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# Urban Mining of Unexploited Spent Critical Metals from E-waste Made Possible Using Advanced Sorting

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- 15
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- 18

#### 19 Abstract

20 The growing number of electronic devices has led to a surge in e-waste, making efficient recycling essential to reduce environmental impact and recover valuable metals. However, 21 traditional recycling methods struggle to extract them due to their low concentrations in e-22 23 waste. Here, we developed a system to sort electronic components from printed circuit boards 24 by elemental composition. It combines a convolutional neural network-based optical recognition with multi-energy X-ray transmission spectroscopy, demonstrating up to 96.9% 25 26 accuracy in controlled conditions. Hence, with elemental enrichments by up to 10,000 for targeted elements, this method renders economically viable the recovery of previously 27 unrecycled critical metals by enriching sorting bags in precious, semi-precious, refractory (Ta, 28 Nb), transition (Co, Cr, Mn, Ni, Zn, Ga, Bi, etc.) or other (In, Sn, Sb) metals. These findings 29 demonstrate the promising applications of this technology in mitigating the environmental 30 impact of e-waste and promoting the sustainable recovery of valuable metals. 31

32

#### 33 1. INTRODUCTION

34 The technological development and the 35 automation of our modern society is 36 supported by an exponential increase in 37 our use of electronics, which leads to a 38 similar increase in the amount of waste 39 from electrical and electronic equipment 40 (WEEE) (Forti et al. 2020). 41 Manufacturing these devices requires a 42 considerable volume of raw materials 43 including valuable ones (gold, palladium, 44 silver...), critical elements (rare earth 45 elements (REEs) (Binnemans, 46 McGuiness, and Jones 2021), tantalum, cobalt...) as well as hazardous products 47 (lead, chromium, bromine...) (Wang and 48 Xu 2015). Thus, making the electronic 49 industry (considered from cradle to cradle) 50 of the most environmentally 51 one impacting (Shakil, Nawaz, and Sadef 52 53 2023).

54 A good way to reduce this industry's55 footprint would be to increase the56 efficiency of its recycling processes. But

1 for this to happen, the major issue of the elemental complexity of the WEEEs' 2 composition needs to be addressed. On 3 4 that front, printed circuit boards (PCBs) (Gorewoda et al. 2020), hold the record 5 with more than sixty chemical elements 6 7 that can be found in them (Maurice et al. 2021). Current methods of treating waste 8 9 PCBs (WPCBs), which consist in first crushing and grinding them, produce a 10 powder with a highly complex and 11 heterogeneous chemical composition. This 12 is then followed by physical separation of 13 some particles (Kaya 2017) and 14 the pyrometallurgical 15 subsequent and hydrometallurgical process steps. With 16 such an approach, although 60% of the 17 value can be retrieved from the WPCBs, it 18 only recycles one quarter of the chemical 19 elements it contains, either the most 20 concentrated or the most valuable (C, Cu, 21 Ni, Fe, Al, Ti, Cr, Au, Pt, Ag, Pd, Pb, Sn) 22 (Wu et al. 2017). Indeed, with such 23 processing, most other elements of interest 24 become too diluted in the overall waste 25 (down to a few ppm levels) and are 26 therefore impossible to extract in an 27 economically viable way. These elements 28 are therefore lost as ash and disposed of 29 through landfilling (U.S. Environmental 30 Protection Agency and Conservation 31 32 2020).

To more effectively access these spent 33 chemical elements an alternative strategy 34 is to dismantle WPCBs to recover the 35 36 electronic components (ECs) and sort 37 them according to their type and chemical composition (Maurice et al. 2021). Recent 38 39 studies have therefore explored some possible electronic components sorting 40 41 and subsequent refining processes, such as 42 optical sorting based on machine vision coupled with artificial intelligence (for 43 example, convolutional neural network -44 CNN). The latter has shown promising 45 46 results for the classification of ECs

47 (Lefkaditis and Tsirigotis 2010; Lu et al. 2022). So far, these methods have been 48 limited to only a few EC categories, 49 50 giving access to only a few more chemical elements. Other processes have privileged 51 a selective robotic disassembly (pick and 52 choose approach), which requires high 53 54 precision for removing the targeted ECs. However, with a speed of a few tenths of a 55 second per component retrieved, it can 56 only be applied to high value ECs to be 57 economically viable (Sauer et al. 2019). 58 However, despite significant 59 the improvement that these recent innovations 60 have brought, these EC sorting techniques 61 all rely on optical recognition, which has 62 its own limitations. Indeed, while it is 63 effective when components looking alike 64 have similar composition (Kopacek 2016), 65 it does not allow for elemental separation 66 when components look similar but have 67 different 68 chemical compositions. Moreover, the elemental composition of 69 PCBs can vary significantly due to their 70 71 diverse origins, as highlighted by previous studies (Smalcerz et al. 2023). For 72 example, this is observed with capacitors, 73 multilayer ceramic capacitors or resistors. 74 To tackle this issue, various spectroscopic 75 methods have been developed such as 76 near infrared (NIR) hyperspectral image 77 78 spectroscopy (HSI) (Rapolti et al. 2021) 79 or laser-induced breakdown spectroscopy (LIBS) (Zeng et al. 2021) giving access to 80 81 some elemental composition of the 82 components' surfaces. Using either spectra analysis or artificial intelligence 83 these methods have shown high accuracy 84 85 in the targeting of specific elements. Other 86 recent studies have showed great progresses in the recognition of e-waste 87 parts by combing optical imaging and HSI 88 (Tan et al. 2022). However, all these 89 approaches can only analyse the surface 90 composition of ECs and do not capture all 91 the internal chemical composition and 92 elements. For this reason, these are useful 93

1 methods for the characterisation and sorting of homogeneous or monolithic 2 3 materials such as plastics. To get access to 4 some internal composition indication, few 5 energies (one to five) X-ray transmission 6 (XRT) spectroscopies have been used in a variety of fields such as mining (Veras et 7 8 al. 2020; Yeyu et al. 2021), food industry, 9 medical imaging (Mesina, de Jong, and 10 Dalmijn 2007) or even plastic waste sorting (Hennebert and Filella 2018), but 11 these approaches do not have enough 12 energy resolution to allow for the 13 elemental sorting of mixture as complex 14 as wasted electronic components from 15 WPCBs. Hence, this study where we 16 present a novel sorting method for ECs 17 using both optical and Multi-Energy XRT 18 (MEXRT) sorting (up to 128 energy 19 levels) in which ECs were sorted on a 20 custom-made prototype using a CNN for 21 22 image recognition followed by MEXRT 23 hyperspectral image spectroscopy 24 analysis. We will show that it allows for 25 the accurate classification of ECs based on 26 their type and elemental composition 27 leading to such an enrichment in many targeted elements, so as to provide access 28 previously unrecoverable 29 to spent elements. 30

#### 31 2. MATERIALS AND METHODS

#### 32 **2.1. Materials**

For this study, model ECs from various 33 manufacturers, with known specifications, 34 were purchased from the distributor RS 35 Components Singapore. They were then 36 37 mixed with other ECs retrieved from the disassembly of spent printed circuit 38 boards of various origins to form the 39 model samples for the study. These PCBs, 40 were extracted from end-of-life electronic 41 products, from various applications such 42 as mobile phones, domestic appliances, 43 computers, or industrial devices, by 44 manually dismantling the appliances. 45

46 These PCBs were then disassembled by
47 heating them in a furnace at ~200 °C for
48 10 minutes, to melt the solder, before
49 being manually shaken or hammered to
50 release and collect loose spent ECs. The
51 latter were used to form the initial e-waste
52 stream for the large-scale study.

Nitric acid (AR, 70 %), hydrochloric acid 53 (AR, 37%), and hydrogen peroxide 54 solution (3%), were purchased from 55 Sigma-Aldrich. Metal standard solutions 56 for ICP were purchased from PerkinElmer. 57 Ultra-pure water was obtained from a 58 water purification system (WaterPro®, 59 Labconco Co.) with a resistivity of 60 18.2 M $\Omega$ ·cm at 25 °C. All the chemicals 61 were used as received. 62

#### 63 **2.2. Sieving**

Once disassembled from the boards, ECs
were segregated into 5 size categories by
sieving. Using Retsch AS 300 shaker with
screens of 40 mm, 10 mm, 5 mm, and
1 mm and a vibration amplitude of 1 mm
for 5 minutes.

#### 70 **2.3. Feeding system**

Each fraction of ECs was then separately 71 loaded into a custom conical bowl feeder 72 (50 cm larger base diameter, 30 cm 73 smaller base diameter, and 20 cm height). 74 The vibration frequency was set up at the 75 harmonic frequency of the bowl for the 76 entire study (55.2 Hz). The amplitude of 77 the vibration is controlled by the input 78 79 voltage delivered by the controller (Variable Frequency Vibratory Feeder 80 Controller SDVC31-M by Nanjing CUH 81 Science and Technology Co., Ltd) and 82 tuned for each EC size to optimise the 83 feeding rate. The tension delivered was set 84 at 110 V for components between 10 mm 85 and 40 mm in size and 95 V for 86 components with a size between 5 mm 87 and 10 mm and 90 V for components with 88 a size between 1 mm and 5 mm. These 89

parameters where determined through an
 empirical investigation for consistent
 feeding of the ECs at a rate of 2 ECs/s. A
 home-made passive rotary feeder was
 installed at the exit of the bowl feeder to
 ensure that the ECs were dispensed one at
 a time onto the conveyor belt.

#### 8 **2.4.** Conveyor system

9 The ECs were conveyed on a custommade 10 cm width and 2.5 m long belt 10 mounted on a 90 cm high aluminium 11 frame. The belt speed was controlled by 12 adjusting the rotational speed of a DC 13 motor. In this study, the rotational speed 14 of the motor was set to 100 rpm for the 15 optical sorting and 57 rpm for MEXRT 16 sorting, which corresponds to a linear 17 18 speed of 26.3 cm/s and 15 cm/s,respectively, for ECs placed on the belt 19 (Figure 1). 20

#### 21 **2.5.** Vision system

Images of ECs were acquired using a 22 BU505MCF Telicam, Toshiba camera 23 coupled with a white LED ring (V2DR-24 i90A-W. 25 Vital Vision Technology). 26 Database of ECs pictures was built using sorted manually components from 27 WPCBs only used for the training of the 28 29 CNN. This database of 3720 images divided into 8 classes of ECs was then 30 used to train the CNN used for the ECs' 31 images classification during the sorting 32 process. Additionally, the dataset was 33 34 augmented by applying random 35 transformations such as rotation, flipping, and cropping to increase the size of the 36 dataset and improve the generalization 37 ability of the model. 70% of the database 38 was used for the training of the model 39 while 30% was used for the validation of 40 the trained model. The CNN was coded in 41 python (version 3.6.8) using the open-42 source library TensorFlow and the keras 43 44 module. The CNN used in this study was a custom implementation based on a 45

46 sequential architecture with three 2D
47 convolution blocks (with respectively 64,
48 128, 256 output filters) each of them
49 coupled with a max pool layer to reduce
50 the output volume.

# 51 2.6. Multi energy X-ray 52 transmission sorting

ECs placed on the conveyor belt were 53 54 forced to pass in between the X-ray X-RAY 55 generator, (VJ (NY) LLC MODEL: IXS160BP200P107 with 56 57 tungsten anode, running at 100 kV and 0.3 mA), and MEXRT detector 58 the (ME100, Detection Technology). 59 The latter was composed of a 128 pixels line 60 each having an acquisition range from 61 19.2 keV up to 160 keV with a 1.1 keV 62 63 binning. Both source-sample and sampledetector distances were 10 cm. For each 64 sample, 50 lines of pixels were acquired 65 on the fly (while the EC was passing 66 the above detector thanks to the 67 conveyor's 68 movement), with an acquisition time of 2 ms per line (which is 69 maximum exposure time that would not 70 cause the detector to become saturated 71 while the generator was operating at its 72 defined energy level), thus creating a 73 hyperspectral 3D image of the EC. To get 74 75 an image with as little distortion as 76 possible, the conveyor speed was set to 15 cm/s. It should be noted that to reduce 77 the data collection size, the acquisition of 78 the MEXRT detector was triggered by the 79 detection of an EC by the upstream optical 80 camera. 81

#### 82 **2.7. Data treatment**

For each pixel of the hyperspectral image, 83 the total number of photons was integrated, 84 giving a 2D grey scale image representing 85 the X-ray transmission of the ECs. The 86 pixel with the lowest photon count was 87 88 considered as being part of the EC and a 3\*3 square of pixels was extracted around 89 conduct the elemental 90 it to

1 characterisation. The k<sup>th</sup> component of the 2 average spectrum  $X \in \mathbb{N}^{128}$  was 3 calculated from the chosen pixels as 4 follow:

$$X_k = \frac{\sum_{i=1}^n \sum_{j=1}^n x_{i,j,k}}{n^2} \quad (1)$$

5 With x<sub>i,j,k</sub> ∈ N the k<sup>th</sup> components of the
6 X<sub>i,j</sub> ∈ N<sup>128</sup> spectrum corresponding to the
7 pixel p<sub>i,j</sub> and n the number of pixels of
8 the studied area (hence, with the region of
9 interest of size 3\*3 pixels used in this
10 study, the sample size n is equal to 3).
11 The average spectrum was then filtered

11 The average spectrum was then filtered
12 using the Savitzky-Golay filter (Python
13 SciPy library, window length = 11,
14 polynomial order = 3) to reduce noise. In
15 X-ray spectrometry, the k-edge energy is a
16 crucial parameter, representing the energy
17 required for an electron to transition from
18 a core level to an empty state in the

conduction band. This leads to a sharp 19 increase in the photons absorption, 20 resulting in a characteristic peak in the 21 spectrum. By comparing this peak's 22 energy with known k-edge energies of 23 different elements, we can identify their 24 presence. Local maxima of the spectra 25 were extracted and compared with the k-26 edge values of the different elements 27 studied with an error of  $\pm 1.1$  keV, to take 28 into account the binning effect, which 29 allows the differentiation of elements 30 whose k-edges are separated by 1.1 keV. 31

#### 32 **2.8. Pneumatic sorting system**

Once classified, ECs were ejected by 33 34 pneumatic nozzles (KNS-R02-100-4 used with compressed azote - 6 bar) from the 35 belt in different bins placed along the 36 37 conveyor belt. Solenoid valves (VQ21A1-5GZ-C6-F) were controlled using the 38 nidaqmx library. 39 python

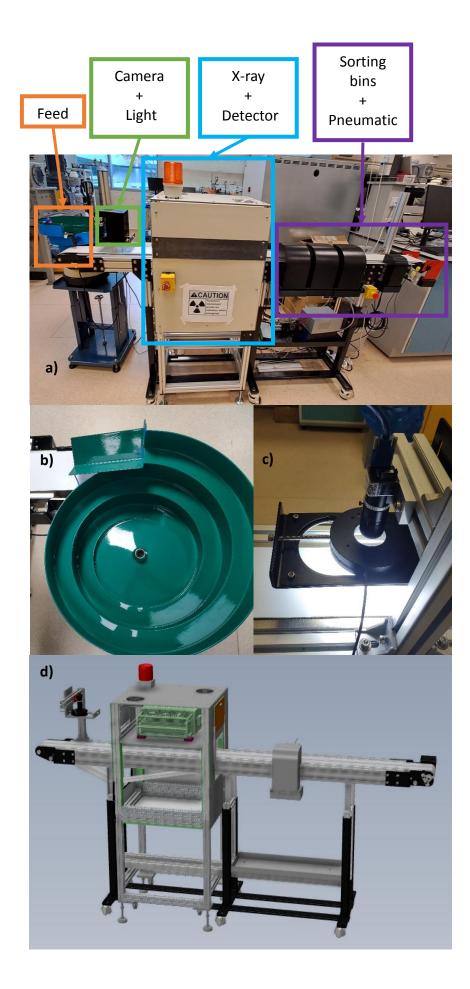


Figure 1: a) Photograph of the whole prototype, close view of the bowl feeder, c) close view of the camera and light and d) 3D CAD view of the prototype

#### 1 **2.9.** Sample characterisation

Sample preparation: Each type of sorted 2 electronic component was shredded 3 separately using a cutting mill (Retsch 4 SM200) with a built-in screen with 2 mm 5 square side holes. The shredded particles 6 were then subjected to either a grinder 7 (Retsch RS200) if they were grindable, or 8 a sample divider (Retsch PT100+DR100) 9 if they contained ductile materials to 10 further increase the compositional 11 homogeneity 12

13

**XRF analysis:** To identify the different 14 elements present in the sorted ECs, a first 15 characterisation of the powder was done 16 using X-ray fluorescence analysis (XRF). 17 The powder prepared was then analysed 18 using Vanta C series, Olympus XRF 19 20 detector (soil method using 3 beams 15 kV, 40 kV and 50 kV). 21

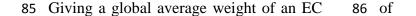
**ICP-OES** analysis: Samples 22 were analysed by Inductively Coupled Plasma-23 24 Optical Emission Spectrometry (ICP-OES). The samples were weighted 25 26 ( $\sim 0.20$  g) and digested in a mixture of 27 aqua regia (10 ml) and hydrogen peroxide (3%, 2ml) at 180 °C for 1 h using a 28 29 microwave digestion system (ETHOS UP, 30 MILESTONE). Metal contents in the 31 digested solutions were measured with an 32 ICP-OES spectrometer, (OP Optima 8000, 33 PerkinElmer) after adequate dilution. 34 Duplicate samples were prepared for each category and each sample was measured 35 by ICP-OES in triplicate so that the mean 36 values could be reported. 37 Standard deviations were estimated 38 by error propagation. 39

#### 40 3. Results and discussion

The cornerstone of the shift in process 41 42 paradigm is to start by a disassembly of the WPCBs, instead of their direct 43 grinding to obtain free spent ECs. As there 44 are many different disassembly processes 45 available at the pre-industrial or even 46 industrial level, which we recently 47 reviewed (Maurice et al. 2021), we did not 48 investigate this step further and emulated 49 the "heating + shaking" method whenever 50 necessary, following the method described 51 52 in the part 2.1

#### 53 **3.1. Sieving**

54 The first step in the sorting process was to separate the disassembled ECs into five 55 size categories using sieves with apertures 56 of 40 mm, 10 mm, 5 mm, and 1 mm. This 57 enabled for a first physical classification 58 of the components, hence reducing the 59 60 diversity of the initial streams and facilitating the subsequent steps of the 61 process. ECs larger than 40 mm were 62 63 mainly connectors, while those smaller 64 than 1 mm were typically tiny multilayer ceramic capacitors (MLCCs), resistors or 65 66 broken parts or solder particles from the disassembly process. This smallest 67 fraction represented less than 1 wt% of the 68 total components (Figure 2). In addition, 69 the MEXRT detector has a resolution of 70 0.8 mm, making it difficult to detect and 71 analyze such small components (<1 mm) 72 with the current geometry. A possible 73 solution to tackle this issue is to use 74 magnification by using a fan-shaped X-ray 75 76 beam and increasing the distance between 77 the sample and the detector. After manually dismantling twelve different 78 WPCBs and counting the different ECs, 79 the average weight of a component was 80 found to be 24.67 g, 9.12 g, 0.83 g, 0.12 g, 81 0.006 g for the ECs with a size 82 above 40 mm, 40 to 10 mm, 10 to 5 mm, 5 83 to 1 mm and less than 1 mm respectively. 84



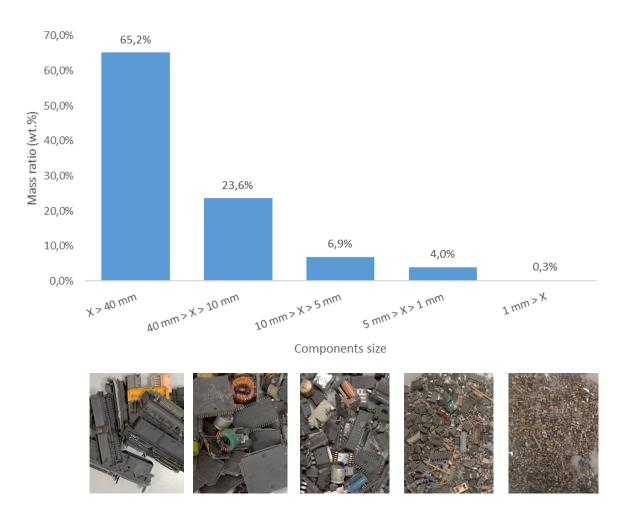


Figure 2: Mass distribution of the components in different size sub-fractions and their corresponding pictures

Given these limitations, the subsequent 1 2 sorting steps performed in this study focused on the three fractions of ECs with 3 sizes comprised between 1 and 40 mm. 4 Overall, the results of the sieving step 5 showed that the system was able to 6 efficiently and accurately separate the ECs 7 into these different size categories, with 8 35 wt.% of the components falling within 9 10 the target size range for further analysis. 11 Fourteen different types of ECs with chemical compositions of interest were 12 13 identified as the most common, of which, 14 between five to nine of them could be 15 found in the different size fractions, 16 therefore already significantly reducing the heterogeneity of the ECs for the 17 subsequent sorting steps. The distribution 18 ECs by category 19 of the can be

20 approximated by the average weight of an 21 EC for each category. In conclusion, the sieving step of our sorting process is an 22 23 effective method and useful step for separating electronic components into 24 different size categories. It allows for a 25 more targeted and efficient subsequent 26 27 sorting process, as the majority of the 28 components fall within the size range that can be effectively classified. The quality 29 of the sieving step is critical to the success 30 of the subsequent sorting steps and to 31 improve the overall performance of the 32 sorting process. 33

#### 34 **3.2. Process validation**

For the next sorting steps, we first
evaluated the performance of a sorting
approach consisting of an initial optical
sorting followed by MEXRT imaging. In
order to validate such a process and
evaluate its performance, a model sample
made of a controlled mixture of ECs was
prepared for each size sub-fraction.

a)

Fourteen EC types were identified (Table 43 SI 1) as the most common found in PCBs, 44 45 which furthermore contained targeted rarely recycled critical elements such as 46 Nd, Ta, Nb, Pd in capacitors, gold in 47 48 CPUs/connectors or aluminum in electrolytic aluminum capacitors. IC chips 49 and connectors were treated together due 50 to their similar visual appearance and 51 elemental composition in the optical 52 53 classification step and sorted together.

Stacking of 2D images One pixel = 1 spectrum x 128 7000 6000 Counts (a.u.) 5000 4000 3000 2000 x 128 1000 0 20 70 120 Energy (keV) b) Counts (a.u.) Count 3000 S 1e6 20000 (a.u.) 10000 120 140 2.0 Tr an 0.20 1.5 edge (Ba) = Κ sm 0.15 0.10 37.4 keV itt 0.05 1.0 an 0.00 100 120 140 160 20 ce 0.5 Tr 0.20 Kedge (Nd) = an 0.15 0.0 43.7 keV 0.10 sm 1 cm 0.05 itt 120 140 100 an Energy (keV) ce

The first sorting step based on optical 1 sorting showed a global accuracy of 2 97.4 % for a total of 8 categories (Table SI 3 4 2, Table SI 3 and Table SI 4). The 5 accuracy of the sorting process was evaluated by monitoring the number of 6 7 ECs in the final bins, taking into account 8 both the optical classification and the 9 mechanical diversion of the different ECs 10 into their respective sorting bins. With a minimum accuracy of 92.5 % for CPUs. 11 This is to be compared to the current state 12 13 of the art in optical sorting, which achieved a maximum of six EC categories 14 with 98 % accuracy on their own training 15 datasets, which could only give access to 16 the concentrated 17 most chemical elements(Lefkaditis and Tsirigotis 2010; 18 Lu et al. 2022). By adding the MEXRT 19 sorting step, which targets the k-edge of 20 elements present in ECs (Figure 3), the 21 system was able to separate optically 22 23 similar components with different elemental compositions. The addition of 24

the MEXRT step gave a total sorting 25 accuracy of the process to 96.9 %. By 26 sorting the components using MEXRT 27 28 and extracting the k-edge of known 29 elements from the HSI (Figure 3). 30 different categories of ceramic capacitors 31 can be separated based on their elemental composition. This is the case for single 32 layer capacitors (SLCC), which can be 33 34 divided into 4 categories: barium-based 35 SLCC, non-barium-based SLCC, neodymium-rich barium-based SLCC and 36 37 tantalum capacitors (Figure 4). The same applies to MLCCs, which can be divided 38 into two main categories: barium-based 39 40 MLCCs and non-barium-based MLCCs. By using the transmittance of the ECs. 41 XRT spectroscopy can easily separate 42 relays from metallized polymer capacitors 43 (MPCs) with 100 % precision which could 44 not be done based solely on the ECs' 45 features using optical sorting (Table SI 5, 46

47 Table SI 6 and Table SI 7).

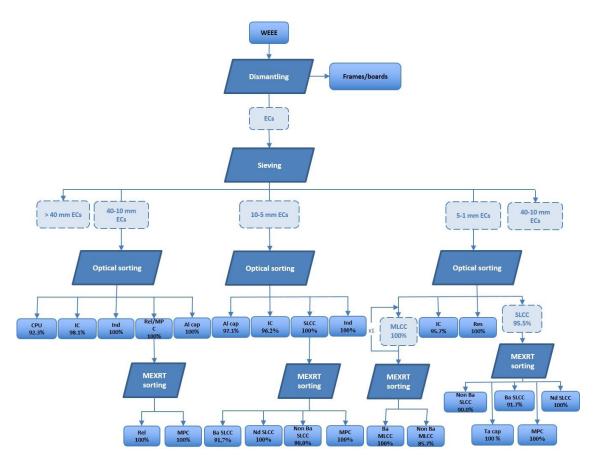


Figure 4: Flowchart of the sorting process for ECs

The optical sorting process can reach a 1 speed of 0.014 s/EC using a camera at a 2 speed of 75 frames per second. The 3 4 MEXRT acquisition speed used was 2 ms per line acquired and 50 lines of 0.8 mm 5 were acquired for a total acquisition of 40 6 7 mm, resulting in a maximum sorting speed of 0.1 s/EC. Considering the data 8 processing of the different images, with 9 the PC configuration used in this study, 10 the sorting speed for optical sorting was 11 12 reduced to 0.017 s/EC and 0.15 s/EC for 13 the MEXRT sorting (approximately 14 59 ECs/s and 7 ECs/s respectively). The image pre-processing method efficiently 15 defines the region of interest using only a 16 minimum transmission finding, allowing 17 accurate EC localization even for low-18 19 density materials, the only issue being the detection of multiple ECs on the same 20 image, which is outside the scope of this 21 22 study. To ensure that consecutive 23 components are not too close to each other during the MEXRT acquisition, a safety 24 25 distance corresponding to at least one

acquisition (100 ms) is left between the 26 components. This resulted in a maximum 27 28 speed of 0.25 s/EC for the MEXRT 29 sorting process. The average weight of an 30 EC was found to be 0.65 g, giving a 31 maximum average sorting speed of 9.36 kg/h. In real conditions, the sorting 32 speed was lowered to 0.5 s/EC (2 ECs/s) 33 34 to better control of the feeding system and 35 avoid stacking of components during MEXRT sorting process. As a result, the 36 37 MEXRT process being significantly 38 slower than the optical sorting, it was reserved for only a sub-fraction of the 39 total ECs for which an elemental based 40 sorting was required. Although the overall 41 42 accuracy may be slightly lower than the pure optical method, MEXRT provides 43 44 the added advantage of being able to differentiate similar-looking ECs with 45 different compositions without impacting 46 47 the high accuracy if the optical sorting.

48 In conclusion, our sorting process using 49 both optical sorting and X-ray transmission techniques is a highly
effective method for accurately classifying
and separating electronic components. The
system has a high sorting accuracy and
speed, making it a valuable tool for
electronic component recycling.

## 56 3.3. Real samples sorting 57 process/Scalability study

58 The sorting process results were then scaled-up to a larger batch of waste by 59 applying the same method to a mixture of 60 61 9056.5 g of printed circuit boards from engine control units. After disassembly, 62 5409.4 g of ECs were recovered and 63 sieved for further sorting (Figure 5). Each 64 65 sorted fraction was shredded into homogeneous powder and then analysed 66 by XRF for a first qualitative analysis of 67 68 the composition, followed by а quantitative analysis of the target elements 69

70 by ICP-OES. The analysis of the sorted 71 ECs in the final streams showed a clear 72 enrichment of the target elements such as gold, Al, Pd, Mn and Cu. The enrichment 73 of gold was found to be up to 0.14 wt.% in 74 75 CPUs while it was below the detection 76 limit in the reference mixture (below 77 0.1 ppm). This represents an increase of four orders of magnitude in concentration. 78 79 It was also found to be up to 53.95 wt.% 80 for Al in electrolytic capacitors with a size between 40 and 10 mm. The segregation 81 of the ECs does not only enrich the final 82 streams in certain elements but also 83 reduces the heterogeneity of some classes. 84 This is the case of the inductors sorting 85 bin (5-10 mm), which is composed of 86 45.6 wt.% of iron, 38.5 wt.% of copper 87 88 and 20.82 wt.% of tin with only traces amounts of other metallic elements (Ni, 89 90 Zn, Al).

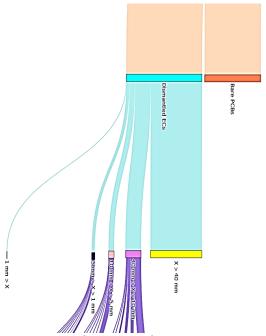


Figure 5: Mass flow diagram for the up-scaled process

From a global point of view of the overall
 process, a convenient way to represent the
 simplification that it allows for is to
 compare the entropies of all the sorting
 bins obtained, which can be estimated
 using the Boltzmann equation:

7  $S = -k_B \sum_{i=1}^{n} x_i \log(x_i)$  (2)

8 Where  $k_B$  the Boltzmann constant and  $x_i$ 9 the mass ratio of the elements present in 10 the ECs measured by ICP-OES. The 11 results of this calculation are shown in 12 Figure 6-a. In the latter, one can compare

the entropy of each sorting bin with the 13 entropy of the reference mixture of ECs 14 plus the composition of a bare WPCB 15 16 (Smalcerz et al. 2023). Any fraction with an entropy lower than the latter, can be 17 said to have a simpler composition. This 18 19 comparison shows that all fractions have a significantly lower entropy than the one of 20 the ground WPCB. This reduction of the 21 22 entropy of the phase is particularly visible 23 for inductors and relays, which are mainly 24 composed of iron and copper. In the cases with the highest entropy, such as the 25 category labelled "5-1 mm other SLCC", 26 this level of entropy comes from the 27 sorting induced enrichment in Mn, Zn and 28 Bi, that are now no longer present in 29 therefore 30 traces. and significantly 31 contribute to the entropy. Same goes for the MPCs sorting bin, which after sorting 32

33 contained up to 47.04 wt.% of tin (Table34 SI 8).

35 Overall, looking at Figure 6-b and comparing the number of major elements 36 37 present either in the initial WPCB or in 38 each of the sorting bins, it can be seen that 39 all the sorted categories have much 40 simpler composition, especially when 41 only considering the number of major 42 elements in their composition (all have 43 less than six elements with concentration 44 over 1 wt.%, to be compared to thirteen in the original WPCB). This demonstrates 45 that the process is not only effective in 46 classifying 47 separating and electronic 48 components, but it also easily and effectively concentrates valuable metals, 49 facilitating 50 thus subsequent chemical 51 separation, a key requirement for making viable. 52 their recycling economically

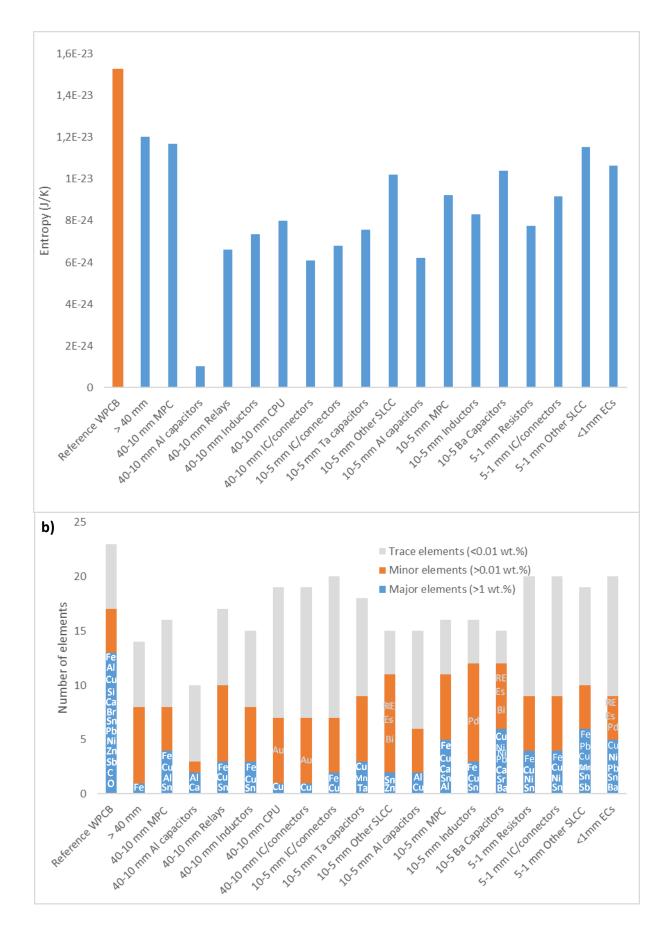


Figure 6: a) entropy of metals and b) number of elements in the different sorted categories. All chemical elements present in the major elements fraction (blue blocks) are listed in white. Significant elements that where either at trace level or below

# 3.4. Throughput and process industrial implementation

This sorting process presents a great opportunity for the development of economically processes. viable The viability of this process was previously reported by (Xia et al. 2022) in the study of neodymium from Nd-based SLCC. Although this study only considered the recovery of Nd-based SLCC, the process was already showing a payback time of less than 5 years. While other recycling processes targeting specific ECs have been developed (Chen et al. 2021; Niu and Xu 2018; Wang and Xu 2017), they often lacked a mean of obtaining concentrated streams of the targeted ECs. This sorting process addresses this issue by effectively classifying and separating ECs, allowing for the recovery of valuable materials in a concentrated form. The ability to recover these materials in a concentrated form significantly improves the economic viability of the recycling process. This technology has the potential to revolutionise the field of electronic waste management and recycling by providing an efficient and cost-effective method of recovering valuable materials from components. discarded electronic However, the limitations of the process lie in the in-line treatment of the ECs, and the sorting speed will be improved in further development of the prototype by using a sorting. We developed parallel a demonstrator at a laboratory scale to validate the feasibility of advanced sorting and its compatibility with industrial plants by testing the sorting capability as a function of belt speed, although a larger, multi-lane industrial sorting tool would use similar conveyor speed. The cost of the prototype was broken down as follows: 50,000 USD for the X-ray system (generator, detector, shielding), 2,500 USD for the control and data acquisition

systems, and 12,500 USD for the mechanical structure, conveyor belt and sorting system. It is important to note that the costs of a laboratory-scale prototype may not be directly transferable to a larger, multi-lane industrial sorting tool. On a 1 meter large conveyor belt, up to 50 ECs can be treated in parallel. Thus, the sorting speed of the prototype would be multiplied by fifty allowing the sorting of up to 500 kg/h. Considering that a country such as Singapore produces an average of 60000 tonnes of e-waste per year, resulting in approximately 1620 tonnes of ECs (PCBs represent 6 wt.% of the global e-waste (Pinho, Ferreira, and Almeida 2018)), 3240 machine hours would be required. equivalent to 125 days. Hence, this process shows promising results for industrialisation.

#### 4. Conclusion

In conclusion. our instrumental developments and integration reported here presents a highly effective sorting process for electronic components (ECs) that combines physical, optical and MEXRTbased tools and process steps. The overall process is capable of concentrating valuable chemical elements in the final streams with, for example, an enrichment of up to 10,000 times its initial content in the WPCBs in the case of gold, including metals that are currently wasted and not recovered in industrial recycling settings. Achieving such levels of enrichment, while combining high accuracy and low operating costs, opens up new opportunities in the recycling industry, especially regarding these elements that are lost in WEEE recycling processes. Indeed, such elementally enriched fractions can either be traded purified or by hydrometallurgy. In the latter case, one still has to face the issue of the variability of the composition of the sorting bags, which can be solved thanks to state of the art microfluidics based rapid process development tools (Maurice et al. 2022; Olivier et al. 2022, 2023).

# Credit authorship contribution statement

М. Charpentier: Nicolas Conceptualization, Methodology, Software, Investigation, Data curation & Original draft Interpretation, Writingpreparation. Maurice: Ange A. Conceptualization, Investigation, Reviewing and Editing. **Dong** Xia: Investigation, curation Data and Interpretation, Writing- Reviewing and Editing. Wen-Jie Li: Investigation. Chang-Sian Chua: Investigation. Andrea **Brambilla**: Conceptualization. Jean-Christophe P. Gabriel: Conceptualization, Methodology, Data Interpretation, Writingand Editing, Reviewing Supervision, Funding acquisition. Project administration.

#### **Declaration of Competing Interest**

NMC and JCG wish to declare that MEXRT detector ME100 used in the work described here is commercialised by Detection Technology under patent licensing from CEA. The authors declare no other competing interests.

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