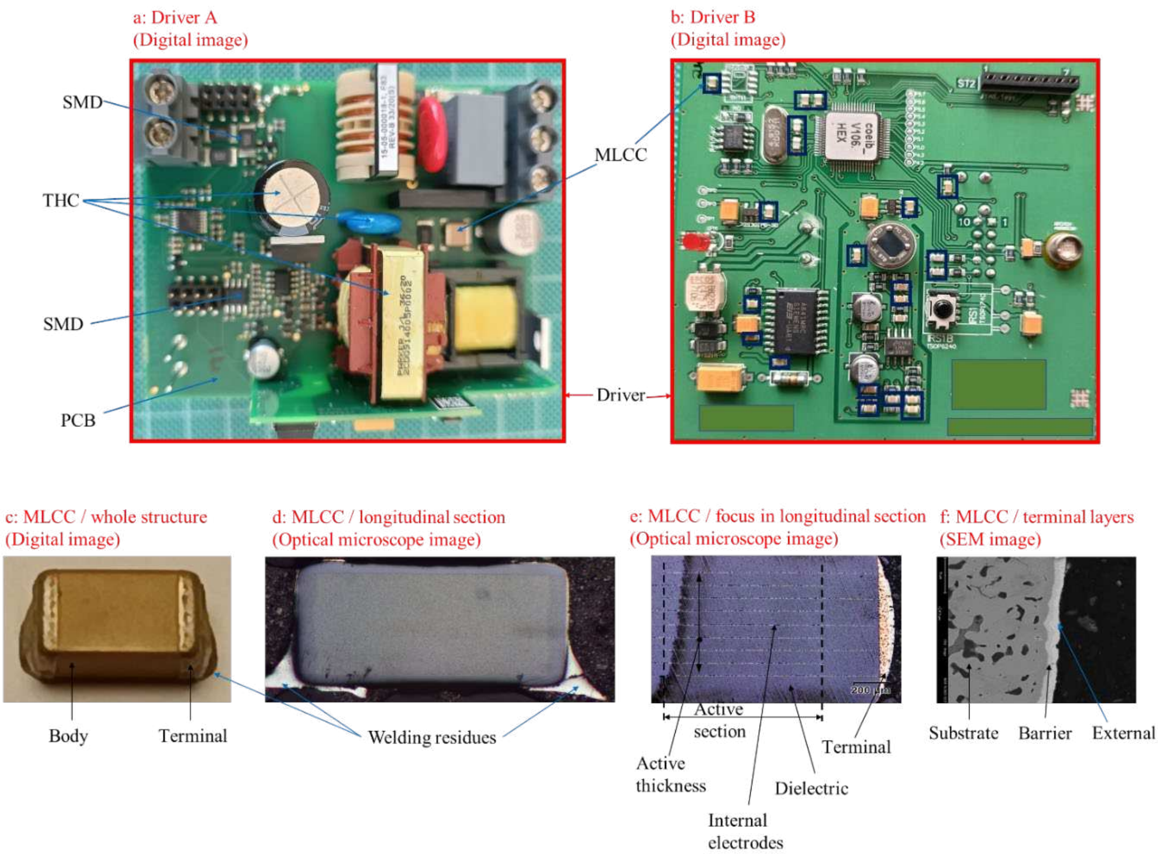

Figure 1. Digital images of drivers (a-b) and an MLCC (c), Optical microscope images of longitudinal section of an MLCC (d, e), f: SEM image MLCC / terminal layers.

Multi-layer ceramic capacitors consist of the main body and terminals (Figure 1c). The main body incorporates metal electrodes and ceramic dielectric (Figures 1d-e). Usually, terminals are formed by three layers (substrate, barrier and external) (Figure 1f). Ni, Sn, Cu, Ag, Pd, and Pt, in single or compound form, are used to manufacture electrodes and terminals. Formerly, Ag and Pd were significantly used to manufacture these structures of capacitors. In the last thirty years, the significant cost increase in their production has led to a technological shift from precious metals to base metals, thus creating BMs-MLCCs. Today, the share of BMs-MLCCs nickel (Ni) accounts for 60% of the total production of MLCCs. Despite the above treatment, there is ongoing scientific interest in creating new materials to eliminate the use of precious and critical metals for cost reduction and mitigate the "balance problem" [3,13,26,31,38–42].

This technological shift led to MLCC's critical parameters deterioration. Since precious metals concentrations were diminished, and unavoidably, the durability of capacitors was negatively affected, and consequently, the whole equipment's service life has been reduced. From a recycling point of view, this change was accompanied by valuable material contents that gradually declined throughout the years [35,42,43]. The challenges in their design, dictated by the requirement for high capacity in small volume and less precious metals use, have been achieved by using rare earth elements as impurities in forming ceramic dielectric (usually $BaTiO_3$). The adoption of these elements led to a) excellent properties related to the dielectric constant (high-k), dielectric losses (low tanδ), and electrical properties (excellent) even for thin dielectric layers (<1μm), b) high performance and reliability of base metals electrodes (BMEs) nickel-capacitors while maintaining reduced production costs, and c) the inclusion of ceramic dielectrics in their design and production [31,44–50]. Many elements in e-parts hinder their recoverability, mainly when their concentrations are extremely low. MLCCs' characterisation, separation and separate recycling make their contents recovery more effective, contributing to natural resources conservation and the electronic industry's partial independence from CRMs' mining and commercialisation monopoly [4,10,32].

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1.3. Artificial lighting and MLCCs

Lighting equipment (LE) maintains inner lighting intensity at a desired level and saves building energy. Depending on its type and specifications, lighting equipment may contain multi-layer ceramic capacitors. MLCC palladium's remarkable concentration adds stored value to drivers in such a case. Mainly due to Pd concentration, capacitors of this type is characterised as a "target component" among other electrical-electronic components of the driver for its selective removal and separate recycling process [16,18,38,42,43].

It is worth mentioning that only a few studies related to the removal and characterisation of multi-layer ceramic capacitors from the drivers of specific e-waste are available [38,51]. Based on the literature review, twelve studies were identified related to their characterization, while only six concern specific capacitors or capacitors from specific e-waste. In particular, capacitors from computer were used by [51], of a specific colour were used by [7], non-magnetic were used by [43], from personal computers were used by [38], X7R-0603 BME-MLCCs were used by [29], and Ni-rich MLCCs were used by [30,39].

The novelties of this study are a) the presence of multi-layer ceramic capacitors in specific lighting equipment and b) the identification and quantification of REEs and PMs in MLCCs. Data can be useful to suggest new recycling protocols for efficiently recovering these metals.

2. Materials and Methods

2.1. Materials

Lighting equipment used included a) light management equipment (LME) corresponding to a 240 m^2 house, b) lamps and luminaires light emitting diode (LED) technology, and c) external drivers for LED strips. The above equipment differed in terms of electrotechnical-photometric characteristics and brand name (BN) and was provided by: a) AEGEAN RECYCLING-FOUNDRIES SA (lamps, luminaires, and drivers), b) FEILO SYLVANIA GREECE SA (lamps), c) ABB Greece and SIEMENS Greece (LME).

2.2. Methodology

The methodological approach followed in this study included six stages: (1) collection and separation of the, (2) disassembly and testing, (3) sample preparation, (4) ICP-OES analysis, (5) EDX analysis, and (6) SV calculation. A detailed description of the laboratory steps from the collection of lighting equipment to the characterisation of MLCCs is presented in Appendix 1. All measurements were repeated three times under the same conditions, and the results were averaged.

2.2.1. 1st stage - Collection and separation

The 1st stage involved the collection, cleaning, visual integrity check and separation of the equipment according to a) e-waste's categories, b) its use (domestic or professional), and c) LED's technology (surface mount devices or filament) (Appendix 2). The lighting equipment corresponded to three categories of e-waste: Cat-3 (lamps), Cat-4 (large equipment), and Cat-5 (small equipment). In this study, the addition of index C to LED lamps indicates that they are lamps containing surface mount devices LEDs (SMD LEDs), while the addition R indicates that they refer to lamps containing filament LEDs. For example, the type of lamp is E27 and is modified to E27-C and E27-R.

2.2.2. 2nd stage - Disassembly and testing

The 2nd stage involved a) initial weighing of the equipment, complete manual disassembly, and weighing of its structures and components for the correlation between its masses, using basic and special tools such as magnifying glasses, precision tweezers, hot air-gun rated temperature (Brand: BOSCH, model: GHG 20-60), balances (Brand: Kern, model: EMB 3000-1, d=0.1), (Brand: KERN, model: EHA 1000-1, d=0.1), and precision balance (Brand: KERN EWJ-300-3, d = 0.001 g), b) for the reliability of the samples, and because the exterior of SMD inductors resembles MLCCs, in addition

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to the macroscopical control during the removal of multi-layer ceramic capacitors from the drivers, their digital control was performed piece-by-piece to exclude the possible presence of SMD-inductors between them, using a digital instrument identifier surface mount devices components (Brand: Mastech, model: MS8910), c) MLCCs' counting and weighing for each item of equipment to generate innovative data on their presence in it, and d) before their magnetisation check [52] a representative sample was removed for EDX analysis, while the remaining capacitors were examined using a Nd-magnet for the initial quantitative assessment of precious metals in their structure according to the methodology followed in [43].

2.2.3. 3rd stage - Samples preparation

To eliminate any possible organic composition on the examined multi-layer ceramic capacitors (e.g., conformal coating), they were burned at 700 °C. Because of this, the 3rd stage involved the preparation of the porcelain crucibles (Brand: JIPO, form: middle) to investigate the difference in mass of the samples before and after their burning, using an analytical balance (Brand: KERN, model: ABP 200-4M), d=0.0001), laboratory oven (Brand: THERMOLYNE, model: 30400), and desiccator. To burn samples more efficiently and avoid contamination, slight breaking of the capacitors was carried out in an agate mortar. Potential mass loss was testing using an analytical balance (Brand: SHIMADZU, model: AUX320, d=0.0001) and the difference was negligible. The temperature and time profiles are presented in Figure 2.

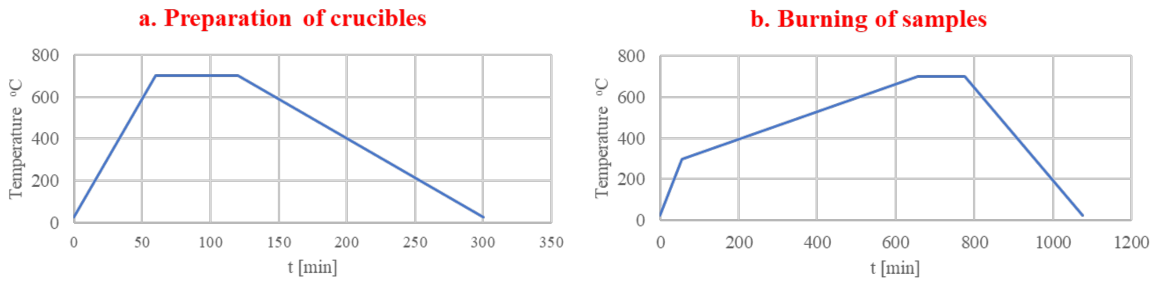

Figure 1. Temperature and time profiles a) preparation of crucibles and b) burning of MLCCs

2.2.4. 4th stage - ICP-OES analysis

The 4th stage involved a) pulverisation of capacitors to create laboratory samples (quartering - ~ 1 g per sample) and make most effective approach of acids in samples, according to the methodology followed in [1,28,38], using a ball mill (Brand: FRITSCH, model: pulverisette 6) with a zirconium oxide planetary ball mill tank (Brand: FRITSCH, volume: 80 ml), and analytical balance (Brand: SHIMADZU, model: AUX320, d=0.0001), b) digestion of the samples was carried out in glass beakers with a solid/liquid ratio 1:40 and on a heating plate at 90-100 °C by manual stirring for a period of 3 h or for as long as required for their digestion. Their digestion was carried out in a chemical fume hood, implementing personal and environmental protection regulations. The determination of Ag and REEs was carried out in nitric acid (HNO_3) and of the remaining PMs in aqua regia ($HCl:HNO_3$ - ratio 3:1), according to the methodology followed in [7,22,24], using acids HNO_3 65% and HCl 35-38%, all analytical grade, (Brand: MACRON and were purchased from Chemix SA), according to the methodology in [1,7,15,53,54], c) calibration of the ICP-OES instrument, using ICP calibration standards: a) REEs (Brand: CPA hem, name: MISA Standard 1 - Rare Earth Metals - 18 components), b) PMs (except Ag), (Brand: CPA hem, Name: MISA Standard 2 - Precious Metals - 6 components, and c) Ag (Brand: hps, name: ICP-AM-MISA6 - 27 component, d) filtration of solutions to protect the ICP-OES instrument, using filter paper (Brand: Ahlstrom-Munksjö, type: Hardened Low-Ash Grades: 391), and ultrapure water (Brand: PanReac, type: LC-MS), and e) ICP-OES solutions analysis to quantify REEs and PMs in samples, according to the methodology followed in [31,51], using the ICP-OES instrument (Brand: Agilent, model: 5110). The ICP-OES instrument was calibrated using five aqueous standard solutions: 0.5, 2, 4, 10, and 20 (mg L^{-1}) for REEs and PMs except Pm, Re, and

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Os. The operating parameters of the ICP-OES instrument were the same for HNO_3 and aqua regia: Radio frequency power 1300 W, Nebulization gas flow 0.8 (L min^{-1}), Auxiliary gas flow 0.2 (L min^{-1}), Plasma gas flow 15 (L min^{-1}), Sample aspiration rate 1.5 (L min^{-1}), and Torch configuration Axial.

2.2.5. 5th stage - SEM-EDX analysis

The 5th stage involved the SEM-EDX analysis of nine multi-layer ceramic capacitors. The capacitors were encapsulated in conductive resin, and their longitudinal sections were subjected to friction and polishing according to the methodology followed in [55], using a grinding and polishing machine (Brand: Struers, model: rotopol 35), optical microscope (Brand: ZEISS, model: HAL 100 with an OLYMPUS DP22 camera), and SEM-EDX (Brand: Thermo Scientific, model: Phenom XL).

2.2.6. 6th stage - Calculation of stored value

The stored value due to the presence of rare earth elements and precious metals and their percentage distribution in an assumed mass of 1 kg of MLCCs from lighting equipment was calculated considering only their usable concentrations of ICP-OES analysis results and their current market prices.

3. Results and Discussion

3.1. Estimation of the chronology of lighting equipment

LE manufacture date is essential information for the recycling sector for two main reasons: a) concentrations of e-waste are linked to the manufacturing technology of the respective period, b) PMs and REEs concentrations change over time. To estimate the chronology of the LE collected in 2021, the following parameters were taken into account: a) the protocol EU-1194/2012 for determining the lifetime of LED lamps, b) the Directive 2009/125/EC and the Regulations 244/2009, 1194/2012, c) the efficiency (lm W^{-1}) of the examined lamps and luminaires compared to their new type counterparts (Appendix 3), concerning the "net zero scenario" 2010-2030 to improve their performance (~ 4 lm W^{-1} per year), (d) the average lifetime specified by the manufacturer, (e) the effects of temperature on the photometric and electrotechnical characteristics of lamps as a function of luminaire types, f) various imponderables, such as failure of materials and components and power line grid surges, (g) average daily operation six hours for household lamps and twelve hours for professional equipment (lamps-luminaires) according to Commission Regulation-EU 1194/2012/ANNEX II, [56–63]. Considering the abovementioned parameters, it has been estimated that the manufacturing date of the examined LED lamps and luminaires is between 2016 and 2021. Based on matching current equipment and sponsor information, the manufacture date of a) the external LED drivers is between 2018 and 2021, and b) the LME is between 2012 and 2020.

3.2. MLCCs in lighting equipment

The average mass per type of lighting equipment, the mean percentage correlation of their masses and in particular between a) driver and LE and b) SMD-EECs and driver are presented in Appendix 4a-c, respectively. External LED drivers, semi-closed type, and G9-R, R7S-R LED lamps, had no multi-layer ceramic capacitors and thus were not investigated further. The absence in the first type is due to the driver's electronic design, whereas in the other two, no driver is required to operate

The experimental and calculated results of the SMD-MLCC's existence in each examined type of lighting equipment are shown in Figure 3. More analytically, the probability of their presence in each type of lighting equipment is shown in Figure 3a. The presence and their number for each type of electrical-electronic equipment are interrelated concepts. In particular, LE's capacitors are significantly smaller than their corresponding number in other e-waste. More thoroughly contained, LED lamps 1-7, LED luminaires 2-7, external LED drivers closed-type (ELEDDCT) 9, and LME 9-60.

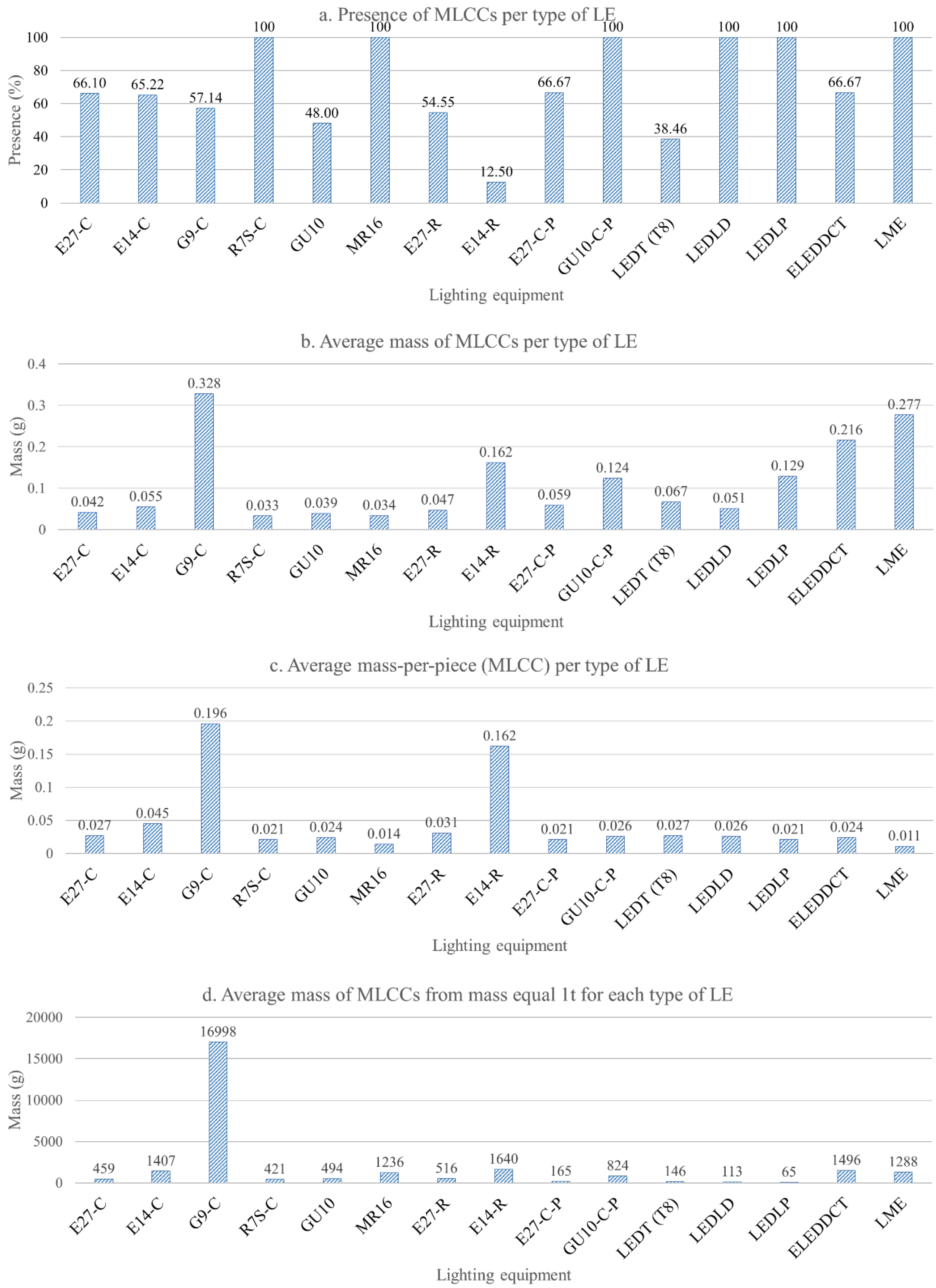

Figure 3. a. Presence of MLCCs in LE, b. Average mass of MLCCs per type of LE, c. Average mass-per-piece of MLCC per LE, d. Average mass of MLCCs per 1 t of LE.

On the other hand, their number in specific equipment corresponds approximately to: TV sets 300, LED TVs and personal computers greater than 1000, personal digital assistants 200, and mobile phones 150-200. Smartphones are the type of equipment with the highest demand of these capacitors between the EEE, with their number depending on their BN and specifications, changing significantly [18,26,28,35,38,48,64]. According to [38] drivers have a multitude of these capacitors in their structure. [23] report that in a classic hybrid driver design, their number, as a percentage versus the number of EECs, corresponds to about 30%.

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The recycling sector focuses on the presence and total mass of capacitors as a function of the types of e-waste. The average capacitor mass per examined lighting equipment type is shown in Figure 3b, highlighting G9-C lamps as the most efficient type with a value that coincides with the maximum of the total configuration that ranged between (0.033-0.328) g. We need to underline that there is no study related to MLCCs' total mass calculation or estimation and no valid results to compare.

Each of the capacitors included, differs in mass, and is characterized by an average mass-per-piece (MpP) value for each type of examined lighting equipment. How high or low the MpP factor is, makes their manual extraction from drivers, profitable or not. Mass per piece value stems from the division of their total mass, and the results are shown in Figure 3c. The product of the two aforementioned factors, i.e., capacitors number × average MpP, contributes to the characterization of their stored value in any of the e-waste. We used the following equation to find the largest capacitors' mass among 1 t of each type of the examined lighting equipment.

$$\text{Mass of MLCCs} = \left(\frac{A}{B}\right) \cdot \text{C} \cdot \text{D}$$

where A is an assumed fixed mass of each type of lighting equipment (in this study, 1 t), B is the average mass per piece of each type of examined lighting equipment (see Appendix 4a), C the presence probability of multi-layer ceramic capacitors in each type of examined lighting equipment, and D the mass of multi-layer ceramic capacitors per piece of examined lighting equipment.

The calculated results highlight LED lamps G9-C as the most advantageous type among lighting equipment used in this study for the collection of multi-layer ceramic capacitors (see Figure 3d).

3.3. Characterisation of MLCCs from lighting equipment via ICP-OES analysis

The calculated results of concentrations and contents of examined multi-layer ceramic capacitors and their necessary accompanying information for the recycling sector are presented in Figure 4.

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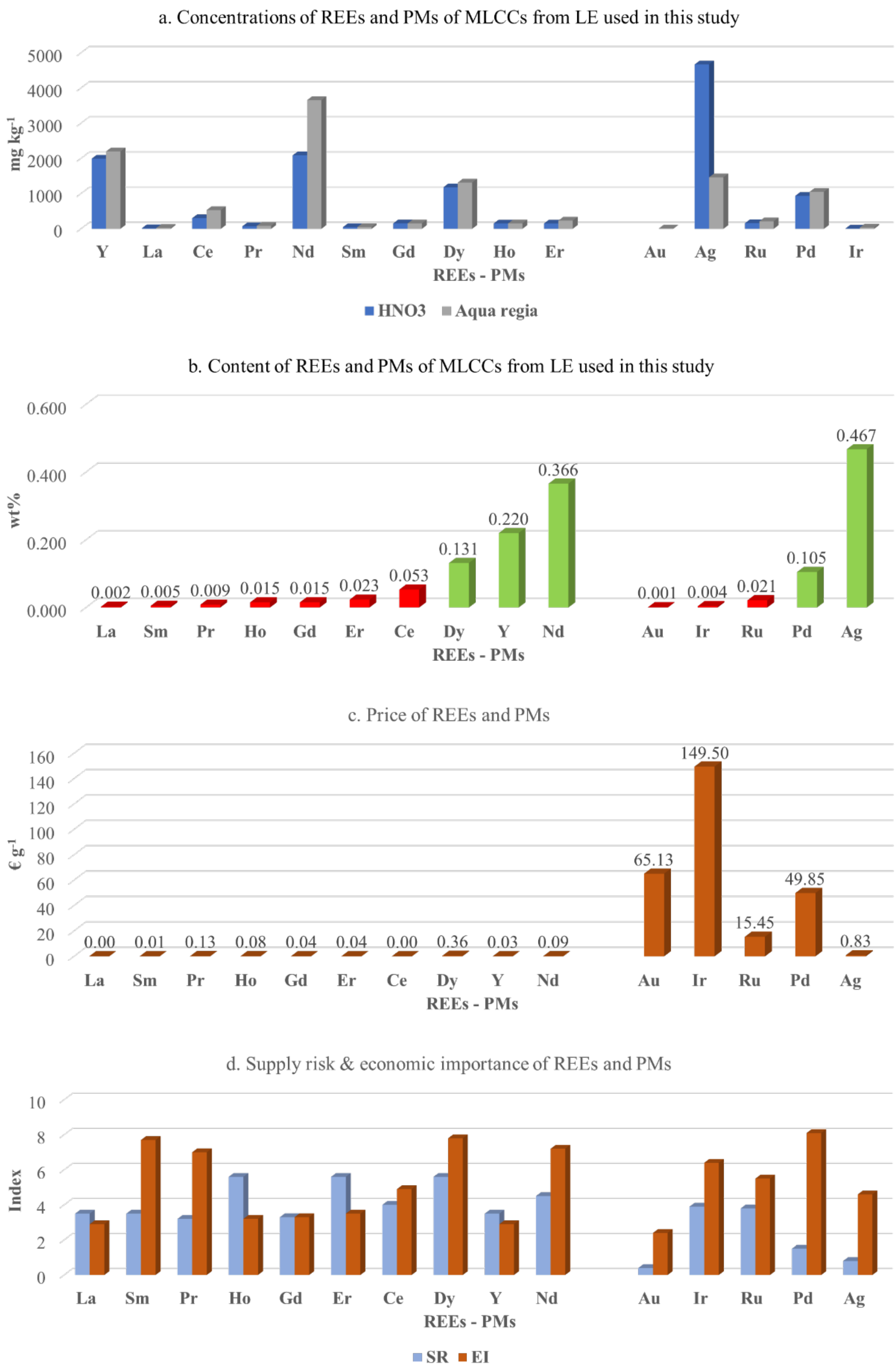

Figure 4. A. Concentrations of REEs & PMs in MLCCs from LE – HNO_3 vs Aqua regia, b. Concentrations of REEs & PMs in MLCCs from LE, c. Content of REEs & PMs in MLCCs from LE, d. Stored value in MLCCs of mass 1 kg due to the presence of REEs and PMs.

Based on the ICP-OES analysis results (mg L^{-1}), the usable concentrations (mg kg^{-1}) (HNO_3 vs aqua regia) of rare earth elements and precious metals are presented in Figure 4a. Sc, Tb, Yb, Lu, and Pt concentrations were out of the quantification limit, and the elements Eu, Tm, and Rh were not detected. The results are a function of the sample specifications and adhesive residues on capacitors'

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terminals. MLCCs' ceramic body colour indicates the presence of specific elements in their structures. In particular, brown is associated with Y, red, purple, and blue with Pd, gold with Ag, and off-white with Ir. The synergy of their chromatic and magnetic separation techniques will create even more specific recycling flow process [7].

The sample consisted of different sizes and colours of magnetic capacitors derived from all the lighting equipment used. The percentage of each type of LE in the sample was: LED lamps (domestic use 45.43%, and professional use 9.53%), LED luminaires 2.84%, ELEDDCT 3.98%, and LME 38.22%.

According to [49,50], dielectric ceramic materials with rare earth elements impurities are used in multi-layer ceramic capacitors. Indicatively, in the following studies are mentioned: **Y** [65], **La** [66], **Ce** [67], **Pr** [68], **Nd** [69], **Sm** [34], **Gd** [13], **Dy** [35], **Ho** [29], **Er** [70], **Yb** [47], and **Lu** [47]. Apart from the REEs, presence of PMs is witnessed in the following studies: **Au** [51], [24], **Ag** [15,24], **Ru** [55], **Pd** [24,26], and **Ir** [7].

As expected, Au's leaching took place only in aqua regia. Ag leaching was 52% higher in HNO_3, while the leaching of the rest of precious metals and some rare earth elements was higher in aqua regia (Ru 14.13%, Pd 5.71%, Ir 61.13%, Y 4.86%, La 14.81%, Ce 27.34%, Pr 7.24%, Nd 27.27%, Dy 5.31% and Er 21.05%). Gd, Ho, and Sm showed small to insignificant differences ranging between (0.40-2.10%) as a function of the acids used. Due to the nature of the sample, the low concentration of Au may be explained as a) its main use of Au in electrical-electronic equipment focuses on the internal structure of integrated circuits and the protection of electrical contacts from environmental conditions such as oxidation; b) gold-plated terminals' MLCCs are used in specific and high cost applications, so their potential presence in the sample is minimal, with external LED drivers and LME being the most likely origin [12,19,71]. Increased concentrations of Ag and Pd may be explained as multi-layer ceramic capacitors is one of the main electrotechnical applications of Ag and, simultaneously, the main electrotechnical application of Pd. Because of this, Pd's concentration in drivers is a function of MLCCs' presence in their electronic circuit. Also, depending on their composition, the welding residues on the terminals of waste-MLCCs form the concentration of Ag [8,15,18]. [31] report that no Pd was detected in their sample examined via ICP-OES analysis. Considering the highest value among the concentrations of each element, MLCCs' contents were calculated (Figure 4b). La, Sm, Pr, Ho, Gd, Er, Ce, Au, Ir, and Ru appear as trace elements (content <0.1%). In contrast, the rest of the investigated elements constitute their main composition. REEs content corresponds to 0.84 wt% and PMs to 0.60 wt%. Based on the literature review, the results of relevant studies, the analytical technique used, and the sample's specifications are listed in Table 1.

Table 1. Results of analysis based on literature on the presence or absence of REEs & PMs in MLCCs structures.

Au wt%	Ag wt%	Pd wt%	Y wt%	Specific (MLCCs or WEEE)	Analytical technique	Ref.
< 0.0001		1.69		Yes [1]	ICP-OES	[51]
0.01	0.13	0.05		No	MP-AES	[22]
	3.48	1.24		No	XRF, ICP-MS	[26] *
	1.99	1.10		No	SEM-EDS	[28]
			(0.3 & 0.8)	Yes [2]	ICP-MS	[7]
	(205-968) mg L^{-1}	(38-193) mg L^{-1}	OoQL	Yes [3]	ICP-AES	[43]
	1.08	0.14		Yes [4]	AAS	[38]
	5.01	0.95		No	XRF, ICP-AES	[18] * [23] *
			68.92	Yes [5]	TEM-EDXS	[29]
	2.02	0.35		Yes [6]	ICP-AES	[30] *, [39] *
< 1	(1-20)	< 1		No	-	[24]

* (data on the main composition of MLCCs). (1) computer, (2) specific colour's MLCCs, (3) non-magnetic MLCCs, (4) personal computers, (5) X7R-0603 BME-MLCCs, (6) Ni-rich MLCCs.

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Part of these studies lists the main composition of the rare earth elements and precious metals of the multi-layer ceramic capacitors they used. Differences between Table 1 results and this study, are possible due to individual or a mix of the following parameters: a) WEEE's type and category, b) brand name, specifications and manufacture date of equipment, c) sample's technology, specifications, and representativeness, and d) laboratory process stages and the analytical technique used [7,15,19,29,31,35,72]. This study includes, the results of presence of eleven additional elements to the ones listed below, nine REEs, and two PMs.

Considering the elements' contents, their market prices, and SR and EI indicators (Figs 4c-d) respectively, as well as the low REEs' recycling rate (<1%), it is clarified that it is imperative try to recover even the traces of the critical raw materials contained in the e-waste, due to their unique properties, natural monopoly, environmental protection, and the economic interest [9,73–77].

3.4. EDX analysis of MLCCs from lighting equipment

EDX-analysis of examined multi-layer ceramic capacitors, indicatively presented in Figure 5, confirmed the existence rare earth elements and precious metals, according to [43]. "Standard terminations" MLCCs (ST-MLCCs) (Figure 5a) and "flexible terminations" MLCCs (FT-MLCCs) (Figure 5b) were found [78,79]. ST-MLCCs terminal's structure (Figures 5a1-a3) confirmed the presence of BMs and in particular tin (Sn), nickel (Ni), and copper (Cu), according to [18,38,39,47].

Of particular interest were the presence of FT-MLCCs in lighting equipment because they are usually used in equipment of significant cost or in equipment subject to vibrations. Specific materials such as conductive polymers were used before 2008 as an additional (fourth) terminals' layer to protect the sensitive ceramic body of multi-layer ceramic capacitors against mechanical stress (Figure 5b). Mechanical stress is likely to be generated by a) vibrations due to careless use of the equipment (poor placement or falls), b) during and because of its operation, and c) local temperature changes created during the manufacture or repair of drivers. Silver conductive polymer (Figure 5b4) aims to absorb mechanical stress and contribute positively to equipment lifetime by capacitors' protection.

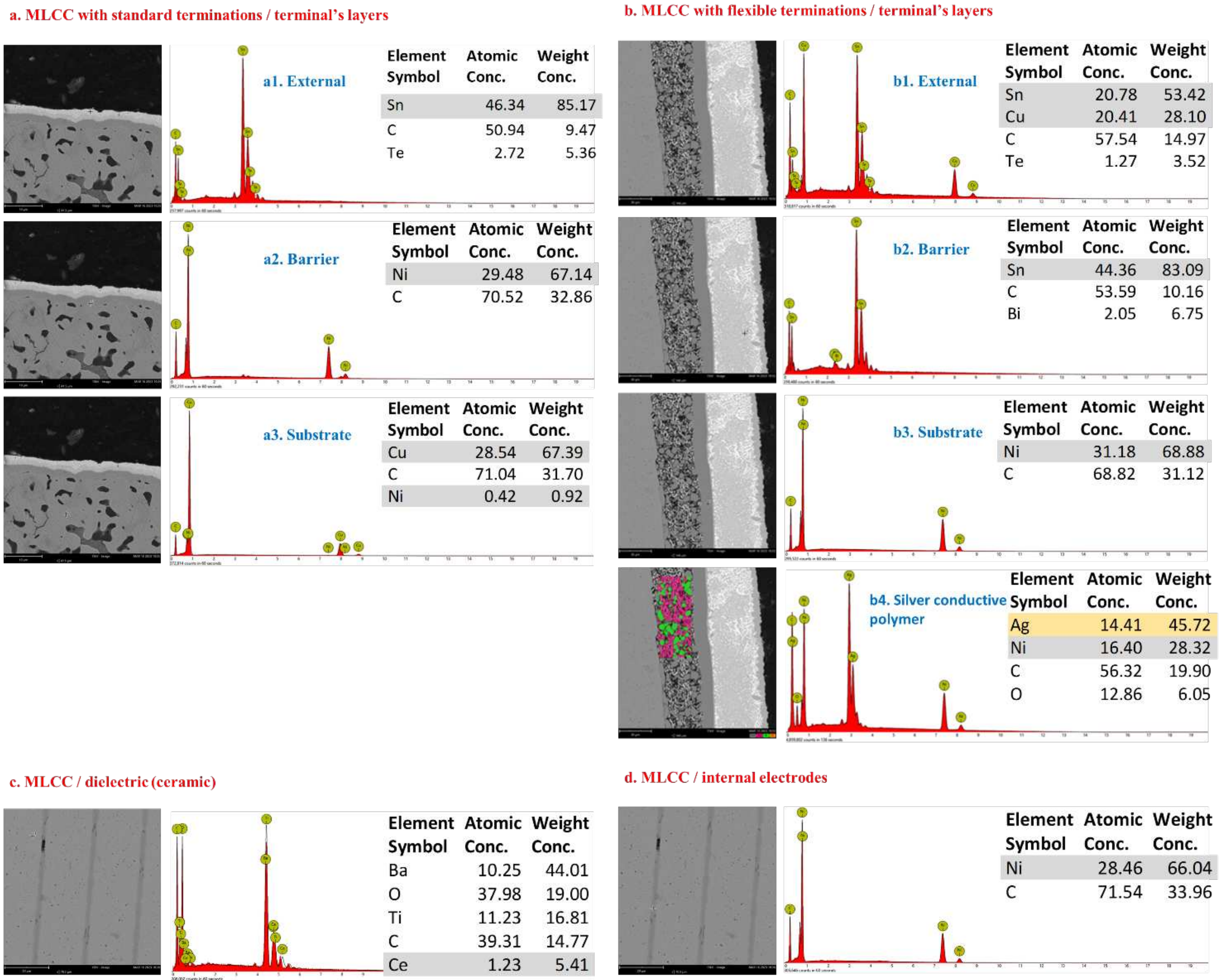

Figure 5. a. MLCC with standard terminations / terminal layers (three), b. MLCC with flexible terminations / terminal layers (four), c. MLCC / dielectric (ceramic), d. MLCC / internal electrodes
.

From the recycling point of view, this additional layer contributes to the concentration of Ag [35,47,79]. Ce, barium (Ba), and titanium (Ti) were detected in ceramic dielectric (Figure 5c), and Ni in electrodes (Figure 5d). Precious metals were not detected in their electrodes, even though Pt and Ag-Pd alloy (70-30) were used for manufacture [16,26,40,41,47,67].

REEs-PMs content of examined multi-layer ceramic capacitors corresponds to 1.44 wt%; therefore, 98.56 wt% corresponds to BMs, TMs, and ceramic dielectric. This significant percentage of weight may include the following elements: Sn 1-2.57%, Cu 1.76-8%, Ni 0.68-66.1%, Ba 17.4-48.56%, Ti 11.4-19.36%, lead (Pb) 1.03-8.82%, bismuth (Bi) 0.18-0.42%, iron (Fe) 3.71%, zinc (Zn) 0.19%, manganese (Mn) 0.23%, aluminium (Al) 0.15%, chromium (Cr) 0.31%, and niobium (Nb) 3.48% [18,22,23,26,30,31,38,39,41].

4. MLCCs as target components and their store value

The ideal-futuristic recycling is related to the individual characterisation of e-waste structures and components and their separate recycling [3]. All concentrations of the examined multi-layer ceramic capacitors, were significantly higher than those of the Earth's crust, while excluding elements (La, Sm, Pr, Ho, Gd, and Er) are also higher than natural mining [80]. Critical raw materials are located in specific structures or components of electrical-electronic equipment [81]. For both MLCCs, SMD-LEDs, and tantalum capacitors (TCs), scientific interest exists for their separate recycling because of their CRMs and TMs contents. Comparing their concentrations about specific elements (Figure 6) shows that multi-layer ceramic capacitors lag significantly behind in Au and Y, are comparable for Ag, and outperform significantly for Ce and Gd. It is worth noting that the literature review shows that SMD-LEDs and TCs do not contain Pd [7,72,81–91].

Calculation and distribution of stored value for an assumed mass of 1 kg of multi-layer ceramic capacitors from lighting equipment due to the presence of rare earth elements and precious metals was performed, taking into account only their usable concentrations and current market prices. The results are presented in the Sankey diagram (Figure 7), where REEs' extremely low participation in stored value is observed. At the same time, the dominant participation of Pd is highlighted. The stored value of these metals has a significant economic impact (~67 € kg-1), which is essentially created by precious metals at a rate of 98.67%, with Pd leading the way in its configuration (Au 0.90%, Ag 5.82%, Ru 4.94%, Pd 78.37%, and Ir 8.64%).

It is worth noting that the high concentration of Ag combined with its low market price of 0.84 (€ g^{-1}) and the low concentration of Ru combined with its high market price of 15.5 (€ g^{-1}) form similar participation in stored value. Correspondingly, the exceptionally high market price of Ir 149.5 (€ g^{-1}) compensates for its extremely low concentration, creating a remarkable contribution. According to [22], in their economic-technical analysis for a mass of 1 kg of multi-layer ceramic capacitors from waste-PCBs, the modulation of overall stored value from precious metals corresponded to 81.4% (Ag 3.8%, Au 20.4%, Pd 57.2%) while the remaining percentage corresponds to the following data which also contribute to the formation of the total stored value of multi-layer ceramic capacitors: Ti 7.2%, Ni 7.5%, and the rest (Cu, Zn, Mn, Al, Sn, Ba, and Pb) at 3.94%. The synergy of chromatic and magnetic separation techniques of capacitors of this type may create even more specific recycling flow processes with significant changes in their stored economic value, contributing to the sustainability of recycling plants [7].

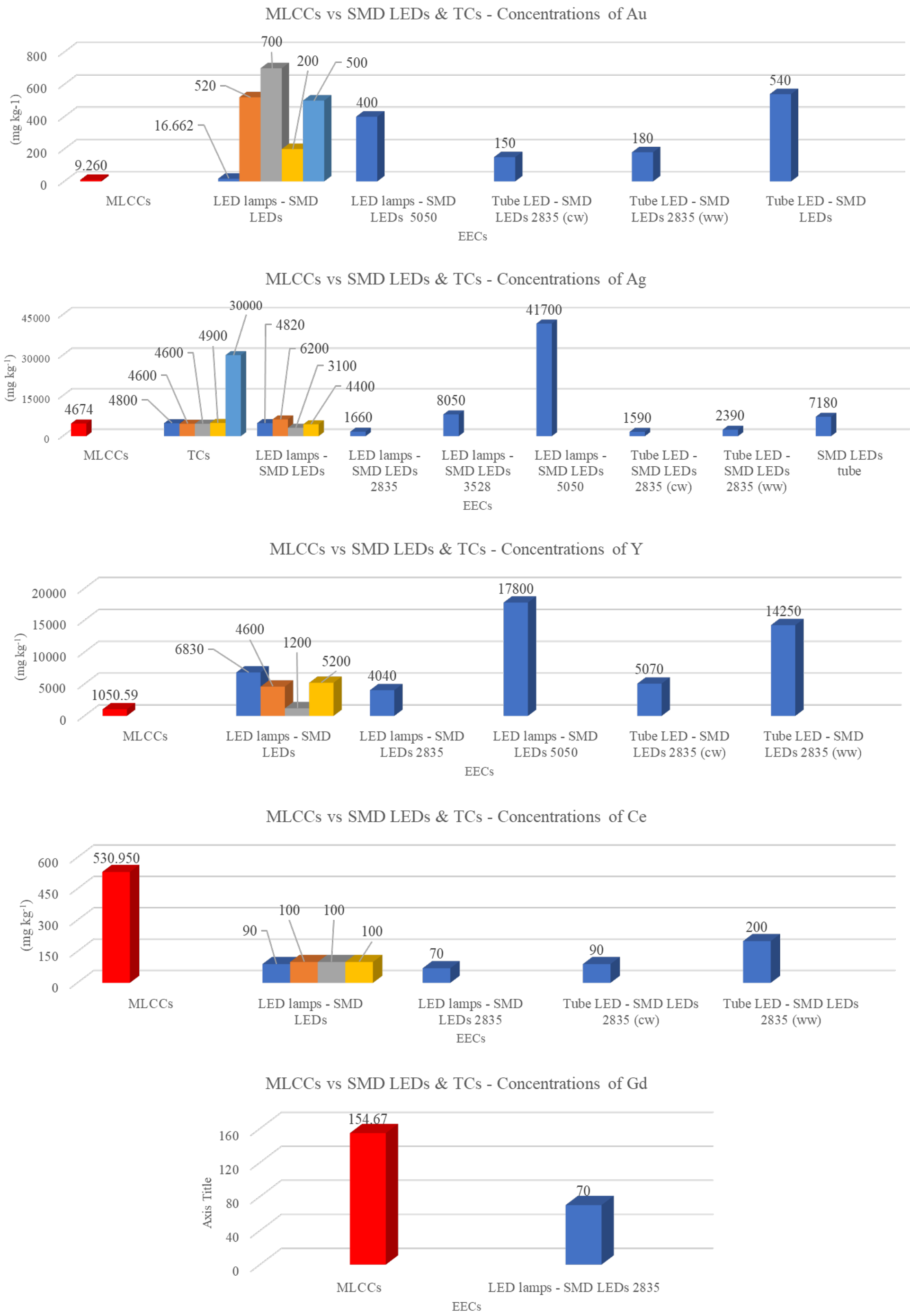

Figure 6. Comparison between concentrations of examined MLCCs (used in this study), different types of SMD-LEDs and TCs.

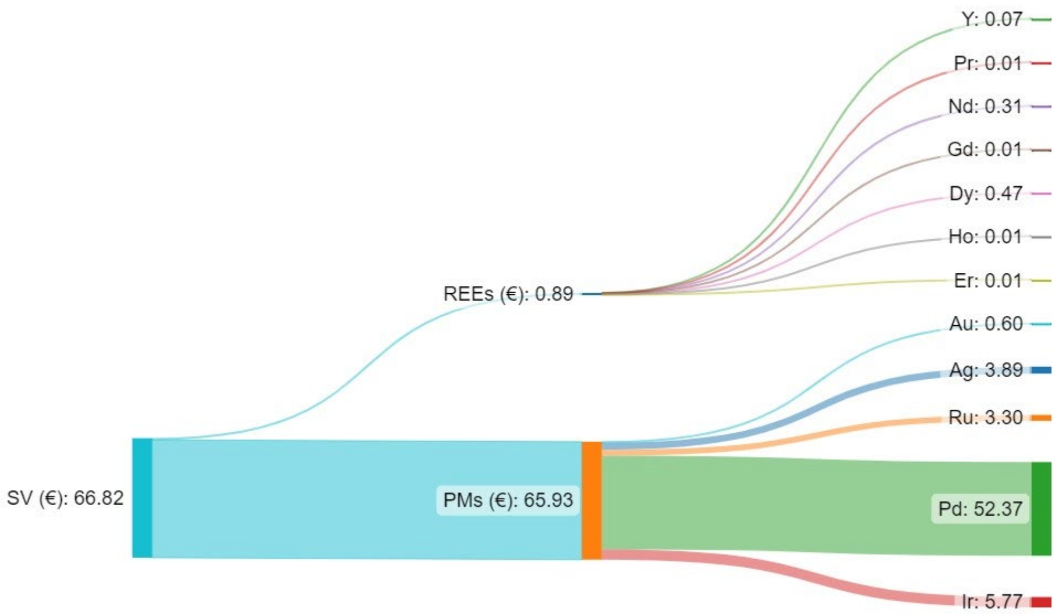

Figure 7. Sankey diagram of Stored value in MLCCs of mass 1 kg due to the presence of REEs and PMs (€).

4. Conclusions

The continuous increase of e-waste is a complex problem that requires special handling. Critical raw materials are found in their specific structures and components and usually provide the ability to separate and recycle them. Multi-layer ceramic capacitors, due to their elemental composition, are of particular interest for their separate recycling. Sorting them based on their magnetic properties and colour can create recycling flows of specific critical raw materials, which will contribute to protecting the environment and natural reserves, the partial independence from the "monopoly" of these metals and the sustainability of recycling plants.

The presence of MLCCs in the examined types of lighting equipment corresponded to 83% (15 out of 18 types), and the probability of their presence in these 15 types ranged between 12.5-100%. Their average total mass per type of examined lighting equipment ranged between 0.033-0.328 g, while their average mass per piece of examined multi-layefr ceramic capacitors as a function of lighting equipment types ranged between 0.011-0.196 g. The participation of Cat.3 of WEEE is extremely low compared to the other five categories. Still, it consists only of lamps, particularly the type G9-C, which is the most essential type for collecting MLCCs among examined lighting equipment with a performance of ~ 17 (kg t^{-1}) (per ton of each type of lighting equipment), compared to the other types whose performance ranged between 65-1640 g.

In the MLCCs used in this study, ten RREs and five PMs were detected with the respective content of 0.84 wt% and 0.60 wt%, respectively, which creates an SV equal to 66.82 (€ kg^{-1}) with Pd dominating with 78.37%. Their main composition concerns the elements Dy 0.131 wt%, Y 0.220 wt%, Nd 0.366 wt%, Pd 0.105 wt%, and Ag 0.467 wt%, while the elements La, Sm, Pr, Ho, Gd, Er, Ce, Au, Ir, and Ru are shown based on their contents as trace elements and with a content ranging between 0.001-0.053 wt%.

Expanding the use of MLCCs with flexible terminations versus MLCCs with standard terminations in driver manufacturing may reduce the growth rate of WEEE through EEE protection and contribute positively to stored value of the drivers.

The results suggest a) their selective conditional removal and b) their separate recycling.

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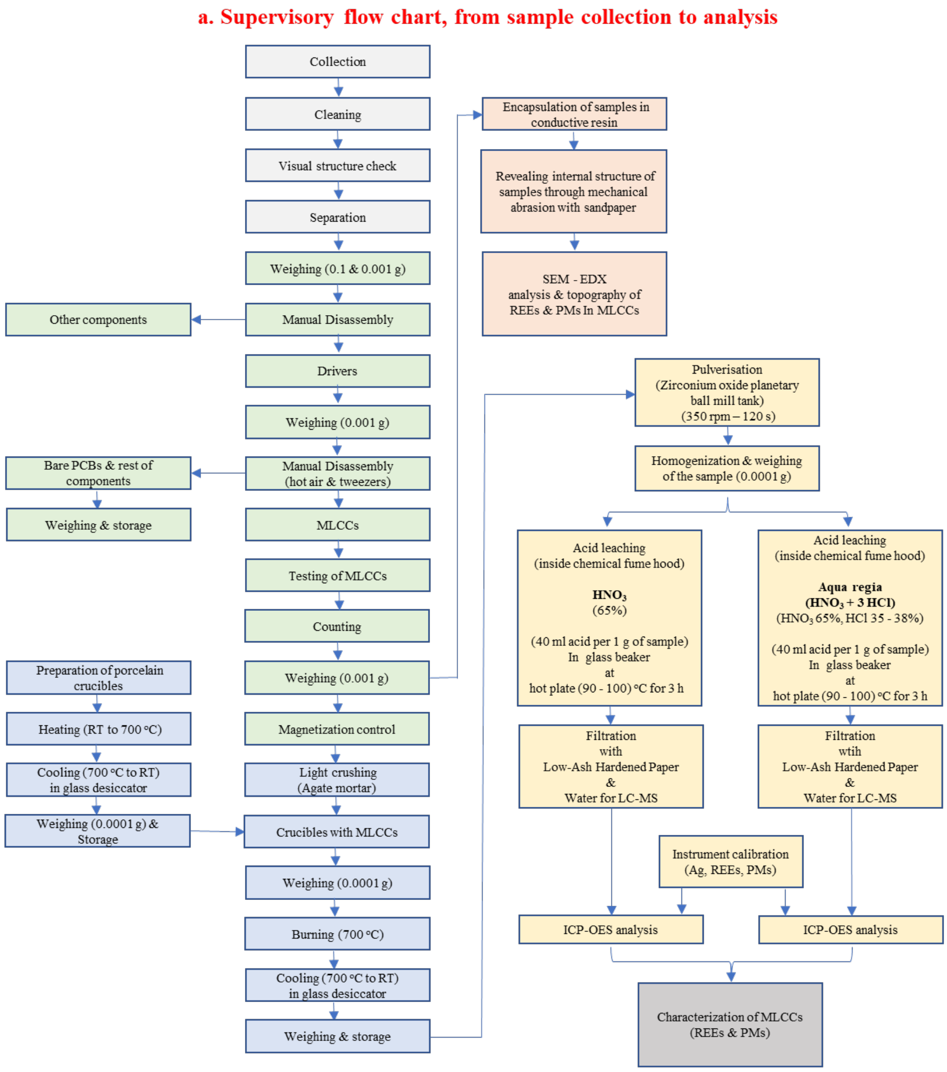
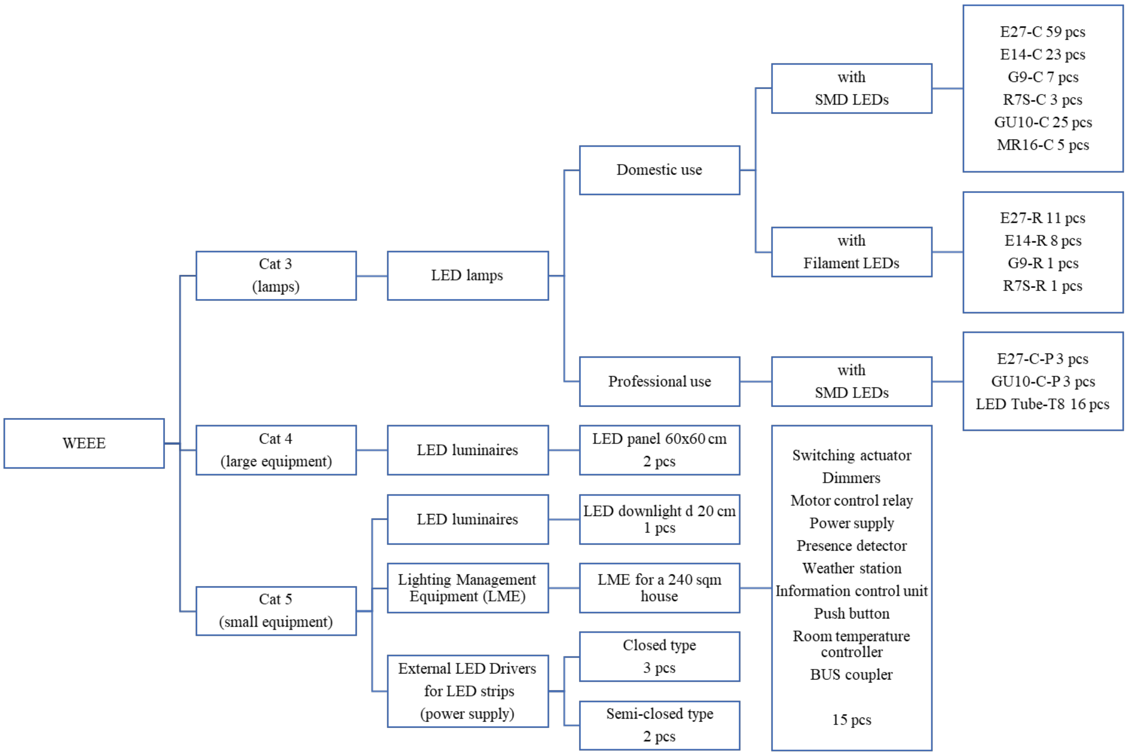
Declaration of competing interest: The authors declare that they have no known competing financial interests or personal relationships that could have influenced the work reported in this study.

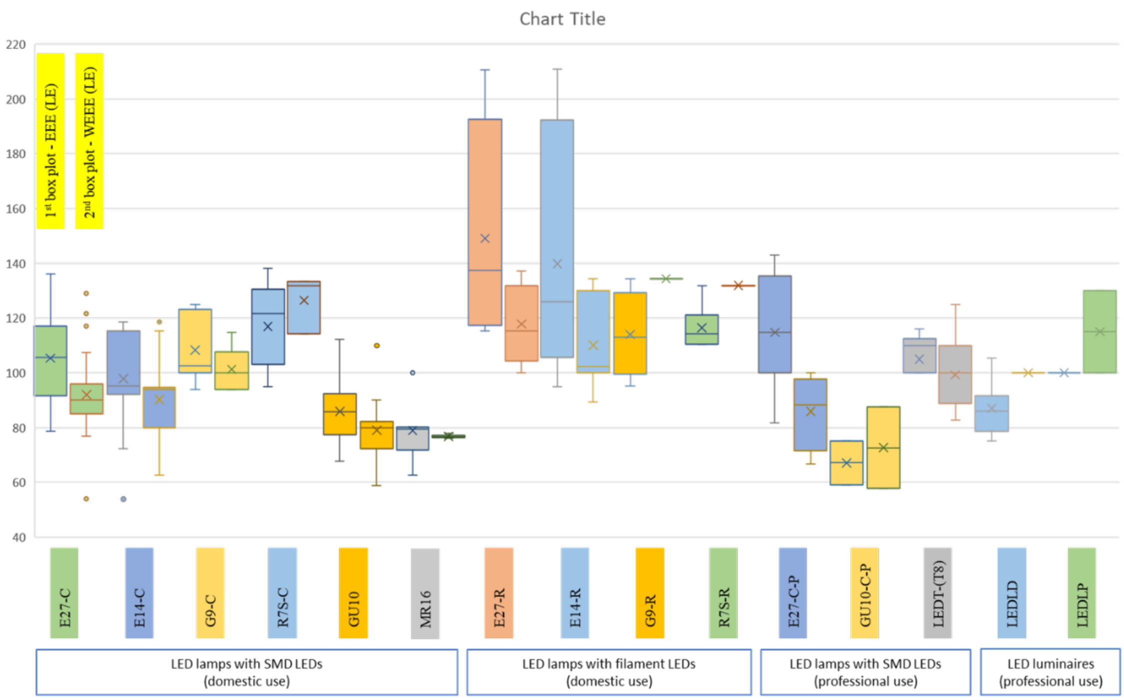
Appendixes



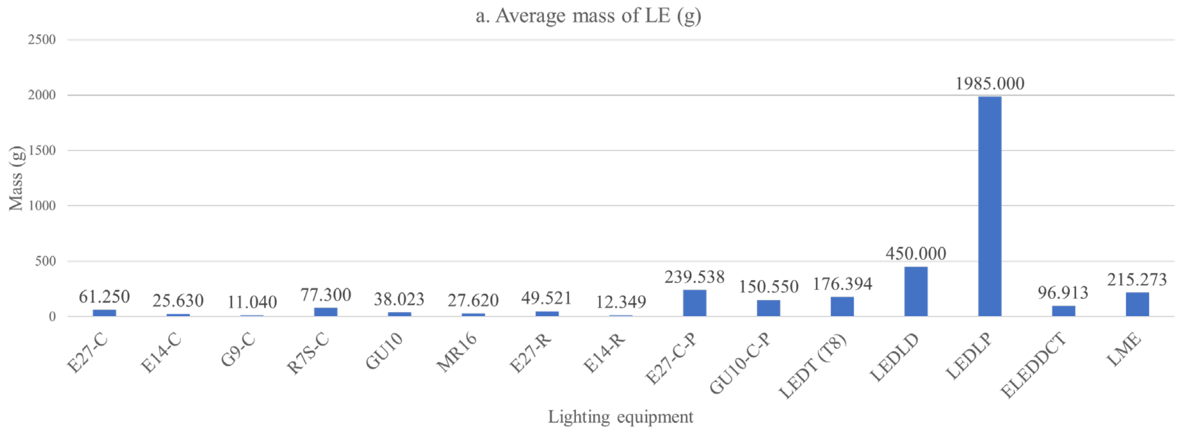
Appendix 1. Supervisory flow chart, from sample collection to analysis.

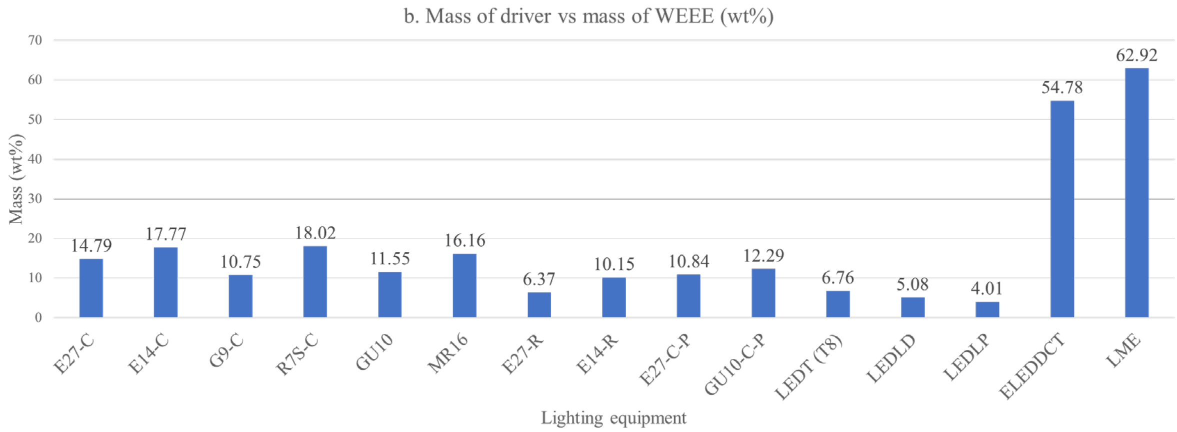


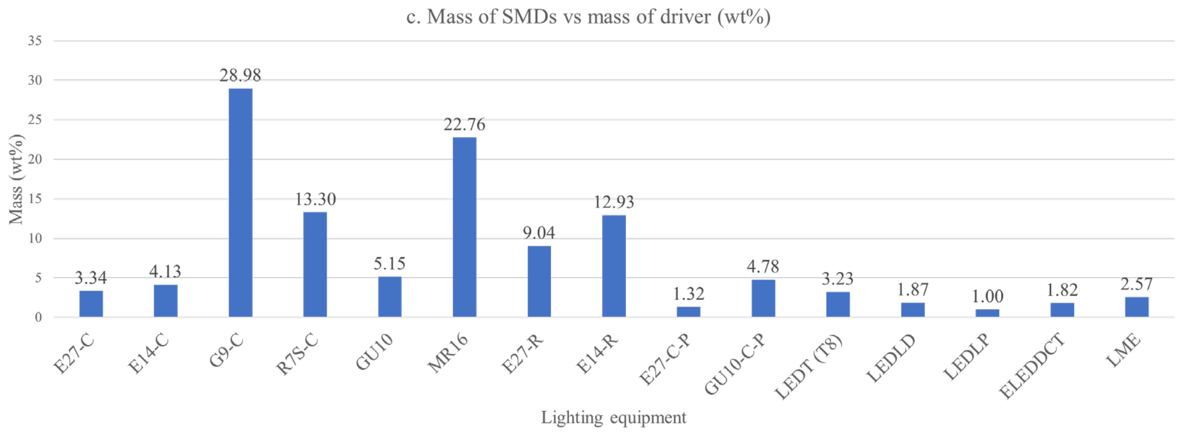
Appendix 2. Separation of LE according to WEEE categories, domestic or professional use and surface mount devices (SMD) or filament.



Appendix 1. Matching the efficiency (lm/W) of LED technology lamps and luminaires to estimate the manufacturing date of the equipment examined in this study.







Appendix 2. a. Average mass of LE (g), b. Mass of driver vs mass of WEEE (wt%), c. Mass of SMDs vs mass of driver (wt%) .

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