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1	Selective separation of plastic LED lamp components using electrodynamic
2	fragmentation for material recovery
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24 Abstract

The recycling of light-emitting diode (LED) lamps and tubes is becoming increasingly 25 important due to their growing market share as energy-efficient lighting technology. Here 26 we report on the use of high voltage electric-pulse fragmentation to recover elementary 27 components such as LED chips and printed circuit boards (drivers). E27 LED lamps with 28 29 plastic bulbs, which represent 48% of deposits collected by a French company, are used as a 30 case study. More than 150 lamps were tested on a laboratory reactor for electrodynamic fragmentation. The technological process in which highly energetic electrical pulses were 31 applied to materials immersed in water was studied in order to separate the components of 32 the LED lamps using a minimal specific energy. The estimated energy necessary to achieve 33 total separation assessed at 64%, without grinding pretreatment, was 5.2 ± 0.6 kWh per ton, 34 35 representing a mass recycling rate of 74%. Based on the disassembled material, the commercial value of the recovered materials was thus estimated. Gold, as the most 36 representative material, was found to represent 0.03% of the mass fraction for 83.6% of the 37 total commercial value. The process disassembling capacity is a key issue to increase the 38 39 recycling rate of current LED lamps and tubes.

40

41 Keywords: Waste LED lighting devices; Material recovery; Selective comminution;
42 Electrodynamic fragmentation;

43

44 **1. Introduction**

To reduce energy consumption and increase energy independence (European Commission, 2009), the technology of light-emitting diodes (LED) has been developed since 2000. By the end of this decade, it is expected that all incandescent and compact fluorescent lamps (CFL)

48 will be replaced by LED lamps, tubes, and panels (ECOSYSTEM, 2019). LED lamps will effectively capture 74% of the market share by 2030, leading to a 46% reduction in energy 49 lighting consumption around the world (Aman et al., 2013; Bruno Gayral, 2017; Natthanon et 50 al., 2018; Doulos et al., 2019). The sharp increase in luminaire efficiency and the gradual 51 reduction in the cost of purchasing LEDs have led to an augmentation in the market share of 52 53 LED lamps and tubes. Rather by the capacitors and electronic components of the printed 54 circuit boards (PCBs or drivers), the lifespan of LED lamps is around 5-7 years, with an 55 average around 25,000 hours - and up to 50,000 hours - depending on the manufacturers (Narendran et al., 2001; Richter et al., 2019). Nevertheless, LED products are expected to 56 end up in the e-waste stream in massive quantities from 2022 to 2030, which represents a 57 challenge for legislators and recycling companies. There is currently no dedicated industrial-58 59 scale technology to treat this e-waste (Mizanur Rahman et al., 2017), and in France, all LEDs are simply stored in the factories such as the ARTEMISE company. ECOSYSTEM, a non-profit 60 ecological organization in France, is in charge of collecting and finding solutions for recycling 61 used LED lamps and tubes. The valuable materials contained in LEDs and their design variety 62 are the main recycling issues (Mizanur Rahman et al., 2019). 63

64 White LED chips are based on a blue multiple quantum well (MQW) and rare earth yellow phosphors materials (Schubert and Kim, 2005; Dussaigne et al., 2020; Shameem et al., 2020; 65 66 Isfahani and Faez, 2021). MQW are grown on III-V semiconductor, most commonly with indium, gallium (Pourhossein and Mousavi, 2018), and nitrogen as ternary compounds, 67 leading to the use of InGaN / GaN MQW. It is worth noting that more abundant materials 68 69 such as zinc oxide-based LED products could be good candidates not only to replace InGaN 70 but also to develop new technologies such as lasing-based white LEDs (Zhang et al., 2013; 71 Nomenyo et al., 2014). However, they are still dealing with stability issues especially in 72 doping that currently prohibit the use of such materials. With the current technology, drivers contain gold, copper, tin, nickel and other precious metals (Lim et al., 2013; Reuter and Van 73 Schaik, 2015; Franz and Wenzl, 2017; Pourhossein and Mousavi, 2018), which constitute the 74 highest valuable fraction. Additionally, to reduce fire hazard, brominated flame retardants 75 76 are used in the plastic matrices (Lim et al., 2011). The European cycLED project (Marwede et 77 al., 2012) explored the mass flows of selected target materials in LED products. Some 78 elements like gold, silver, palladium, aluminium, copper, tin, and antimony were found in 79 the electronic components. Moreover, other resources are used in a limited number of specific components in the LED chips, such as semiconductors (gallium and indium) or rare 80 earth metals (phosphor) (Pilotto Cenci et al., 2020; Martins et al., 2020). This study focuses 81 on the recycling of E27 LED light devices to facilitate the recovery of parts or the extracted 82 83 materials, including precious and strategic metals (Fouquet and Martel-Jantin, 2014). One of the key issues is determining how to separate the optical systems (bulbs) and bodies without 84 mechanical crushing of components containing metals parts. In this respect, only a recycling 85 process adapted to the specificity of the LED lamps could undertake this separation to 86 87 recover all the elementary components individually such as LED chips, LED modules and 88 drivers.

In the context of the development of new recycling routes for some categories of WEEE, such as solar panels, printed circuit boards or lithium-ion batteries, some researchers have tested the high voltage electric-pulse fragmentation process to increase the recovery rate while reducing production costs compared to mechanical crushing processes.

Indeed, it was noted that using high voltage technology allowed for the selective
fragmentation of concrete and composite materials (Van der Wielen et al. 2013; Shi et al.
2014; Touzé et al. 2017; Bru et al. 2018). This technology is based on the rapid return of

96 highly concentrated electrical energy stored in capacitors by creating a fracturing shock wave allowing selective separation (Zhao et al. 2015; Nevala et al. 2019). How about, EDF 97 was tested for the recycling of waste solar photovoltaic panels by considering the energy 98 consumption (Song et al., 2020). It was found that the optimal condition of high voltage 99 100 fragmentation is 160 kV for 300 pulses with an energy consumption of 53.6 kWh/t, crushing 101 the PV panels into particles of 4.1 mm on average. Printed Circuit Boards waste have also 102 been studied using this technique. It was observed that an increase in the voltage and the 103 pulse number improved the metal recovery rate with a reduction in the size of the crushed products (Zhao et al., 2015). Another study suggests that the metal and nonmetal materials 104 could be dissociated with minimum tensile strength, which makes the circuit boards 105 106 gradually break into block pieces (Duan et al., 2018). Finally, we can report on the study of 107 lithium-ion batteries (Tokoro et al., 2021) which employed EHF for separating cathode particles and aluminum (AI) foil. One of the main results is that using a voltage of 25 kV, 108 93.9% of the cathode particles are separated from the Al foil, and these particles were easily 109 recovered by appropriate sieving. The parameters studied in these studies were the voltage 110 111 (20-200 kV), electrode gap (10-40 mm), pulse repetition frequency and number of electrical 112 pulse (1-1000) (Bru et al., 2018).

These results suggest that high voltage electric-pulse could offer a new and promising approach to the recycling of specific WEEE waste such LED lamps. So, this study deals with the potential of EDF technology to selectively separate the electronic components of plastic E27 LED lamps without damaging them, which would allow their reuse or further recycling to recover the constituent materials and more specifically precious metals. After reviewing the type of reactor used in this study, we will show how the technique allows for the

119 comprehensive disassembling of the most frequently collected products: i.e. plastic E27 LED120 lamps.

121

122 **2. Materials and methods**

123 2.1. Study materials: Plastic E27 LED lamps

124 To determine the trends and repeatability of the used LED light products collected by the 125 ARTEMISE company, we carried out a monthly sampling campaign starting in September 126 2019 and then continuing on a quarterly basis until June 2020. The results are shown in Fig. 1. We characterized six big bags of LED lamps and spotlights with an average weight of 336 127 kg and a total number of 29,600 LED units. As a result of this sampling, E27 LED lamps were 128 129 found to be the most common type of lamp, accounting for 58-73% of the deposit 130 depending on the sampling period. GU10 LED spotlights were the second most represented type with 16-29% followed by E14 LED lamps accounting only for 3-7%. These three models 131 constitute 88-92% of the total deposit. A similar trend was observed by Mizanur Rahman et 132 al. (2019) who concluded that E27, E14, and GU10 LED lamps represented 90% of LED 133 134 products. The mapping of the deposits (Fig. 1-a) reveals that E27 LED lamps with small 135 plastic parts are the predominant model (between 25% and 52% and 38% on average). E27 LED with large plastic parts are also frequently represented in the deposit (between 6% and 136 137 14% and 10% on average). These differ from the small plastic E27 LED lamps by the size of the bulb, which is composed of polycarbonate (PC) (Cheng. 2002; Martins et al., 2020). This 138 model could not be explored further in this study because of these dimensions. Another 139 140 model is the GU10 LED spot, which is also common on average it accounts for 22%. Finally, 141 we chose to examine E27 LED lamps with small plastic optical system. The body of these

lamps is made of aluminium with a typical thickness of 0.8 mm and covered by a polymericmaterial with a typical thickness of 0.9 mm (Fig. 1-b).

This type of LED lamp represents around 38% of total deposits (Fig. 1-c) and thus constitutes
a very representative sampling.

146 **2.2. Laboratory tests**

147 There are two types of high-voltage fragmentation. In the first, known as localised fragmentation, the discharge occurs within the material(s): this is electrodynamic 148 fragmentation (EDF). The second, known as delocalized fragmentation, consists of 149 transmitting a shockwave to the material(s), through a dielectric liquid: this is 150 electrohydraulic fragmentation (EHF) (Justin Martin, 2013). The voltage rise in the case of 151 152 EDF is very fast (< 500 ns) with applied values in the order of 90 to 200 kV, whereas the 153 voltage rise in the case of EHF is slower (> 500 ns) with values in the order of a few tens of kilovolts (30-50 kV) (Pestalozzi et al., 2018). EDF technology was chosen in our study 154 because, as shown above, it has been the most widely used technology in the fragmentation 155 of some WEEE. 156

As shown in the Fig. 2 (a, b), the EDF laboratory-scale tests were carried out using the SELFRAG laboratory-scale device for the batch processing of materials. The system was manufactured by the Swiss company SELFRAG. It is based on a 5.3 L capacity vessel (SELFRAG, 2020) containing 2.78 L of deionized water. The equipment is designed to process products weighing up to approximately 1 kg and measuring between 80 mm (diameter) and 40 mm (height).

163 The discharge electrode is immersed in the upper part of the container, while the bottom 164 constitutes the ground electrode (Fig. 2-c). The device produces high voltage electrical 165 discharges between the two electrodes. The operating parameters are the voltage (90-200

166 kV), electrode gap (10-40 mm), pulse repetition frequency (1-5 Hz), and number of electrical
pulses (1-1000) (Bru et al., 2018).

The unit was used to fragment 164 plastic E27 LED lamps. These lamps represent a total 168 mass of 7.7 kg with an average mass per sample of 47.0 ± 11.6 g. The studied parameters 169 were the voltage and the number of discharges, while the interelectrode distance was kept 170 171 constant at 40 mm throughout the study. In fact, the dimensions and shape of these E27 LED lamps did not allow us to test a distance of less than 40 mm. The objective of the laboratory 172 scale tests was to study the influence of the generator energy (specific energy in kWh/t for 173 plastic E27 LED lamps) on the fragmentation of samples. As shown in Equation (1), the (1)174 energy of the generator only depends on the variation of the voltage and the number of 175 pulses applied, and not to the capacitance, which is fixed at 37.5 nF for this laboratory 176 machine. The applied voltage for this study was set between 90 kV and 180 kV. 177

$$E_{gen} = n x \frac{1}{2} x C x U^2$$

E_{gen} represents here the generator energy (J), n the number of pulses, C the capacitance (F),
and U the pulse voltage (V) (Bru et al., 2018).

181

182 2.3. Methodology

As will be shown below, it was possible to disassemble the plastic E27 LED lamps into four components: bulb, LED module, driver and body with the Edison screw. Based on this observation, we defined criteria to assess the quality of the disassembled sample. The fragmentation can be considered validated by a recycling company if:

187 The bulb and body are completely separated;

188 The LED module is separated with at least 50% of the LED chips still attached to the 189 aluminium support;

190 The driver is completely separated from the LED module and base;

191 The Edison screw is still attached to the lamp body.

For all series, we counted the number of detached bulbs, LED modules, and drivers in order to determine the percentages of selective fragmentation. A total of 164 identical plastic E27 LED lamps were individually fragmented. For the most favorable voltage-pulse pairs, eight experiments were carried out to ensure the reproducibility of the validated fragmentations. Reproducibility was assessed by observing and counting the total liberation of the drivers and the LED modules with at least 50% of LED chips still placed on the aluminium modules.

Let us now define the condition criteria for the electrical components, namely the LED modules and drivers. Four different conditions were defined for both the module and the driver. For the LED module, the condition: excellent, good, bad, and very bad indicates that 75-100%, 50-75%, 25-50%, and 0-25% of the chips are still attached to the aluminium plate respectively. For the drivers, their condition is simply based on a visual analysis with excellent, good, bad, and very bad respectively referring to their unfractured, weakly, significantly, and totally fractured state.

205

3. Characterization of selective fragmentation of plastic E27 LED lamps elements

207 **3.1. Selective fragmentation**

208 3.1.1. Results

We tested plastic E27 LED lamps (110 mm height) with a standard diameter optical system (40 mm). They were diagonally positioned in the vessel. Using a logic of experimentation and experimental observations, the voltage range was between 90 kV and 180 kV, with electric discharges ranging from 1 to 30 pulses at a frequency of 5 Hz. The average specific energy
was 5.2 ± 0.6 kWh/t. The mapping of the tests shown in Fig. 3-a represents the areas that
were tested to determine the area for the dismantling and deconstruction of the plastic E27
LED lamps. The aim here was to validate the most suitable parameters for the voltage and
number of pulses. Considering the criteria established in the Methodology section above,
Fig. 3-b (150 kV; 15 pulses) and 3-c (150 kV; 5 pulses) do not validate the criteria, whereas
Fig. 3-d (150 kV; 4 pulses) and 3-e (140 kV; 5 pulses) do.

After selecting the most favorable dismantling conditions, we counted seven voltage-pulse pairs for which eight tests were carried out. In this case, 56 plastic E27 LED lamps were investigated, corresponding to a total sample mass of 2.6 kg. Table 1 and Fig. 4 show the selective fragmentation of the plastic E27 LED lamp elements based on visual inspections. Overall, the results of the seven experimental series A to G are very encouraging, resulting on an average overall separation of 64%.

225

226 **3.1.2. Discussion**

227 The B series has an average total fragmentation percentage of 96% with electronic 228 components in very good state and the average specific energy evaluated at $6.2 \pm 1.0 \text{ kWh/t}$. 229 The validated tests remain very promising for the design of a future experimental prototype, 230 particularly with a larger vessel more suited to LED devices. This potential solution will also make it possible to increase the distances between the electrodes, which will be better 231 232 adapted. The idea is to guarantee an efficient electric discharge path in water and 233 consequently develop an electric arc of selective fragmentation that is sufficient for the 234 treatment of all types of LED lamps. This is an essential criterion for recycling companies.

235 Moreover, the notion of reproducibility for the seven series of tests was evaluated. This 236 relates to the total separation of the drivers and the LED modules with at least 50% of the 237 LED chips still found in the aluminium modules as shown in Fig. 4-h. The average 238 reproducibility is estimated to be 78% for LED supports and 64% for PCBs.

An estimation of the reproducibility is essential to evaluate whether a series of tests has been validated. In fact, differences were observed for tests conducted with strictly identical operating modes. Significant variations were found in terms of the separation of the constituent elements:

LED supports released with all the LED chips attached or detached;

LED supports not released with the LED chips detached or sometimes even fractured;

245 Entire separate drivers and in a relatively good state;

Drivers not separated and still soldered or nested to the LED modules and bases, and finally,exploded drivers.

For this reason, we counted the number of LED chips that remained attached to the LED modules after the selective fragmentation. Overall, 75% of the LED chips were still available after the selective fragmentation. For their optimal recovery in the reuse or recycling scenario, it would be preferable to keep the LED chips on their aluminium supports. This would facilitate the sorting step and thus increase the recovery rate. In addition, keeping the LED chips on their LED modules, as close as possible to their initial forms and states, helps prevent losses of mass and energy (Reuter and Van Schaik, 2015).

The plastic E27 LED lamps with large diameter bulbs could not be fragmented. However, the validated parameters of this study should be applicable to all LED devices composed of plastic bulbs and plastic-coated aluminium bodies such as E27, B22, and E14 LED devices (about 55% of deposits; Fig. 2-c) (Martins et al., 2020).

The reproducibility criteria used for the separation of components ensures their recyclability in an industrial recycling process which, in turn, could be economically viable and profitable. The separation and subsequent recycling processes help make the overall process and sector more environmentally friendly. It is worth mentioning that the eco-design strategies used for LED lamps is also essential in order to efficiently dismantle these new light products (Dzombak et al., 2017; Pilotto Cenci et al., 2020; Martins et al., 2020).

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267 **3.2.** Quality of the bodies and electronic components

268 **3.2.1. Results**

We also considered the state of the aluminium bodies covered with a polymeric coating composed of polybutylene terephthalate (PBT) (Martins et al., 2020). A visual analysis was performed (Fig. 5) on all the sample bodies. The examination criteria were perforation and deformation due to electrical discharges.

273 In the validated series (voltage and pulses), 97% of the bodies were deformed and 76% were 274 pierced. Therefore, in the case of validated voltage-pulse pairs, the fragmentation energies 275 were optimized. The electric discharges passed through the samples to obtain favorable 276 selective separations for the entire study.

The observations were also made with regard to the electronic components, which constitute the added value of such LED devices. For this reason, we analyzed the fragmentation of the LED modules and the drivers (Fig. 6). In the two analyses of the series of validated voltage-pulse pairs, the fragmentation criteria correspond to the states of the electronic components at the end of the tests, which could be excellent, good, bad, or very bad. In the case of the LED modules, the criteria were whether or not the LED chips remained attached to their aluminium support. In the case of drivers, we visually examined
the state of the electronic components (coils, capacitors, transformers, etc.).

Fig. 6 summarizes the statistical study conducted on the electronic components, namely the LED modules and the drivers. The main idea was to classify the disassembled parts according to their condition.

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- 289
- 290

291 **3.2.2. Discussion**

Regarding the LED modules (Fig. 6-a to 6-e), it was found on average 64% were in excellent 292 condition, meaning that 75% to 100% of the LED chips are still positioned on the module. 293 294 This value can be compared to the average percentage obtained when considering the overall sampling material (i.e., 56%). Furthermore, 15%, 10%, and 10% of the LED modules 295 were respectively in a good (50-75%), bad (25-50%), and very bad condition (0-25%) (vs 13%, 296 297 10%, and 21% for the overall sampling material). It should therefore be noted that, on 298 average, 79% of the LED modules are in excellent or good condition (vs 69% of the overall 299 sampling material). By contrast, 20% are in a bad or very bad condition (vs 31% for the overall sampling material). 300

Considering these findings, we show that using the same parameters detailed in Table 1, two-thirds of the LED lamps can be treated; in other words, the lamps can be disassembled without deteriorating the LED modules.

As shown in Fig. 6-f to 6-j, the same kind of analysis can be applied to the drivers. Our findings show that on average 31% of the drivers were in excellent condition meaning that 75% to 100% of the drivers were undamaged, which compares to the 25% for the overall

307 sampling material. Furthermore, 34%, 10%, and 25% of the drivers were respectively in a 308 good, bad, and very bad condition (vs 28%, 17%, and 31% for the overall sampling material). 309 Therefore, it was found on average that 65% of the drivers were in excellent or good 310 condition (vs 53% for the overall sampling material). These results show that EDF was less 311 effective for the drivers than for the LED modules, although the technology is still relevant.

312 To conclude, the comparisons with the overall sampling material shows the effectiveness of 313 the determined parameters (Table 1) for sample fragmentation. Considering these two 314 analyses, we identified the optimal conditions to dismantle the electronic components while preventing their degradation. Additionally, having the components as close as possible to 315 their initial state will help minimize mass and energy losses in the new proposed recycling 316 317 process (Reuter and Van Schaik, 2015; Pilotto Cenci et al., 2020). At the end of the tests, we 318 observed that when the LED chips were still positioned on the aluminium support, they were found to be functional. A more systematic study is currently underway to assess the effect of 319 fragmentation treatment. This observation could nevertheless favor the reuse and/or the 320 321 material recovery (recycling) of the LED chips. Regarding the drivers, based on our 322 observations, recycling seems preferable. In fact, the damage caused by selective EDF 323 deteriorates the electronic components of the drivers in such a way that they cannot be reused as such. To consider a reuse scenario for the drivers or their components, only the 324 325 manual separation of the LED devices is conceivable at this stage.

A final aspect relates to a technical constraint in the treatment of this type of LED product using selective fragmentation. This concerns the inclusion of air in the bulbs, which forms an "air chamber" and prevents the lamp from being correctly positioned in the reactor. A piercing action is thus needed to avoid the floating of the plastic E27 LED devices. If not, this situation would cause a short circuit in which the path of the electrical discharge would only

partially impact the bulb, or even not at all. All the electrical energy would then be
concentrated on the ground electrode. It is worth noting that in the case of EHF, the final
shock wave fragments the samples by generating internal stress such as compressive strain
(Vaasjoki and Lindqvist, 2001; Justin Martin, 2013; Touzé et al., 2017; Pestalozzi et al., 2018).

336 **3.3. Materials recovery and recycling rate**

337 **3.3.1. Results**

Fig. 7-a shows the quantities and percentages of the recovered material for the total study 338 with all different energy levels. The studied sample series is composed of plastic E27 LED 339 lamps, which will represent more than 30% of deposits over the next decade (ECOSYSTEM, 340 2019). In total, 34.2 g of LED chips, 1231.1 g of LED modules, and 923.6 g of drivers (Fig. 7-b) 341 342 were recovered. Assuming that our samples are comparable to those of Pilotto Cenci et al. (2020), we applied the material coefficients to the recovered electronic components of the 343 plastic E27 LED lamps to estimate the quantities of precious metals. The equation considered 344 for this analysis is below (2). 345

346 Amount of precious metal
$$j = \sum_{i=1}^{n=3} (Masses of electrical components_i x metal rate ij) (2)$$

The Sankey diagram visualizes these results (Fig. 7-b). The main idea is to integrate the LED lamps into a loop (Baldé et al., 2017) with the aim of reusing and/or recycling them using an efficient and safe process that mitigates environmental impacts and generates economic benefits. Using this integrated closed-loop circuit, waste management will be more straightforward and efficient by minimizing the generation of waste or increasing the economic benefit by considering e-waste as a real source of secondary raw materials (Pilotto Cenci et al., 2020). 354 Regarding plastic E27 LED lamps, their valuable materials include precious metals, some of which are considered critical with a high supply risk (Hennebel et al., 2015; Isildar et al., 355 2018). Gold, silver, copper, gallium, indium, and rare earth elements can be found in the LED 356 chips, LED modules, and drivers (Reuter and Van Schaik, 2015; Franz and Wenzl, 2017; 357 358 Pourhossein and Mousavi, 2018). The mass fractions of these materials in plastic E27 LED 359 lamps were estimated to be 0.052%, 0.482%, and 0.381% for gold, silver, and gallium, 360 respectively (Pilotto Cenci et al., 2020). These recoverable materials are mainly found in LED 361 chips. Nevertheless, due to the lack of specific recycling processes for LED lamps (Mizanur Rahman et al., 2017), any new technological process to recover such materials should take 362 economic viability into account. The current study aimed at understanding how the 363 electronic components can be dismantled using EDF tests. 364

365

366 **3.3.2. Discussion**

Based on the results of the fragmentation study and the percentage composition as shown 367 in Fig. 7-a, let us estimate the recoverable fraction of materials in 1,000 kg of plastic E27 LED 368 369 lamps. The first step of the recycling process is the carcass segregation (optical systems, 370 bodies, and bases), which represent around 720 kg. The second step involves generating a material flow of the LED modules, drivers, and LED chips (around 280 kg). In supplementary 371 372 data, Fig. S2 summarizes the mass fractions of precious metal ores. More precisely, segregating the carcasses of 1,000 kg of LED lamps results in 160.0 kg of LED modules, 120.0 373 374 kg of drivers, and 4.4 kg of LED chips. From 284.4 kg of components, only 158.8 kg of 375 precious metals is recoverable, corresponding to 102.4 kg in LED modules, 53.4 kg in drivers, 376 and 3.0 kg in LED chips. Hence, the recycling rates of the components are 64.0% for LED 377 modules, 44.5% for drivers, and 68.4% for LED chips. Other studies have reported a 28%

378 recycling rate for cellphones (Reuter et al., 2018), 30% for PCBs (Isildar et al., 2018), and 30% 379 for LCD screens (Van Schaik and Reuter, 2016). In terms of the economic value, the 380 components and materials with the largest mass fraction may not be the most economically 381 important in terms of recycling (Pilotto Cenci et al., 2020). It is worth analyzing the main 382 components and materials from an economic perspective.

383 Gold is the most valuable material (Murakami et al., 2015), while the LED chips are the most 384 valuable components. Despite the low quantity of gold in LED lamps, it remains the most 385 important material from an economic perspective. Indeed, gold is potentially 0.03% (71 g) of the electronic component mass (ADEME, 2017) but it represents 83.6% (Magazine 386 RECYCLAGE RECUPERATION, 2020) of the total commercial value, whereas aluminium is 387 33.1% (94.1 kg) of the electronic component mass but it only represents 3.0% of the total 388 389 commercial value. The small portion of gold in the LED lamps is therefore relevant in terms of the total economic value of the product. These results for the economic strategy of 390 recovering gold from LED lamps were also mentioned in the study of Kumar et al., 2019. 391

In addition, tin and copper have an important distributed mass fraction in the electronic components, and they are interesting materials from an economic perspective. The mass fraction of tin is 8.9% (25.4 kg), representing 9.2% of the total commercial value. As for copper, it is 9.0% (25.6 kg) of the total mass fraction for 3.5% of the total commercial value.

Moreover, it is relevant to define the most valuable electronic components according to the normalized economic value per kilogram of LED chips, LED modules, and drivers. Even though the LED chips are the electronic component with the smallest mass fraction, this product is the most valuable component compared to the LED modules (47% of the value of the LED chips) and the drivers (39% of the value of the LED chips). This highlights the importance of using efficient industrial processes to separate the components to ensure and 402 improve the recovery of highly valuable materials and hence maximize the mass fractions of403 recovered materials.

The concentration of materials in e-waste represents a potential of €55 billion with €40 billion of metals (Baldé et al., 2017). This requires appropriate industrial processes to recycle the materials (Kumar et al., 2017). Besides the intrinsic value of this secondary source of materials, recycling also reduces the need for primary raw materials (Pilotto Cenci et al., 2020). This kind of analysis allows recycling companies to define their industrial strategy and concentrate on the most profitable secondary raw materials depending on geopolitical factors.

411

412 **3.4. Recommendations to improve the recycling process**

413 The selective fragmentation of plastic LED lamps in this study demonstrated the need to 414 focus on the recovery of electronic components which contain precious metals to increase 415 economic profits. The process will be all the more efficient with eco-designed products 416 whose fragmentation would be facilitated. Thus, the recovery of precious metals would be 417 easier to sort and recycle. The added value of these products is contained in their electronic 418 components and the pulsed powers allow separation of these products while avoiding their physical, and therefore potentially functional, deterioration. These components contain the 419 420 precious metals which define the economic profitability.

The expected increase in the volume of LED lamp waste, as well as their wide variety should lead to adapting the process to operate in industrial conditions of continuous treatment. For example, to achieve a flow rate of up to 10 t/h and to accept up to 7 types of LED lamps (Fig. 1-a) in the same stream. This observation is encouraging for an industrial-type device.

425

426 **4. Conclusion**

The main objective of this study was to identify the optimal conditions for disassembling, and estimate the recycling potential, of LED-based lighting devices. As a test sample, we chose plastic E27 LED lamps with the goal of reaching a global recycling rate of 80%. The main results obtained in this study are:

The efficient separation of the components was achieved without any pretreatment such asgrinding;

433 Implementation of a protocol to qualify defragmentation as acceptable or not for recycling434 companies;

The average specific energy to achieve selective and efficient fragmentation was 5.2 ± 0.6 kWh/t, with the average overall separation estimated at 64%. These results to be compared with the 152 kWh/t of the shredding process, with a segregation percentage close to zero;

Gold represents 0.03% and 83.6% of the mass and commercial value of the recoveredelectronic components respectively.

In order to control the quality of the fragmentation, we could use artificial intelligence, based on image analysis, and recognize the optimal fragmentation of the components to sort and recover the under-products which not be correctly fragmented. They would be retreated by loops in the process. This technology would improve the industrial process.

444 Considering these findings, EDF emerges as a very promising process for LED lamp recycling, 445 since it allows a better valorization of the components through their preliminary 446 disassembly. This research highlights the relevance of recycling this new complex e-waste 447 and the opportunities provided by new technological alternatives.

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456 **Conflicts of Interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

459

460 Supplementary data

461 Supplementary data associated with this article can be found in the online version

462 **References**

463

- 464 ADEME « Equipements électriques et électroniques » rapport annuel 2017.
- 465 https://www.ademe.fr/sites/default/files/assets/documents/registre-eee-donnees-2017-201711-
- 466 rapport.pdf
- 467

Aman, M.M., Jasmon, G.B., Mokhlis, H., Bakar, A.H.A., 2013. Analysis of the performance of
domestic lighting lamps. Energy Policy. Volume 52, pages 482-500.
https://doi.org/10.1016/j.enpol.2012.09.068.

471

Baldé, C.P., Forti, V., Gray, V., Kuehr, R., Stegmann, P., 2017. The global e-waste monitor 2017, United Nations University (UNU), International Telecommunication Union (ITU) &
International Solid Waste Association (ISWA). Bonn/Geneva/Vienna.
https://doi.org/10.1016/j.proci.2014.05.148.

476

Bru K., Touzé S., Auger P., Dobrusky S., Tierrie J., Daniel B. Parvaz., 2018. Investigation of lab
and pilot scale electric-pulse fragmentation systems for the recycling of ultra-high
performance fibre-reinforced concrete. Minerals Engineering 128, 187-194.
https://doi.org/10.1016/j.mineng.2018.08.040.

481

Bruno Gayral, 2017. LEDs for lighting: Basic physics and prospects for energy savings.
Comptes Rendus Physique 18 pages 453-461. https://doi.org/10.1016/j.crhy.2017.09.001

Cheng, S.Z.D., 2002. Handbook of thermal analysis and calorimetry. Volume 3, Applications
to polymers and plastics. Elsevier. Department of polymer science university of Akron, OH
44325-3909 USA.

488

Doulos, L.T., Kontadakis, A., Madias, E.N., Sinou, M., Tsangrassoulis, A., 2019. Minimizing 489 490 energy consumption for artificial lighting in a typical classroom of a Hellenic public school aiming for near Zero Energy Building using LED DC luminaires and daylight harvesting 491 492 systems. Energy & Buildings 194 201-217. pages https://doi.org/10.1016/j.enbuild.2019.04.033 493

494

Duan, C., Han, J., Zhao, S., Gao, Z., Qiao, J., Yan, G., 2018. The stripping effect of using high
voltage electrical pulses breakage for waste printed circuit boards. Waste Management 77
(2018) 603-610. https://doi.org/10.1016/j.wasman.2018.06.003.

498

Dussaigne, A., Barbier, F., Damilano, B., Chenot, S., Grenier, A., Papon, A. M., Samuel, B., Ben
Bakir, B., Vaufrey, D., Pillet, J. C., Gasse, A., Ledoux, O., Rozhavskaya, M., and Sotta, D., 2020.
Full InGaN red light emitting diodes. Journal of Applied Physics 128, 134704.
https://doi.org/10.1063/5.0016217

503

Dzombak, R., Padon. J., Salsbury, J., Dillon, H., 2017. Assessment of end-of-life design in solid-state lighting. Environ. Res. Lett. 12, 084013. https://doi.org/10.1088/1748-9326/aa7ab1.

507

508	ECOSYSTEM. Le Recyclage Des Lampes Et Luminaires à LED, 2019. Available online:
509	https://www.recylum.com/recyclage-lampes-luminaires-leds/ (accessed 5 August 2020).
510	

European Commission, 2009. FAQ: Phasing out Conventional Incandescent Lamps. Available
online: http://europa.eu/rapid/press-release_MEMO-09-368_en.htm (accessed 23 August
2020).

514

Franz, M., Wenzl, F.P., 2017. Critical review on life cycle inventories and environmental
assessments of led-lamps. Crit. Rev. Environ. Sci. Technol. 47, 2017-2078.
https://doi.org/10.1080/10643389.2017.1370989.

518

Fouquet, Y., Martel-Jantin, B., 2014. Rare and Strategic Metals. Deep Marine Mineral
Resources. Springer, Dordrecht. https://doi.org/10.1007/978-94-017-8563-1_3.

521

Hennebel, T., Boon, N., Maes, S., Lenz, M., 2015. Biotechnologies for critical raw material
recovery from primary and secondary sources: R&D priorities and future perspectives. N.
Biotechnol. 32, 121-127. https://doi.org/10.1016/j.nbt.2013.08.004.

525

526 Isildar, A., Rene, E.R., Van Hullebusch, E.D., Lens, P.N., 2018. Electronic waste as a secondary

527 source of critical metals: Management and recovery technologies. Resour. Conserv. Recycl.

528 135, 296-312. https://doi.org/10.1016/j.resconrec.2017.07.031.

Justin Martin. Study and characterization of pressure waves generated by an electric discharge in water. Application to the electric fracturing of rocks, thesis defended on June 14, 2013 at the University of Pau et des Pays de Adour.

533

Kumar, A., Holuszko, M., Espinosa, D.C.R., 2017. E-waste: An overview on generation,
collection, legislation and recycling practices. Resour. Conserv. Recycl. 122, 32-42.
https://doi.org/10.1016/j.resconrec.2017.01.018.

537

Kumar, A., Kuppusamy, V.K., Holuszko, M., Song, S., Loschiavo, A., 2019. LED lamps waste in
Canada: Generation and characterization. Resour. Conserv. Recycl. 146, 329-336.
https://doi.org/10.1016/j.resconrec.2019.04.006.

541

Lim, S.R., Kang, D., Ogunseitan, O.A., Schoenung, J.M., 2011. Potential environmental impacts of light-emitting diodes (LEDs): Metallic ressources, toxicity, and hazardous waste classification. Environ. Sci. Technol. 45, 320-327. https://doi.org/10.1021/es101052q.

545

Lim, S.R., Kang, D., Ogunseitan, O.A., Schoenung, J.M., 2013. Potential environmental impacts from the metals in incandescent, compact fluorescent lamp (CFL), and light-emitting diode (LED) bulbs. Environ. Sci. Technol. 47, 1040-1047. https://doi.org/10.1021/es302886m.

550

551 Magazine « RECYCLAGE RECUPERATION » - n°38 - 16 November 2020 - Recycled materials
552 prices.

553

554 Martins, T.R., Tanabe, E.H., Bertuol, D.A., 2020. Innovative method for the recycling of end-555 of-life LED bulbs by mechanical processing. Resources, Conservation & Recycling. 161 556 104875. https://doi.org/10.1016/j.resconrec.2020.104875.

557

558 Marwede, M., Chancerel, P., Deubzer, O., Jordan, R., Nissen, F.N., Lang, K.D., 2012. 559 Electronics Goes Green 2012+ Joint International Conference and Exhibition September 9 -560 12, 2012 in Berlin, Seminaris Dahlem Cube. Fraunhofer. Technische Universität Berlin. 561 cycled.

562

Mizanur Rahman, S.M., Kim, J., Lerondel, G., Bouzidi, Y., Nomenyo, K., Clerget, L., 2017.
Missing research focus in end-of-life management of light-emitting diode (LED) lamps.
Resour. Converv. Recycl. 127, 256-258. https://doi.org/10.1016/j.resconrec.2017.04.013.

566

Mizanur Rahman, S.M., Kim J., Lerondel G., Bouzidi Y., Clerget L., 2019. Value retention
options in circular economy: Issues and challenges of LED lamp preprocessing. Sustainability
11, 4723. https://doi.org/10.3390/su11174723.

570

571 Mohammad Hossein Alam Varzaneh Isfahani & Rahim Faez, 2021. The most optimal barrier 572 height of InGaN light-emitting diodes. Applied Physics A pages 127-144. 573 https://doi.org/10.1007/s00339-021-04306-1

574

575 Murakami, H., Nishihama, S., Yoshizuka, K., 2015. Separation and recovery of gold from 576 waste LED using ion exchange method. Hydrometallurgy 157, 194-198. 577 https://doi.org/10.1016/j.hydromet.2015.08.014.

Narendran, N.; Bullough, J.D.; Maliyagoda, N.; Bierman, A. What is useful life for white light
LEDs? Journal of the Illuminating Engineering Society. 2001, 30, 57-67.
https://doi.org/10.1080/00994480.2001.10748334.

Natthanon Phannil, Chaiyan Jettanasen and Atthapol Ngaopitakkul, 2018. Harmonics and
Reduction of Energy Consumption in Lighting Systems by Using LED Lamps. Energies 2018,
11, 3169. https://doi.org/10.3390/en11113169

586

Nevala, S.M., Hamuyuni, J., Junnila, T., Sirviö, T., Eisert, S., Wilson, B.P., Serna-Guerrero, R.,
Lundström, M., 2019. Electro-hydraulic fragmentation vs conventional crushing of
photovoltaic panels - Impact on recycling. Waste Management. 87 (2019), 43-50.
https://doi.org/10.1016/j.wasman.2019.01.039.

Nomenyo, K,. Gadallah, A., Kostcheev, S., Rogers, D., Lerondel, G., 2014. Enhanced
stimulated emission in ZnO thin films using microdisk top-down structuring. Appl. Phys. Lett.
104(18), 181104. https://doi.org/10.1063/1.4875744.

594

Pestalozzi, F., Stefan, E., Woidasky Jörg, W., 2018. Benchmark Comparison of High Voltage
Discharge Separation of Photovoltaic Modules by Electrohydraulic and Electrodynamic
Fragmentation. MDPI recycling. https://doi.org/10.3390/recycling3020013.

598

Pilotto Censi, M., Christ Dal Berto, F., Schneider, E-L., Veit, H-M., 2020. Assessment of LED
lamps components and materials for a recycling perspective. Waste Management 107 (2020)
285-293. https://doi.org/10.1016/j.wasman.2020.04.028.

- Pourhossein, F., Mousavi, S.M., 2018. Enhancement of copper, nickel, and gallium recovery
 from LED waste by adaptation of Acidithiobacillus ferrooxidans. Waste Management 79
 (2018) 98-108. https://doi.org/10.1016/j.wasman.2018.07.010.
- 606
- Reuter, M.A., Van Schaik, A., 2015. Product-centric simulation-based design for recycling:
 Case of LED lamp recycling. J. Sustain. Metall. 1,4-28. https://doi.org/10.1007/s40831-0140006-0.

610

Reuter, M.A., Ballester, M., Van Schaik, A., 2018. Limits of the circular economy: Fairphone
modular design pushing the limits. World Metall. 71, 68-79.

613

Richter, J.L., Van Buskirk, R., Dalhammar, C., Bennich, P., 2019. Optimal durability in least life
cycle cost methods: The case of LED lamps. Energy Effic 12, 107-121.
https://doi.org/10.1007/s12053-018-9662-4.

617

Schubert, E.F., Kim, J.K., 2005. Solid-State Light Sources Getting Smart. Science 308, 12741278. https://doi.org/10.1126/science.1108712.

620

SELFRAG. Waste to value. Electrodynamic fragmentation technology. Available online:
http://www.selfrag.com/ (accessed 5 December 2020).

623

Shameem Ahmada, Shalendra Kumarb, Savaş Kayac, Alvid, P.A., Siddiquia, M.J., 2020.
Improvement in efficiency of yellow Light Emitting Diode using InGaN barriers and modified

electron injection layer. International Journal for Light and Electron Optics 206, 163716.
https://doi.org/10.1016/j.ijleo.2019.163716

628

- Shi, F., Manlapig, E., Zuo, W., 2014. Progress and Challenges in Electrical Comminution by
 High-Voltage Pulses. Chemical Engineering Technology. 37, No. 5, 765-769.
- 631 https://doi.org/10.1002/ceat.201300660
- Song, B.P., Zhang, M.Y., Fan, Y., Jiang, L., Kang, J., Gou, T.T., Zhang, C.L., Yang, N., Zhang, G.J.,
 Zhou, X., 2020. Recycling experimental investigation on end of life photovoltaic panels by
 application of high voltage fragmentation. Waste Management 11 (2020) 180-187.
 https://doi.org/10.1016/j.wasman.2019.10.015.

636

Tokoro, C., Lim, S., Teruya, K., Kondo, M., Mochidzuki, K., Namihira, T., Kikuchi, Y., 2021.
Separation of cathode particles and aluminum current foil in Lithium-Ion battery by highvoltage pulsed discharge Part I: Experimental investigation. Waste Management 125 (2021)
58-66. https://doi.org/10.1016/j.wasman.2021.01.008.

641

Touzé, S., Bru, K., Ménard, Y., Weh, A., Von der Weid, F., 2017. Electrical fragmentation
applied to the recycling of concrete waste - Effect on aggregate liberation. International
Journal of Mineral Processing 158, 68-75. https://doi.org/10.1016/j.minpro.2016.11.009.

645

Vaasjoki, M., Lindqvist, K., 2001. Electrodynamic fragmentation tests aimed at separation of
mineral species for mineralogical and isotopic. Mineralogical laboratory. M41.1/2001/2.

Van Schaik, A., Reuter, M.A., 2016. Recycling indices visualizing the performance of the
circular economy. World Metall. 69, 5-20.

651

- Van der Wielen, K.P., Pascoe, R., Weh, A., Wall, F., Rollinson, G., 2013. The influence of
 equipment settings and rock properties on high voltage breakage. Minerals Engineering 4647 (2013) 100-111. https://doi.org/10.1016/j.mineng.2013.02.008
- Zhang, L., Li, Q., Shang, L., Wang, F., Qu, C., Zhao, F., 2013. Improvement of UV
 electroluminescence of n-ZnO/p-GaN heterojunction LED by ZnS interlayer. Optics Express
 21(14), 16578-16583. https://doi.org/10.1364/OE.21.016578.
- 658
- 259 Zhao, Y., Zhang, B., Duan, C., Chen, X., Sun, S., 2015. Material port fractal of fragmentation of
- waste printed circuit boards (WPCBs) by high-voltage pulse. Powder Technology 269 (2015)
- 661 219-226. https://doi.org/10.1016/j.powtec.2014.09.006.







Fig. 1. Deposit study: Typology of the six big bags analyzed between September 2019 and June 2020 (a),
photographs of small plastic E27 LED lamps (b), and average typology of the big bags (c).



Fig. 2. Laboratory machine: Schematic representation (a) and photograph (b) of the SELFRAG unit and diagram
showing the plastic E27 LED lamp in the vessel (c).

-





(b)

(c)



Fig. 3. Fragmentation tests: Mapping of the fragmentation tests (a), non-validated fragmentation at 150 kV
with 15 pulses (b) and 150 kV with 5 pulses (c), and validated fragmentation at 150 kV with 4 pulses (d) and 140
kV with 5 pulses (e).



Fig. 4. Sample study: Unfractured bulb (a), fractured bulb (b), unfractured driver (c), fractured driver (d),
 fractured body (e), unfractured base (f), fractured base (g), and reproducibility of the selective fragmentation
 for the seven validated series (h).





Fig. 5. Photographs of the bodies: undamaged body (a), deformed but unpierced body (b), deformed and
 pierced body (c), and state of bases for the seven series (d).

- - -





(e)





Fig. 6. Electronic components in 7.7 kg of LED lamps: Examples of LED modules classified as a function of the
 remaining number of LED chips with 100% (a), 67% (b), 44% (c), and 0% (d) of LEDs remaining on the module.
 Classification of LED modules condition by 25% steps depending on the number of chips remaining according to
 the seven series of EDF treatments (e). Examples of drivers classified as a function of their condition:
 unfractured (f), weakly fractured (g), significantly fractured (h), and totally fractured (i). Classification of the
 driver conditions according to the seven series of EDF treatments (j).



Fig. 7. Quantities investigation of the constituent elements that are potentially recoverable by selective
 fragmentation: Percentage graph showing the composition of plastic E27 LED lamps (a). Sankey mass diagram
 of precious metals recovered from LED modules, drivers and chips in 7.7 kg of plastic E27 LED lamps (b).

Table 1

Summary of the selective fragmentation results for the different constituent elements and the corresponding

750 parameters (voltage and number of pulses). In supplementary data, Fig. S1 shows the photographs.

Tests series	Voltage (kV)	Pulses	Average specific energy (kWh/t)	Bulbs (%)	LED Modules (%)	Drivers (%)	Total output (%)
Α	145	5	5.7	60	70	80	70
В	140	6	6.2	88	100	100	96
С	140	5	5.5	25	50	100	58
D	140	4	4.6	63	38	38	46
E	145	4	5.0	38	50	75	54
F	150	4	4.8	75	50	75	67
G	155	4	4.9	63	63	38	54
Average series A-G	-	-	5.2	59	60	72	64