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Liberation and Separation of Valuable Components from LED Modules: Presentation of Two Innovative Approaches

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Abstract: The rapid development of light-emitting-diode (LED) technology is attributed to its superiority over light sources of earlier generations. Although LED lamps, compared to compact fluorescent lamps, are considered less harmful to the environment, there is still no efficient solution to deal with them at the end of their lifecycle. The first part of the study provides a detailed characterisation of LED lamps, focusing on their most interesting component: the LED module. LED packages attached to the module are highly enriched with Ga, In, Pd, Ag, Au, Sr, Y, Ce, Eu, Gd, and Lu, with the content of each element varying greatly depending on the LED technology. In the second part of this research, two new approaches for liberation and concentration of valuable components from LED modules are presented and compared: a chemical route and a thermal route. The chemical treatment leads to a highly selective separation of LED chips and encapsulation. Enrichment factors up to about 125 are achieved, and a concentrate is obtained containing approximately 14 wt% of the aforementioned valuable components. However, the process requires aromatic solvents, which are viewed as toxic. The thermal treatment results in separation of the aluminium heat sink from all other components of the LED module. Enrichment is approximately ten times lower, but the approach is technically feasible.

Keywords: LED lamps; recycling; waste electrical and electronic equipment; gallium; precious metals; rare earth elements



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1. Introduction

LED lamps belong to the newest generation of light sources. Due to their long lifetime, low energy consumption, compactness, very low IR and UV emission and mercury-free design, they are rapidly replacing conventional lamps [1]. According to Nikulski et al. (2021) [2], by 2030, LED lamps will completely replace previous-generation light sources in the European Union, with over 1000 million units expected to be sold every year from 2026 onwards solely for general household use. Due to its high flexibility and efficiency, LED technology is also widely used in other areas (e.g., in the automotive industry or street lighting), meaning that the number of LED products coming onto the market will be even higher in a rapidly growing market [3].

Once damaged, LED products represent a new material flow that contains valuable components. Additionally, they can be hazardous for people and the environment if improperly disposed of [4]. Efficient and selective recycling of modern products, such as LED lamps, is a huge challenge due to their complex design, small scale and material composition (plastics, ceramics, metals, glass and semiconductors) [5]. Companies specializing in the recycling of previous-generation light sources have to respond to this rapidly advancing technological change. Nevertheless, there is still a lack of development for specific recycling methods [6] and research on this topic [7].

The most valuable part of an LED product is the LED module, which is comprised of gallium, indium, precious metals and rare earth elements [5]. However, compared to the mass of the whole product, or even to the mass of the LED module, their concentration is very low. According to Reuter and Schaik (2015) [8], common recycling processes cannot recover valuable components from ppm levels of complex waste products such as LED lamps. Efficient liberation and concentration steps are of crucial importance. Nevertheless, there are few studies dealing with both issues, as summarized in Table 1.

Table 1. Studies on liberation and concentration of valuable components from LED bulbs/lamps.

Reference	Comminution, Liberation or Particle-Size Reduction	Sizing and Separation of Particles by Screening or Classification	Obtained Fractions
LED professional (2016) [9]	Electrohydraulic fragmentation	Manual sorting	LED packages, mixed fraction (glass, metals, plastics, ceramics, residual, electronic comp.)
Kumar et al. (2018) [10]	Electrohydraulic fragmentation	Sieving, manual sorting of fractions +6.3 mm	−0.30 mm; 0.30–1.18 mm; 1.18–6.3 mm; +6.3 mm fractions
Mizanur Rahman et al. (2019) [11]	Manual and automatic disassembly (hydro-comminution and shredding)	Manual sorting, sieving	Aluminum, plastic, LED modules, printed circuit boards
Martins et al. (2020) [12]	Comminution in a hammer mill	Sieving, electrostatic separation, magnetic separation, gravity separation (non-magnetic fraction)	Capacitors, coils, wires, polymeric housing, polymeric cover, metallic housing, Edison screw, printed circuit boards, heat sink with LEDs

Liberation and particle-size reduction have been achieved by manual disassembly [11], milling [12], electrohydraulic fragmentation (EHF) [9,10], hydro-comminution and shredding [11]. Depending on process parameters, EHF allows coarse as well as fine fragmentation. Using only a few high-voltage pulses, LED lamps are fragmented into metal pieces, ceramic parts, circuit boards, LED modules and plastic parts, which can be treated again in order to yield finer fragmentation and to liberate LED packages [9]. The study proposed how to deal with the generated material mix in order to separate single components; however, separation was done manually, so no experimental results were presented. The breakage behaviour of different types of LED lamps by EHF was also studied by Kumar et al. (2018) [10]. The authors concluded that metallic components can be liberated at a coarser fraction. However, the method is unsuitable for LED lamps with metallic casings, as no breakage was observed. Mizanur Rahman et al. (2019) [11] performed fragmentation manually and automatically (hydro-comminution and shredding). Manual disassembly and hydro-comminution generated clean fractions of aluminium, plastics, LED modules and printed circuit boards. However, hydro-comminution damages single lamp components, such as the LED modules. Shredding generates a mixed, plastic-rich fraction, hindering the proper recycling of valuable components. Recently, Martins et al. (2020) [12] considered the whole process chain. The authors showed that it is possible to liberate and separate single components of LED lamps effectively by comminution in a hammer mill, sieving and electrostatic, magnetic and gravity separation.

In summary, following fragmentation of waste LED lamps and separation of their components, either LED packages or modules are produced as a process stream. Following the physical processing stage comes the extraction and/or further separation of valuable components from obtained fractions. Review of research articles identified that a wide variety of different kinds of materials and waste from the LED industry have been treated, such as semiconductor materials [13–17], semiconductor materials and wires [18,19], wires [20] or LED chips and phosphors [21]. However, so far no solution has been provided to deal with the whole LED modules.

Our study fills this gap and shows how to liberate and concentrate valuable components contained in LED modules. This can be achieved by soaking an LED module in an

organic solvent and subsequent ultrasonication, or by thermal treatment. Both methods show advantages as well as disadvantages, which we discuss in detail. In the first part of this research, waste LED lamps are disassembled manually, and the mass balance of main components is determined. Then, a comprehensive analysis of the assembly of LED modules as well as of the content of valuable metals in the most important LED components are provided. Finally, the two approaches for liberation and separation of valuable components are presented, and benefits resulting from recycling of LED lamps are introduced.

2. Materials and Methods

2.1. Manual Disassembly and Sorting

The LED lamps used in this study were provided by LAREC GmbH, located in Saxony (Germany). One hundred LED lamps were randomly sorted from a collection container and disassembled manually, generating five different fractions: base assembly and heat sink, diffuser, driver, LED module and Edison screw.

2.2. Characterisation of LED Packages, Chips and Encapsulation

The structures of the LED packages from the three different LED modules depicted in Figure 1a were characterized using mineral liberation analysis (MLA).

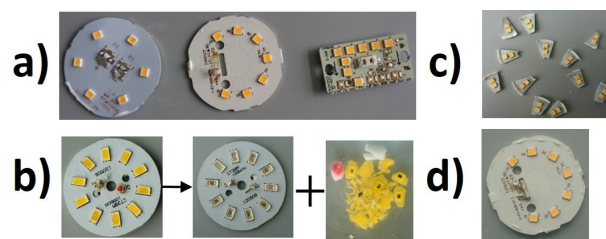


Figure 1. (a) LED modules used for the characterization of the construction of LED packages by MLA; (b) detachment of LED chips and encapsulation for chemical analysis; (c) LED module pieces used in tests; (d) LED module used in comparative experiments.

Furthermore, the chemical composition of LED chips and encapsulation was determined (Figure 1b). For this purpose, five different LED modules were soaked in acetone for 24 h, and the LED chips and encapsulation were subsequently detached mechanically. Following this, total dissolution of the substrate was carried out in three steps. Initially, aqua regia digestion was carried out. The residue was then oxidized at 1200 °C in a muffle furnace for two hours, and finally, the calcine was fused with lithium metaborate (1050 °C; 30 min). The hot crucible with melt were immediately immersed in an aqueous solution of HNO₃ (5 wt%), and finally, 5 wt% of HCl was added in order to achieve total dissolution. The chemical composition of the obtained solution was determined by ICP-MS (Thermo Scientific XSERIES 2, internal standards: 10 µg/L Rh and Re, Waltham, MA, USA).

2.3. Approaches for Liberation and Concentration of Valuable Components from LED Modules

Liberation and concentration of valuable components were investigated by two different approaches: thermal and chemical treatment. In the first part of this experimental study, the LED module pieces depicted in Figure 1c were mixed with 5 mL of the following soaking media for 24 h: an inorganic base (NaOH, 2 mol/L), an oxidizing agent (H₂O₂, 30 wt%), carboxylic acids (acetic and naphthenic acid), alcohols (isopropanol and ethylene glycol), a ketone (acetone), a glycol ether (1-methoxy-2-propanol acetate), kerosene and aromatic solvents (ethylbenzene and toluene). Samples soaked in 1-methoxy-2-propanol acetate, kerosene, ethylbenzene and toluene were subsequently ultrasonicated for eight hours (Bandelin Sonorex DT 103 H; ultrasonic frequency: 35 kHz). Additionally, three alternatives for aromatic solvents were tested: ethyl-isobutyrate, limonene and *p*-cymene. Soaking time was 24 h, and the samples were ultrasonicated for eight hours. In thermal treatment experiments, an LED module piece was heated in a muffle furnace at temperatures between 250 °C

and 600 °C for two or four hours. Finally, in comparative experiments, the efficiencies of both approaches were compared using the LED module depicted in Figure 1d (chemical treatment: soaking medium—toluene, soaking time—24 h, ultrasonication time—eight hours; thermal treatment: heating temperature—600 °C, heating time—four hours).

3. Results and Discussion

3.1. Disassembly and Sorting Process

The five main fractions obtained by manual disassembly and sorting are depicted in Figure 2a.

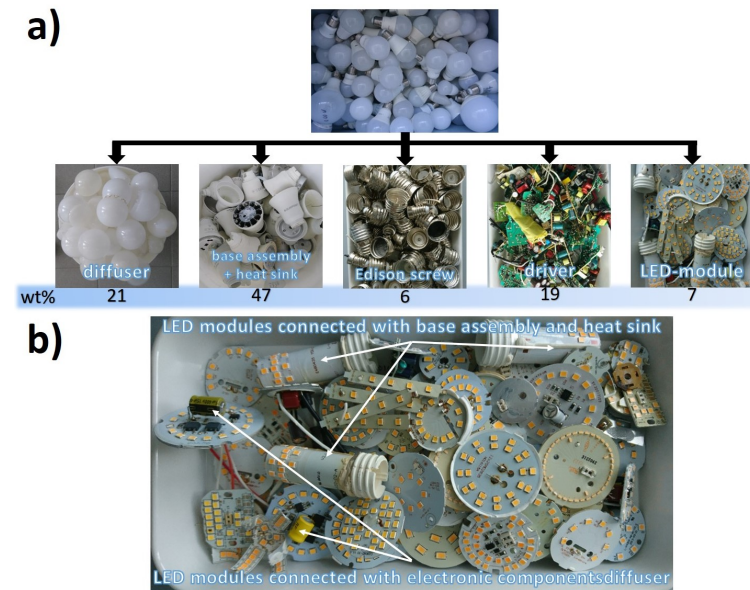


Figure 2. (a) Fractions and their mass percentage generated by manual disassembly of LED lamps; (b) all separated LED modules.

The average mass of a single LED lamp amounts to approximately 53 g and consists of base assembly and heat sink (47 wt%), diffuser (21 wt%), driver (19 wt%), LED module (7 wt%) and Edison screw (6 wt%). The only component that is constructed solely from plastic is the diffuser. The simple composition of the metallic part of the base assembly and heat sink (over 95 wt% of aluminium [12]) significantly facilitates recycling. This is also the case for the Edison screw, which consists mainly of iron (approximately 82%) and nickel (approximately 17%) [12]. The driver and the LED module are the most complex and precious components. This study focuses on the LED modules depicted in Figure 2b, which have an average mass of 4 g and an average number of LED packages of 13.3. A total of 92% of LED modules could be easily detached from all other parts, 3% of LED modules remained connected to base assembly and heat sink and the last 5% to electronic components.

3.2. Characterisation of LED Packages, Chips and Encapsulation

Two of three of the LED packages analysed by mineral liberation have the same arrangement, which can be seen in Figure 3.

The LED package is comprised of a reflector cup, encapsulated phosphor particles and wires (Ga, Sr, Y, Al, Ag), electrode (Cu), solder (Sn), LED chips (Ga) and die-attachment material (Ag). The third LED package shows the following differences: wires consisting of gold and thick silver layer on Cu electrode. The most-valuable components of an LED module (Ga, Ag, Au, Sr, Y) are included primarily in the LED chips and encapsulation. To gain more specific information, the LED chips and encapsulations detached from five different LED module types were dissolved, as described in Section 2.2, and their chemical composition is provided in Table 2.

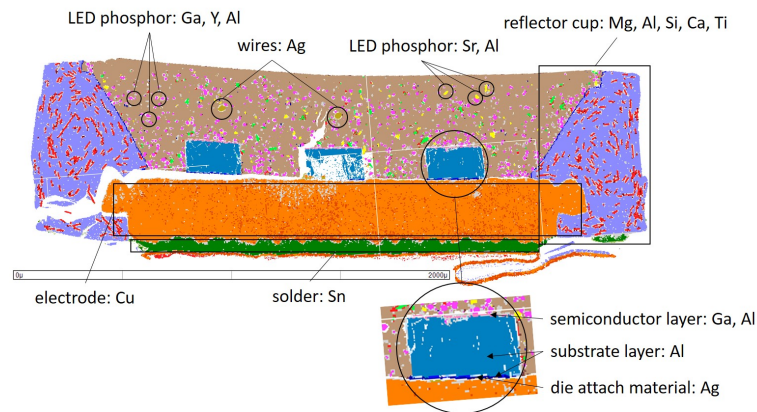


Figure 3. The profile of the LED package as determined by mineral liberation analysis (MLA).

Table 2. Characterized LED modules and elemental composition of LED chips and encapsulation.

LED Module	S1	S2	S3	S4	S5	Average Value
LED module before treatment						
LED module after treatment						
Detached LED chips and encapsulation						
Mass of LED module in g	5.44	4.95	5.49	1.95	6.02	4.77
Mass of LED chips and encapsulation in mg	61	55	63	66	63	62
Number of LED packages	20	40	10	25	12	21.4
Mass of LED chips and encapsulation per LED package in mg	3.1	1.4	6.3	2.6	5.3	3.7
Main components [wt%]						
Ga	4.03	0.61	0.25	2.95	0.62	1.69
Sr	1.02	0.35	1.09	0.65	0.25	0.67
Y	7.96	0.01	4.20	4.81	0.06	3.41
Lu	<0.01	15.47	0.01	<0.01	17.19	6.54
Other relevant components [ppm]						
In	66	60	47	39	38	50
Pd	1623	4	842	942	13	685
Ag	1589	18	977	621	1377	916
Au	12	6	2	4	2	5
Eu	320	251	197	233	308	262
Gd	13	6	15	9	10	10

In addition to the elements detected by mineral liberation analysis, significant amounts of In, Pd, Ce, Eu, Gd and Lu were found. The average content of the following components does not exceed a value of 5 ppm: Sc, Co, Ni, La, Pr, Nd and Tb. The content of main components in the substrate varies strongly, indicating differences in the LED technology. The Ga content ranges from 0.25 wt% to 4 wt% and has two different applications: as a compound of semiconductor materials [22] as well as of garnet phosphors [23]. The high Ga content in samples S1 and S4 of about 4 wt% and 3 wt%, respectively, indicates the presence of garnet phosphors, for instance $\text{Y}_3(\text{Al,Ga})_5\text{O}_{12}:\text{Ce}^{3+}$, which belongs to the phosphors emitting yellow-green light [23]. This suggestion confirms the detected high Y content of 8.0 wt% and 4.8 wt%, respectively. By contrast, the high concentration of Lu in samples S2 and S5 suggests the presence of a Lu-rich phosphor garnet such as $\text{Lu}_3\text{Al}_5\text{O}_{15}:\text{Ce}^{3+}$, which

also emits yellow-green light [23]. In sample S3, neither significant amounts of Ga nor Lu were found, it did have over 1 wt% Sr. These LED packages most likely include nitride red-emitting, nitrogen oxide cyan-emitting or blue-cyan-emitting phosphors. Besides Lu and Y, other rare earth elements (Ce, Eu, Gd) were detected in lower concentrations, acting as a component of different kinds of LED phosphors and serving as doping agents [24]. The presence of Ag and Au can be explained by the application of both elements as die attachment material (e.g., Ag as silver paste) and as wire bonding [22].

3.3. Approaches for Liberation and Concentration of Valuable Components from LED Modules

3.3.1. Chemical Treatment

First, chemical treatment was investigated. Figure 4a provides the results of soaking LED module pieces in different media for 24 h. Sodium hydroxide solution (1), hydrogen peroxide solution (2) and acetic acid (3) are not suitable for the liberation of LED chips and encapsulation. In all three cases, the decomposition of the heat sink/upper layer occurs instead that of the LED package. After soaking in naphthenic acid (4), isopropanol (5), ethylene glycol (6) and kerosene (7), no visible changes to the LED module piece were observed. In contrast, soaking in acetone (8), 1-methoxy-2-propanol acetate (9), ethylbenzene (10) and toluene (11) resulted in partial detachment of the LED encapsulation. However, even after a week of soaking, there was not complete detachment. The best results were achieved using toluene.

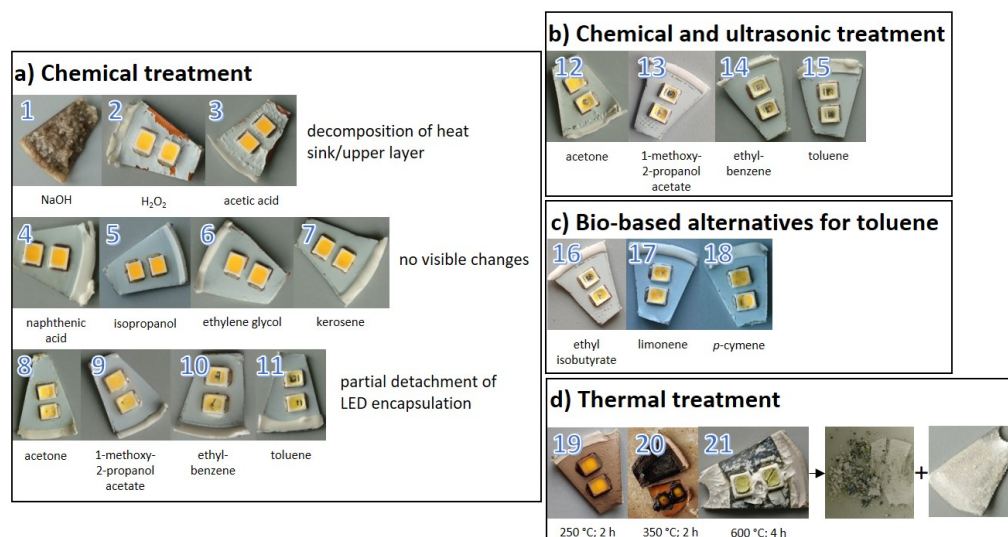


Figure 4. Liberation experiments.

In further experiments, LED module pieces treated in acetone, 1-methoxy-2-propanol acetate, ethylbenzene and toluene were ultrasonified for eight hours, leading to enhanced detachment (Figure 4b). In the case of ethylbenzene (14) and toluene (15), the LED substrate separated almost completely.

The use of aromatic solvents is viewed critically due to their toxicity. In recent years, attempts have been made to develop green alternatives. According to the literature, terpenes, their derivatives [25] and bio-based esters [26] have properties similar to petroleum-based chemicals and can be substituted for them in certain applications. However, as shown in Figure 4c, the tested bio-based compounds were less efficient than toluene or ethylbenzene even after eight hours of ultrasonication (16: ethyl isobutyrate, 17: limonene, 18: *p*-cymene).

3.3.2. Thermal Treatment

In other experiments, the LED module pieces were treated thermally. Depending on temperature and heating time, they decompose to different extents (Figure 4d). At 250 °C, only a slight decomposition of the upper layer occurs (19). At 350 °C, the reflector cup

degrades and the LED piece splits into two parts: aluminium heat sink with insulating layer and other upper components (20). At 600 °C, conductive and insulating layers as well as LED packages split from the heat sink, forming a fragile and easy to pulverise substrate (21). Thus, for thermal treatment, only the separation of the metal base plate from all other LED components is possible.

3.3.3. Comparative Experiments

For the quantitative comparison of both approaches, two identical LED modules (depicted in Figure 1d) were treated chemically (soaking medium: toluene, ultrasonication time: 8 h) and thermally (heating temperature and time: 600 °C and 4 h). The elemental composition of both concentrates and the enrichment factor EF are provided in Table 3.

Table 3. Elemental composition of concentrates obtained after chemical and thermal treatment, and enrichment factor (EF) as percentage of the mass of the LED module.

	Chemical Treatment		Thermal Treatment	
	[wt%]	EF	[wt%]	EF
Main high value components				
Ga	2.67	78.0	0.13	3.60
Y	4.56	119	0.34	6.20
Lu	5.67	128	0.36	6.25
Other valuable components				
	[ppm]	EF	[ppm]	EF
Sr	6719	102	26	6.26
In	67	0.38	872	6.24
Pd	889	117	67	6.18
Ag	3238	1.30	4216	5.96
Au	4	5.10	3	3.34
Eu	294	69.9	114	6.14
Gd	15	34.9	3	5.68
Base metals				
	[wt%]	EF	[wt%]	EF
Al	10.8	0.14	7.4	0.10
Fe	0.08	0.28	0.57	1.48
Cu	0.19	0.03	31.5	6.20
Zn	0.38	14.2	0.12	3.20
Sn	0.54	0.10	24.6	6.25

The concentrate resulting from chemical treatment consists of significant amounts of Ga, Y and Lu. It bears resemblance to substrate S4 characterized in Table 2, and the content of all valuable components amounts to approx. 14 wt%. The enrichment factor varies from 1.30 for Ag to 128 for Lu, respectively.

In comparison, the amount of valuable components in the concentrate obtained in the thermal treatment, at 1.37 wt%, is nearly ten times lower. The enrichment factor ranges from 3.34 for Au to 6.26 for Sr. Main components of the concentrate are Cu and Sn, with total mass fractions of over 55 wt%. In contrast, the main component of the concentrate obtained after chemical treatment is Al, with a content of approximately 11 wt%.

3.4. Estimates for Resource Recovery from LED Lamps in the European Union from 2022 and 2030

The described approaches are neither technically nor economically fully developed. However, there is no question that more attention should be paid to the recycling of end-of-life products. This section covers the benefit and importance of the recycling of LED lamps, estimating how much Ga, In, Pd, Ag, Au, Sr, Y, Ce, Eu, Gd and Lu can be recovered from LED lamps within the EU between 2022 and 2030, and how many primary resources can be saved by doing so. For the calculation, the following assumptions were considered:

- The annual number of LED lamps sold in the EU is 968 million [2];

- The average number of LED packages per LED lamp is 21.4, and the average mass of LED chips and encapsulation per LED package is 3.7 mg (see Table 2);
- The average mass percent of a component X (X = Ga, In, Pd, Ag, Au, Sr, Y, Ce, Eu, Gd and Lu) in the concentrate is given in Table 2;
- The construction and the chemical composition of LED lamps will not change during this time period;
- The content of Lu in monazite is 30 ppm [27], and the content of monazite in the Bayan Obo deposit is 3.36 wt% [28];
- The content of Y in tailings from alluvial tin mines in Malaysia is about 0.56 wt%, and the content of Y₂O₃ in xenotime is 60 wt% [29];
- The content of Ga in bauxite and zinc ores is about 100 ppm [30].

The annual average amount $Prod_X$ of each component X calculated by Equation (1) is given in Table 4.

$$Prod_X = 968 \text{ million} \times 21.4 \times 3.7 \text{ mg} \times \frac{\text{wt\%}_X}{100} \quad (1)$$

Table 4. Annual amounts of Ga, In, Pd, Ag, Au, Sr, Y, Ce, Eu, Gd and Lu required for the production of LED packages sold in the European Union between 2022 and 2030 in kg ($Prod_X$) and the corresponding amount of primary resources needed for the production of Ga, Y and Lu in t (Res_X).

	Ga	In	Pd	Ag	Au	Sr	Y	Ce	Eu	Gd	Lu
$Prod_X$ in kg	1307	3.84	53	71	0.40	519	2634	113	20.21	13.69	5049
Res_X in t	13,074	-	-	-	-	-	995	-	-	-	5×10^6

With over 5000 kg/y of Lu and 5×10^6 tonnes of Bayan Obo ore required each year, these are the highest estimated values. According to Emsely (2011) [27], about 10 tonnes of Lu are produced from primary resources each year (data from 2011). This would imply that 50% is consumed in manufacturing processes, but we believe that, at present, the production capacity is higher. The second-most abundant element in the obtained LED concentrate is Y, with the estimated amount of approximately 2600 kg/year. For this quantity, nearly 1000 tonnes of tailings from alluvial tin mines in Malaysia are needed each year. Compared to the world production of Y as Y₂O₃, which in 2013 was between 8000 t and 12,000 t [31], the estimated value represents only a small percentage of the total. The calculated values for Ga are approximately 1300 kg and 13,100 t per year, respectively. When compared to the world production of Ga from primary resources (in 2020 about 300,000 kg), the estimated number is of little significance.

4. Conclusions

This study deals with the liberation and concentration of valuable components from LED modules by chemical and thermal treatment, and with characterization of the assembly and the chemical composition of LED packages. Moreover, the benefits and importance of recycling LED lamps are discussed. It has been found that the content of valuable components in LED chips and encapsulation is remarkably high and varies significantly depending on the LED technology. The most-abundant elements are Lu, Y, Ga and Sc, with average contents of 6.54, 5.41, 1.69 and 0.67 wt%, respectively. Additionally, significant amounts of In, Pd, Ag, Au, Ce, Eu and Gd were detected. The introduced approaches do not show industrial maturity, and further improvements are required. The main drawback of chemical detachment is the application of toxic aromatic solvents. Green alternatives considered in this study did not provide satisfactory results. Another conceivable option is the optimization of the frequency and power of the ultrasonic waves to improve detachment in non-aromatic solvents, e.g., acetone. By contrast, the content of valuable components in the concentrate produced by thermal treatment is ten times lower. The main elements are Cu and Sn, with mass fractions of over 55 wt%. Although thermal treatment is technically feasible, separation of valuable elements from the low-grade concentrate entails much

higher efforts. Due to the high importance of the covered topic (recycling of all LED lamps in the European Union could save over 5×10^6 t of primary resources each year) more research should be carried out in this field.

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Abbreviations

The following abbreviations are used in this manuscript:

EF	enrichment factor
EHF	electrohydraulic fragmentation
EoL	end-of-life
EU	European Union
ICP-MS	inductively coupled plasma mass spectrometry
LED	light emitting diode
MLA	mineral liberation analysis

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