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The effect of crusher type on printed circuit board assemblies' liberation and dust generation from waste mobile phones

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ABSTRACT

Waste electrical and electric equipment entering our recycling system poses various challenges in different stages of the whole treatment chain. Losses of valuable metals in the crushing of electrical devices through dusts and fines have been identified as one such challenge. Crushing is nevertheless crucial in order to liberate metals from each other for efficient separation in the sequential mechanical unit processes. This study investigated the relation of crushing mechanism on Printed Circuit Assemblies' (PCAs) liberation and fines generation in the size reduction of waste mobile phones using two crusher types. The results revealed that a fast-rotating hammer mill produced better liberated PCAs with an overall PCA grade, presenting the purity of PCA fraction of 77% compared to a slow-rotating cutting mill with 58% PCA grade. However, the hammer mill produced over two times the amount of fines compared to the cutting mill. The fines fraction mainly comprised silicon and base metals but also noble metals and harmful elements which need to be taken into account when further treatment is considered. Even though the same elements could be found in the fines from both crushers, differences in the concentrations were observed. In the cutting mill, higher concentrations of ductile materials such as gold and copper were observed in the fines fraction with particle size below 4 mm compared to the hammer mill.

1. Introduction

Electronics has been one of the key industries in the promotion of wellbeing in today's society. The working environment has long been reliant upon electrical devices, which are also being increasingly used in leisure time. Consumer demand for versatile devices together with the trend in decreasing prices has led to a drastic reduction in the lifespan of electronic devices (Baker and Schuit, 2017; Tanskanen, 2013). This trend in turn has led to a situation where the generation of Waste Electrical and Electronic Equipment (WEEE) has increased substantially, resulting in a global WEEE generation of 53.6 million tonnes in 2019. It has been estimated that by 2030 the global WEEE generation will reach 74.7 million tonnes (Forti et al., 2020).

Electronics consumption and ownership are linked with the income level of the country. It has been reported that, as the level increases the number of devices owned per capita increases as well. For example, in a high-income country with an average purchasing power of over USD 51 000 per capita, a person owns on average 1.6 laptops, 0.7 fridges, 1.4 mobile phones and 16 lamps. By contrast, in a low-income country with an average purchasing power of slightly over USD 1 000 per capita, the ownership figures are 0.1 laptops, 0.02 fridges, 0.6 mobile phones and 4 lamps. (Forti et al., 2020) The gap between the low- and high-income countries is substantial, and when the populations in low-income countries adopt similar lifestyles to those which the high-income countries currently have, the global metal demand would be 3–9 times greater than the current metal consumption (UNEP, 2013). This projection creates pressure on sustainable raw materials' production, which calls for new circular economy business models and technology solutions, including also recycling. As a major consumer, the electrical and electronics industry demands solutions especially for copper, precious and scarce metals (Bachér et al. 2020, ICSG, 2020, Oguchi et al., 2012; Glöser et al., 2013, U.S. Geological Survey, 2021).

The vast range of different types of electrical devices requires different metals to function. These devices may contain up to 69 different elements from the periodic table (Forti et al., 2020). In particular, so-called high-tech devices such as mobile phones require various metals. In addition, the grade of the materials used in the applications needs to be high (UNEP, 2009). Consequently, high-tech applications provide a desirable raw materials source for the recycling industry. As an example, a mobile phone comprises metals with a share

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of between 22 and 35 w%, plastics between 40 and 60 w% and ceramics around 10 w% depending on the phone model and manufacturing year (Bachér et al., 2015; Chancerel and Rotter, 2009; Huisman, 2004; Ueberschaar et al. 2017a; UNEP, 2009, 2013). The metal content comprises mostly copper, aluminium and iron followed by other industry metals such as nickel, tin and zinc, as well as precious metals, but it also includes technical and scarce metals such as cobalt, gallium, neodymium and tantalum (Ueberschaar et al. 2017a).

Legislation that steers the management and handling of the WEEE varies depending on the continent. In Europe, the handling of WEEE and waste mobile phones is governed through the WEEE Directive (Directive 2002/96/EC), which states that equipment entering the end-of-life stage should primarily be reused as a whole or on the component level. However, if reuse is not possible, the collected WEEE must be recycled. The collected WEEE is commonly treated with a series of mechanical unit processes such as crushing, sieving, magnetic, eddy current, density and sensor separation. In addition, manual dismantling can be applied, especially if hazardous components must be removed prior to processing. The separated metal concentrates are sent to various metallurgical recovery and refining processes to produce metal goods. The plastics can be partly recycled if the quality is good and no harmful substances are present, whereas the low-quality plastics go to energy recovery or disposal (Buekens and Yang, 2014; Maisel et al. 2020). In each stage of the treatment chain, losses of materials and metals occur due to, for example, the physical and chemical boundary conditions in the unit processes. Consequently, in order to avoid metal losses in waste mobile phone recycling, the mechanical treatment step is most often bypassed. Within this approach, batteries are removed manually, if possible, and then the entire phone is fed into a sophisticated integrated copper smelter where the plastics are used as a fuel and as a reducing agent (EPA, 2009; UNEP, 2013). However, since 2018 the European Union has targeted the recovery rate for group 6 (small IT and telecommunication equipment) to be 75% and the recycling rate 55% (Directive 2012/19/EU). This leads to a situation where plastics need to be recycled and separated, which in turn requires a mechanical pre-treatment or manual dismantling step, as the targets are defined on mass-based calculations.

The mechanical treatment unit processes of the WEEE recycling chain aim to enrich selected metals for subsequent metallurgical treatment. Several authors have reported the size reduction process as an origin for metal losses when targeting the optimal enrichment (Bachér et al., 2015; Bachér and Kaartinen, 2017; Chancerel et al., 2009; Marra et al. 2018a; van Schaik and Reuter, 2014; Ueberschaar et al. 2017b; UNEP, 2013). However, for effective mechanical treatment, size reduction of the feed is required as the particles are separated based on their physical properties such as magnetic susceptibility or density. During the crushing procedure, the materials are disengaged from each other, and various liberation stages occur with the remaining particles. Liberation describes how well the materials are disconnected from each other (Castro et al., 2005; Gay, 2004; Heiskanen, 2014; van Schaik et al., 2004). A poorly liberated particle is composed of several materials which all affect its physical properties. This is likely to weaken the separation based on the physical property (i.e. density, colour, magnetism) (UNEP, 2013). Alternatively, a poorly liberated particle might be separated into incorrect fraction, as it may contain materials with properties that have been used to separate the particles prior to separation into the desired fraction. This might lead to the loss of the material if further treatment of the fraction does not recover it. For example, a PCA may end up in the magnetic fraction, if the magnetic parts such as screws have not been disengaged during the crushing stage. As steel refining cannot recover copper, tin and precious metals, and it is the subsequent process for the magnetic fraction, it would mean that a PCA and its metals would be lost. Furthermore, some of these metals are considered as an impurity in steel production (Björkman and Samuelsson, 2014).

The liberation of materials can be generally improved by decreasing the particle size through size reduction (Castro et al., 2005; Menad et al. 2013; Quan et al., 2012; Zhang and Forsberg, 1997). However, the mechanical treatment processes may have particle size limitations for separation efficiency. As an example, in eddy current separation, the efficiency starts to decrease when the feed particle size is below 5 mm (Cui and Forssberg, 2003). This leads to optimization between the particle size and liberation of materials. Even though the optimal particle size with good liberation of materials is reached, some small particles and dusts are always generated in size reduction. These fines have raised concern as a potential source for losses of valuable metals (Bachér et al., 2015; Bachér and Kaartinen, 2017; Chancerel et al., 2009; van Schaik and Reuter, 2014; UNEP, 2013). This relation between the losses through fines and the liberation of desired components in mobile phone recycling becomes relevant when mechanical treatment is carried out to reach recycling targets.

During recent decades, studies on the size reduction of WEEE have been widely conducted. Various mills of different scales have been studied, such as the large-scale vertical hammer mill on WEEE by Eichert et al. or the smaller scale impact mill on Printed Circuit Board (PCB) to separate copper by Hou et al. (Eichert et al., 2008; Hou et al., 2013). Koyanaka et al. studied further the liberation and breakage of PCB particles in relation to the origin device in an impact mill. They found that PCBs from different devices behave differently in a size reduction process (Koyanaka et al., 2000, 2006). In addition, Quan et al. studied a two-step crushing process of PCB to liberate metals from non-metals. They observed that metals are liberated at a particle size of<0.59 mm, whereas non-metals tend to aggregate in a smaller than 0.15 mm size fraction, which can cause difficulties in further treatment. (Quan et al., 2012) A recent study on the effect of feeding rate of PCB and milling time on the metals recovery and enrichment in a hammer mill showed that Cu and Al recovery in coarse fraction was related to their selective agglomeration, while in the fines fraction the recovery was related to the selective size reduction (Otsuki et al. 2019). In addition, Otsuki et al. studied different analysis methods to investigate the liberation of copper and other materials after milling of PCB with a hammer mill. The study revealed that for a particle size fraction of 0.125–0.320 mm, optical microscope and tomography analysis produced a liberation degree of 89% and 81% for metals respectively, whereas the micro-XRF resulted in a 54% liberation degree. (Otsuki et al. 2020) Besides the research focusing on size reduction of PCB, Zhang and Forssberg studied the liberation of different components and elements in the size reduction of personal computers in a hammer mill. These studies revealed that metallic particles achieved almost complete liberation at below 2 mm, whereas in coarser particles copper gained poorer liberation degrees than aluminium and ferrous metal (Zhang and Forsberg, 1997; Zhang and Forssberg, 1999). Similar results have been presented by Menad et al. who investigated the liberation as well as shapes of particles from a recycling plant treating WEEE with a hammer mill as a size reduction process (Menad et al., 2013).

Further, an investigation into breaking mechanisms in different crushers with different feed material characteristics has gained interest. Sander et al. studied the behaviour of scrap metals in a high-speed swing-hammer type shredder. They identified four stages that occur during the shredding. In the first stage, single fragments are torn off from the feed, whereas in the second stage intense deformation of the comparatively large platy fragments occurs leading to the formation of flaws. In the third stage, smaller fragments smash against the wall of the housing, causing deformation and further formation of flaws. In the last stage, the fragments compact until they become spherical and the formation of fines takes place. (Sander and Schubert, 2003; Sander et al. 2004) In turn, Woldt et al. studied the breaking mechanisms in low-speed rotary shear with aluminium (AlMg3) and polypropylene samples. The study revealed that the comminution in rotary shear results from shearing, cutting and tearing stresses, and it is composed of deformation and friction processes. (Woldt et al. 2004) A recent study by Heibeck et al. further researched the material liberation of multi-material structures, namely steel and composites, after shredding with a rotary shear. The study tracked changes in joint characteristics, material composition and particle size over a course of two-sequence shredding, which revealed that material liberation is dependent on many design and shredding parameters. The work was complemented with numerical simulation modelling to provide information for designers on how design configurations affect the liberation behaviour. (Heibeck et al. 2021).

In a broader perspective, to improve recycling Maisel et al. highlight the importance of crushing of WEEE for the efficient separation of plastics, the communication between different actors on particle size standards for intermediate products, as well as steering fines to appropriate actors (Maisel et al. 2020). In summary, the liberation and comminution behaviour in a size reduction process of WEEE differs and involves complex characteristics.

As for the fines and dusts from the mechanical treatment of WEEE, research has for the most part focused on occupational health matters (Deng et al., 2014; Tesar et al., 2014; Xiang et al., 2007; Zheng et al., 2015). In order to identify possible losses of valuable metals through fines/dusts, some studies on mineralogical characterization, dust generation and metallurgical treatment have been published (Bacher and Kaartinen, 2017; Borowski et al. 2018; Marra et al. 2018b; Wang et al. 2015). However, the linkage of material liberation and fines generation in relation to the crushing mechanism of WEEE has not yet been addressed.

In this study, the relation of crushing mechanism on PCA liberation and fines generation in the size reduction of waste mobile phones was investigated with two crusher types, a high-speed hammer mill and a slow-rotating cutting mill. Waste mobile phones were selected for the study due to their high precious and industry metal content in order to detect clear variation in the fines concentrations. In addition, the aim was to reveal which metals and materials are lost if they end up in the crusher instead of feeding them directly into the pyrometallurgical processes, as currently is the case.

2. Material and methods

2.1. Mobile phone sample and dismantling

For this study, two identical spent mobile phone samples were collected. The samples of 9.8 kg each were composed of 21 different phone models from the time period between 2000 and 2012. In total, 124 phones per sample were identified which mainly dated from 2004, 2005, 2007 and 2009 (Fig. 1).

The phones in the samples were classified into three different categories based on their characteristics: basic phone, communicators and smart phones. Basic phones were typically small and light, composed of plastic casing and with a small screen. These phones were mainly used for making calls and sending text messages. Communicators were larger and more sophisticated than basic phones, which could be used for websurfing and taking photographs. However, these phones did not have a touch-screen. Often these phone models had two sides, the traditional basic phone user interface on one side and then a more extensive interface with large keyboard and screen when the phone was opened as a book. The final category was composed of smart phones with large touch-screens, several cameras and high-end processing capacity.

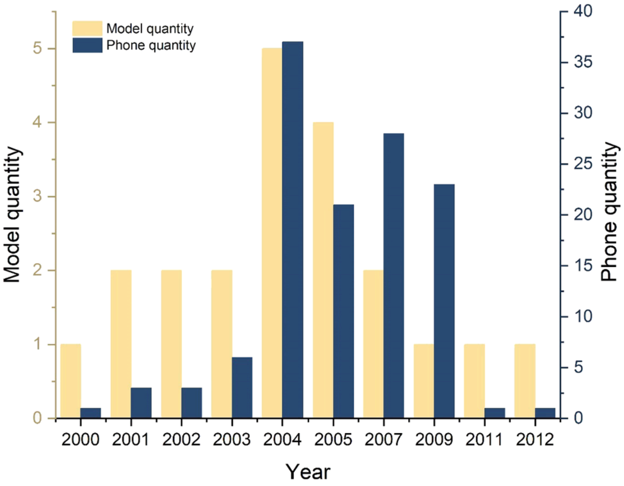

Fig. 1. Phone quantity and model distribution in the two identical samples.

In order to identify the component composition and to investigate the effect of different crushing mechanisms on the liberation of printed circuit assemblies (PCAs), one of each phone model was sent for dismantling analysis. In this analysis, the phone was hand-dismantled into seven different component groups: plastic casing, frame, PCA, laminated printed circuit board, screen, screw, other. Each of the components were weighed to study the weight-based component composition.

Besides the two identical samples for the size reduction experiments, a smaller 2.3 kg sample of mobile phones was gathered for a pre-trial with the hammer mill to assess agitation speed of the rotor for the phone sample. This sample was divided into two 1.15 kg sub-samples, both comprising 15 phones. These phones were not introduced to similar extensive characterization with hand-dismantling as the primary mobile phone samples since the liberation efficiency was only determined from the crushed samples.

2.2. Size reduction experiments

Prior to size reduction, all the mobile phones were examined to check if any batteries were still installed in the phones, and if so they were removed. The size reduction was carried out with a Rivakka hammer mill and a Weima WL2 labor single-shaft cutting mill. The Rivakka hammer mill is composed of a rotor with four blades on which three hammers per blade are mounted, adding up to 12 hammers in total. The diameter of the rotor is 53 cm. The Weima cutting mill is composed of a 37 cm wide rotor with a diameter of 18 cm including 22 knives. The output size opening was 100 mm for the Rivakka hammer mill, which was used without an internal screen/grate, but for the Weima cutting mill, the construction design is such that the output opening size cannot be determined. However, its internal screen/grate was not mounted. The opening size was selected in order to minimize damage to the PCA in the mobile phones whilst liberating them from the other main parts.

The Rivakka hammer mill operates in continuous mode with an adjustable agitation speed of the rotor with attached small movable hammers from 0 to 3 000 rpm. The processing capacity for mobile phone type material was a few kg/h. The feeding was carried out manually one mobile phone at a time. The crushed sample, including the dust fraction, was gathered in a filter bag. In order to find a suitable agitation speed for the mobile phone sample, a pre-trial with two agitation speeds was carried out. In this trial, a smaller sample amount was crushed at 1 500 and 3 000 rpm. The crushed samples were analyzed similar to the main sample but without the dust/fines chemical analysis, as the main purpose for this pre-trial was to find a suitable agitation speed to maximize liberation of the PCA particles.

The Weima cutting mill is a slow-rotating single-shaft mill which operates in continuous mode with a fixed agitation speed of 70 rpm. The processing capacity for mobile phone type material was roughly 10 kg/h. The feeding was carried out manually, whereby several mobile phones were fed simultaneously in batches. The crushed sample, including the dust fraction, was gathered in a container.

After the size reduction, the crushed samples were sieved into particle size fractions with the following sieve sizes: 31.5 mm, 20 mm, 14 mm, 10 mm, 8.0 mm, 5.6 mm, 4.0 mm, 2.0 mm and 1.0 mm.

2.3. Liberation analysis of PCA

The liberation characterization (Fig. 2) was performed for the fractions from all crushing experiments above 4.0 mm. The liberation characterization of PCA particles was carried out by hand dismantling and demolishing every PCA particle. In this study, PCA refers to printed circuit boards that still have surface components attached. Within this procedure, all the PCA particles in each size fraction were dismantled with pliers and screwdrivers to separate the PCA from the other main mobile phone parts such as casing and screens. After dismantling, both the separated parts and liberated PCA were weighed, and the mass-based liberation degree of the PCA was calculated with Equation (1) below. This value describes how well the PCA is disengaged from the other parts. Based on the result from Equation (1), each PCA particle was distributed into the following liberation classes: <15%, 15–25%, 25–35%, 35–45%, 45–55%, 55–65%, 65–75%, 75–85%, 85–95, 95–100% and 100%. Based on the liberation data and mass distribution of PCA particles, the PCA recovery and grade were calculated with Equation (2) and (3) in Fig. 2, where *l* denotes the lower liberation class value with the exemption of class < 15% where 5% liberation value has been used and *m* the mass of the entire PCA liberation class in Fig. 2. The number and mass of analyzed particles, for the liberation analysis, is presented in Table 1. For the pre-trial experiments, the amounts of PCA particles analyzed were lower.

$$DegreeofliberationforPCAparticle = \frac{m_{PCA}}{m_{PCA} + m_{rest}} \tag{1}$$

where m_{PCA} denotes the mass of detached PCA in the particle and m_{rest} the mass of other parts detached in the same particle.

2.4. Chemical analysis of dust and fines fraction

The dust (<1.0 mm) and fines (1–2 and 2–4 mm) fractions from both crushing experiments were characterized to provide information on the elemental composition of the dust and how the crusher type affects it. Prior to the elementary analysis, roughly 20 g subsample of each size fraction was divided with a rotary sampler and milled with a Fritsch Pulversette 19 to under 1 mm. An assay sample of roughly 0.2 g was taken from the subsample and dissolved including microwave-assisted digestion with hydrofluoric (HF), nitric (HNO3) and hydrochloric (HCl) acid mixture for subsequent determination of elements in the sample. The eluates were analyzed for Ag, Al, Cu, Fe, Pb, Sb, Si, Sn and Zn by Inductively Coupled Plasma Mass Spectrometry (ICP-MS). After the dissolution and analysis, the undissolved leaching residue was weighed. Depending on the sample, the residue presented between 11.6 and 22.1 w% of the assay sample. In addition, precious metals (Au, Pd and Pt) were determined with the Pb-Fire Assay method combined with Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES) technology. For this analysis, between 3.81 and 10.04 g assay sample was used.

Table 1
Summary of the analyzed PCA particle amounts and masses in different experiments.

	Pre-trials with hammer mill		Actual size reduction experiment	
	1500 rpm	3 000	Hammer mill	Cutting mill
Number of PCA particles analyzed	155	311	1 661	1 266
Mass of PCA particles analyzed [g]	476.3	308.6	2 266	3 285

3. Results and discussion

3.1. Mobile phone composition

The dismantling analysis revealed that spent mobile phone samples were mostly composed of plastic casing, frames and PCA (Fig. 3). These three components contribute to over 81 w% of the phones. When considering the valuable precious metals (Au, Pd, Pt) and copper, which are located in the PCA, mobile phones represent a truly valuable product

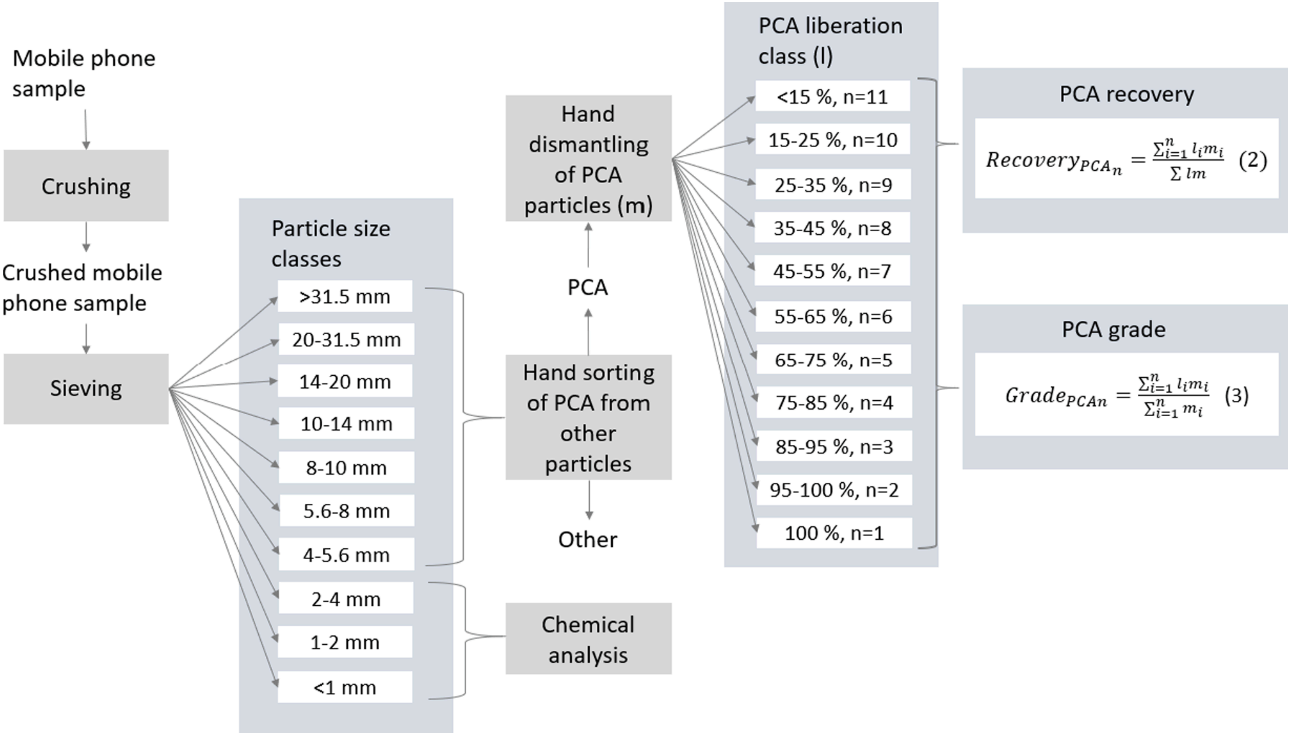

Fig. 2. An overview of the liberation characterization methodology.

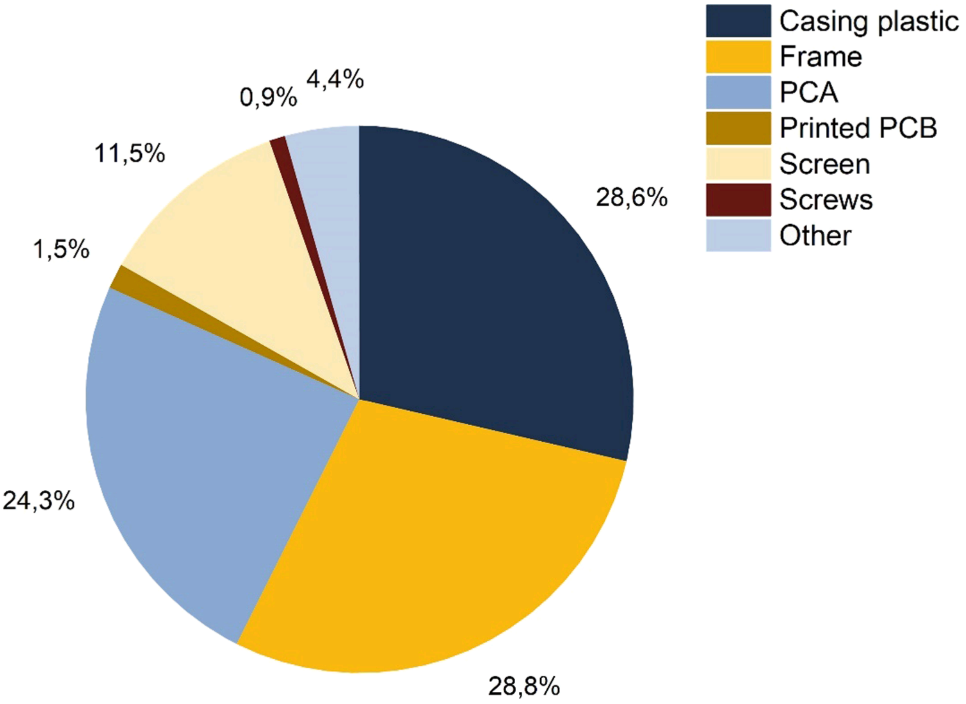

Fig. 3. Component composition of the entire spent mobile phone sample computed as average from all three phone categories namely regular phone, communicator and smart phone.

category with their high PCA share (Chanceler et al. 2009; Oguchi et al. 2011; UNEP 2013).

The component composition varies significantly depending on the mobile phone category (Fig. 4). One distinguishing characteristic is the share of screen in the phone. With the move towards smart phone technology, the share of screen increases dramatically. This is logical considering the mobile phone development with the shift towards touch-screen phones in which the screen acts partly as a casing. Another notable change is in the share of PCA in the phone. Higher shares can be detected for basic phone models. The reason for this originates from the design, where the PCA acts as a platform/frame for the phone. The other parts are constructed around the PCA. As for communicators and smart phones, the PCA is a separate component that is connected to other parts. Laminated/printed PCBs are nearly solely found in only communicators beneath the keyboard. This is due to individual design choices which appear only for products produced in a certain period. This is a

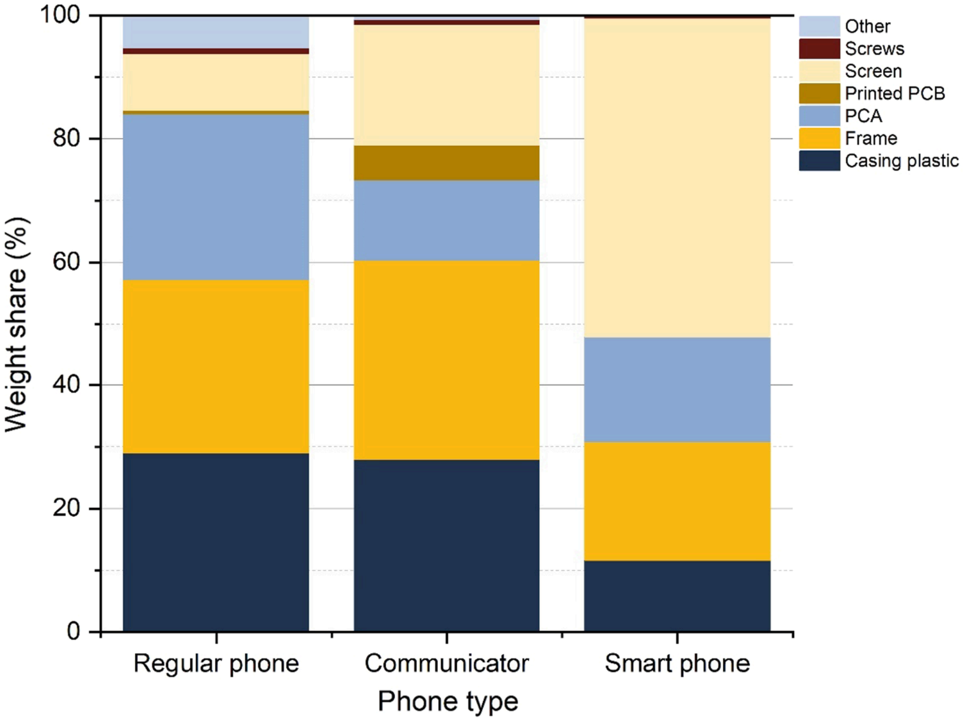

Fig. 4. Component composition in different phone categories.

good example of the dynamic nature of mobile phone waste and in larger context the Waste Electrical and Electronic Equipment (WEEE).

When looking at the change in share of key components in basic phones over the time period under examination, it can be noticed that while the share of PCA has decreased during the decade, the share of screens has increased (Fig. 5). An even greater shift towards screen-dominated spent mobile phone waste occurs when taking smart phones into consideration. As for casing and frames, no clear change in the shares can be detected. The dynamic nature of mobile phone waste, where metal-rich PCA shares are decreasing and silicon-rich screens are increasing, will not ease the recycling of valuable metals. In fact, with regard to the size reduction process, brittle screens will break more easily in the crushing and thus increase the share of fines and dust.

3.2. Pre-crushing test

The pre-crushing test with the hammer mill was to assess the optimal rotor agitation speed in terms of liberating the PCA components from other mobile phone parts. The sieving analysis of crushed samples revealed that the D50 value, representing the particle size where 50 % of the material is below this size, for high agitation (3 000 rpm) was 14.4 mm, whereas for slow agitation (1 500 rpm) the value was 33.7 mm. The clear increase in the D50 value already indicates a lower liberation of components in the slow agitation mode. As for the share of dust (<1.0 mm), the high-speed agitation produced roughly 3% of material under 1 mm, whereas the slow agitation produced roughly 1% of dust.

In more detail, the size distribution of PCA particles and other parts such as frames, casing and screens in Fig. 6 shows a clear difference between the agitation speeds. In general, PCA particles are larger than other particles, indicating a more tensile and resistant nature in terms of adsorbing the stresses in a collision between the particle and the hammer of mill. On the other hand, PCA components are often situated in the centre of mobile phones protected by the parts around them, which absorbs the main energy. When comparing the agitation speeds, it seems that in slow agitation the energy introduced to the system does not break the phones in a such manner that the PCAs undergo transversal breakage, which further leads to lower liberation. This is supported by the international literature in which it has been reported that, in order to generate intense deformation and transversal breakage, the energy demand influenced by the circumferential velocity of the impacting tool needs to be high enough (Sander et al. 2004).

The effect of the hammer mill's agitation speed on the PCA grade and recovery is presented in Fig. 7, which shows that high agitation provides enough energy to liberate PCA particles with a grade of above 86%. In slow agitation, the grade remains low, and only slightly above 30% of PCA particles are completely liberated compared to high agitation, which produced 77.6% of fully liberated PCA particles.

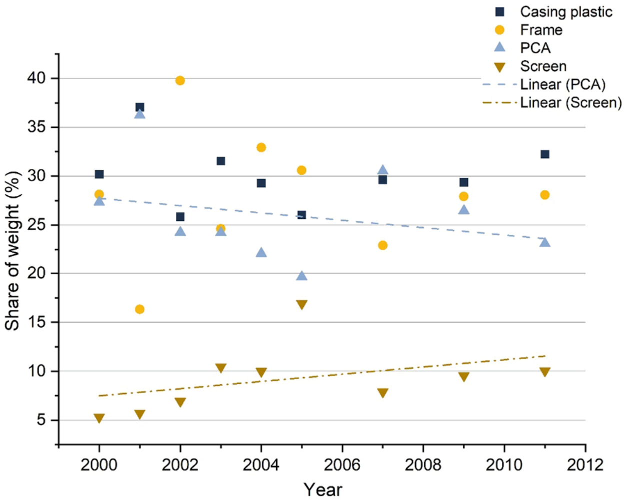

Fig. 5. Share of key components in basic phones (number of phones 18) and communicators (number of phones 2) used in this study during the time period under examination (excl. 2012 with smart phone).

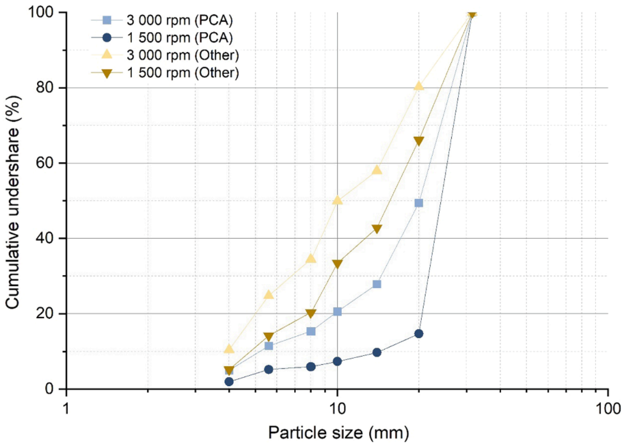

Fig. 6. Particle size distribution of PCA and other parts (incl. frames, casing, screens etc.) with hammer mill rotor agitation speeds of 1 500 and 3 000 rpm.

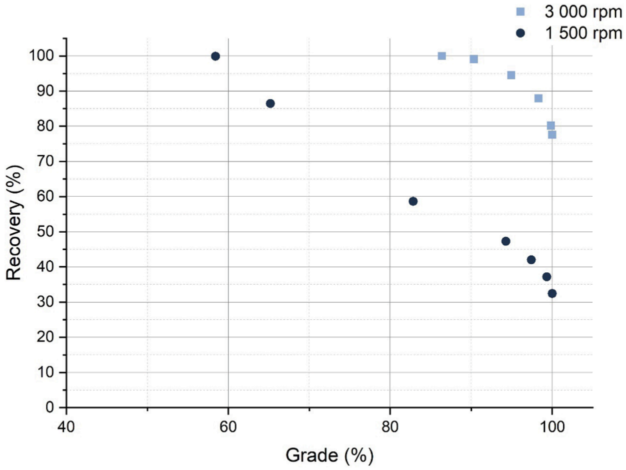

Fig. 7. The recovery and grade of PCA particles from both agitation speeds. The amount of PCA particles analyzed in the liberation characterization was 155 for the 1 500 rpm sample and 311 for the 3 000 rpm sample.

It was found that the higher agitation speed increased the liberation of PCA particles from other components. The overall PCA grade for 3 000 rpm agitation speed was 86.4%, whereas for the 1 500 rpm agitation speed it was 57.9%. Therefore, the agitation speed of 3 000 rpm was selected for the main crushing experiment.

3.3. Particle size comparison between hammer mill and cutting mill

The sieving analysis showed that the hammer mill produced finer material than the cutting mill (Fig. 8). After the cutting-mill crushing, nearly 45 w% of the material was>31.5 mm, while after hammer-mill crushing, only approximately 30 w% of particles had particle size above 31.5 mm. The D50 value for the hammer mill was 20.5 mm, whereas for the cutting mill the value was 28.6 mm. To some extent, the difference may originate from slightly different output opening sizes of the crushing chambers. In any case, the breaking stresses are not similar in the crushers. In the cutting mill, the stresses are channelled on to a smaller area at which time more precise breakage can occur. Usually, the cutting mill's breaking stresses are more controllable than in the hammer mill. In the hammer mill, the mobile phone sample shoots

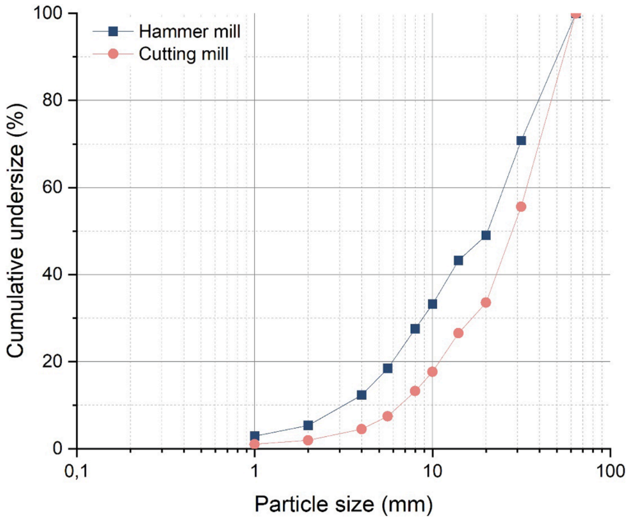

Fig. 8. Cumulative particle size distribution of crushed material from the hammer and cutting mills.

uncontrollably in the crushing chamber, while in the cutting mill the mobile phones set between the knife and anvil in a more controlled way. This may also be affected by the agitation speed of the rotor. As a result, a greater amount of material is broken down in the hammer mill when the stress is spread over a larger area. In fact, the hammer mill produced 2.9 w% of dust fraction (<1.0 mm), whereas the cutting mill produced 1.1 w%.

The size distribution in terms of PCA and all other components from both mills (Fig. 9) shows that the PCA particles remained larger than other components. The D50 value for the PCA from both mills was slightly above 22 mm. The size distribution curves of the PCA particles are rather similar for both mills, whereas for other components the hammer mill produced smaller sized material. Especially the brittle types of materials, which are used for example in screens, are presumably pulverized more easily in the hammer mill than in the cutting mill and generate more finer material for the other component fraction. From the size distribution point of view, the hammer mill can be considered slightly more selective than the cutting mill, as clearer difference between distribution of PCA and other particles exist. However, pure fractions are not produced in either case, as in all size fractions both PCA particles and other particles can be detected.

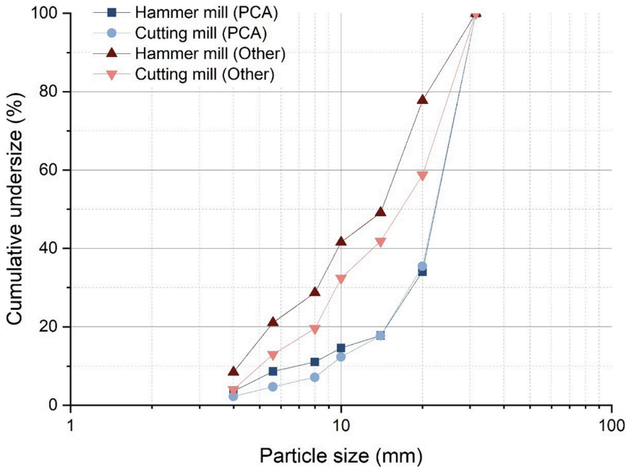

Fig. 9. The size distribution of PCAs and other components for the hammer and cutting mills.

3.4. Liberation of PCA

Fig. 10 shows the liberation distribution of the PCA from the hammer and cutting mills. The results clearly indicate that a greater amount of liberated PCA particles is generated with the hammer mill. This is in line with the sieving analysis (Fig. 8) presented in Section 3.3, as generally a smaller particle size has higher liberation (Wills and Napier-Munn, 2005). For the hammer mill, over 50 w% of PCA particles is completely liberated, while for the cutting mill only 31.3 w% of PCAs are fully liberated. Furthermore, the hammer mill produced only a minor amount (0.4 w%) of PCA particles with a liberation degree of below 25%, whereas for the cutting mill 11 w% of PCAs had a liberation degree below 25%. If considering the composition of the sample where the PCA accounts for roughly 25 w% of the mobile phone, which is the starting level for the liberation, the cutting mill produces particles with a lower liberation degree than the mobile phone itself. One possible reason for this can be the larger output opening size in the cutting mill compared to the hammer mill. However, every mobile phone in the sample was crushed, and none of them passed the crushing chamber undamaged. When observing the distribution shape, it can be noticed that a secondary peak occurs for the cutting mill in the liberation degree class of 35–45%. For the hammer mill, such a distribution was not detected. Overall, it can be stated that the hammer mill is more selective than the cutting mill. The reasons behind selectivity are manifold, but the breaking system can be considered as one of key reasons. In the hammer mill, the system mainly utilizes an impact type of stress, whereas in the cutting mill shearing, compression and attrition types of stresses are present. When a mobile phone is positioned under a cutting head in the cutting mill, the head will cut through the phone from the position in question regardless of the materials (ductile, brittle) (Fig. 11). In the literature it has been proposed that the zone of deformation is around the knife edge and the failure occurs right next to the edge (Schubert and Bernotat, 2004). In this manner, components and parts will not detach from each other where the connection is weakest such as physical and surface joints or phase boundary regions, as presented in the literature (Heibeck et al. 2021; Richard et al. 2005). This is because, in the hammer mill, the energy from the hammers complemented with heavy bending and torsion is distributed on a larger area of the phone than in the cutting mill, which directs the energy on a smaller area, as has been presented in the literature (Kirchner et al. 1999; Sander et al. 2004; Schubert and Bernotat, 2004).

The liberation distribution was also examined in different size fractions (Table 2). Commonly, the liberation degree increases as the fraction becomes finer, which was seen in both of the crushers as well.

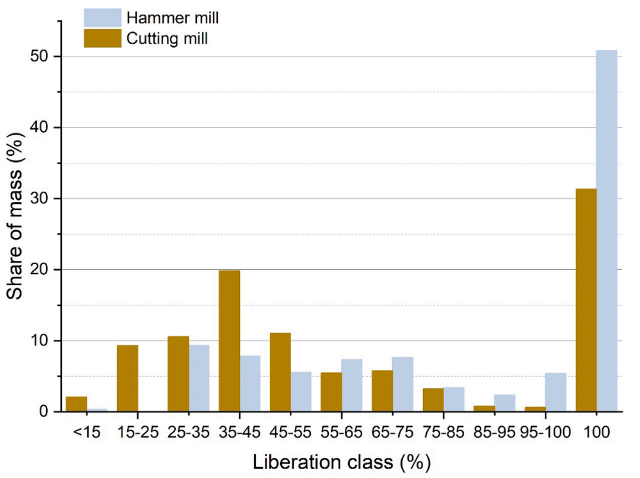

Fig. 10. Liberation distribution of PCA particles from the hammer and cutting mills. The amount of PCA particles analyzed in the liberation characterization was 1 661 for the hammer mill sample and 1 266 for the cutting mill sample.

Fig. 11. Photograph of a mobile phone cut in half in the cutting mill.

Table 2
The liberation distribution in two different size classes for the hammer and cutting mill, respectively.

PCA liberation class [%]	Hammer mill		Cutting mill	
	>31.5 mm	<31.5 mm	>31.5 mm	<31.5 mm
100	31.3%	88.5%	9.3%	71.4%
95	5.6%	4.9%	0.4%	0.9%
85	2.2%	2.6%	0.7%	0.9%
75	4.5%	1.3%	4.5%	1.0%
65	11.6%	0%	7.9%	1.9%
55	10.7%	0.8%	6.1%	4.3%
45	7.9%	1.0%	14.2%	5.3%
35	11.9%	0%	28.1%	4.7%
25	13.9%	0.6%	14.2%	3.9%
15	0%	0%	13.1%	2.4%
<15	0.4%	0.2%	1.4%	3.3%
Mass	66%	34%	65%	35%

However, a clear difference in the distributions was detected. In the larger size fraction (>31.5 mm), below 10 w% of the PCA was fully liberated with the cutting mill, whereas for the hammer mill the figure is above 30 w%. While roughly 34 w% of the PCAs have a liberation of 55% or less for the particles from the hammer mill, for the cutting mill the share is as much as around 71 w%. This is significant when the notable mass share of the large size fraction (>31.5 mm) is taken into account. For the finer size fraction (<31.5 mm), the fully liberated particles are clearly dominant, but roughly 20 w% of PCA particles from the cutting mill have a liberation degree of maximum 55%. This indicates that the cutting mill does not produce as clean fractions as the hammer mill. However, the grade of the PCA can be affected with a grate mounted in the output opening to generate a smaller particle size.

The recovery grade curves of PCA particles from both mills are presented in Fig. 12, which shows a distinct difference between the mills in the curves. The overall PCA grade (when the recovery is 100%) for the hammer mill is around 77%, whereas for the cutting mill it is around 58%. As the grade increases, the recovery starts to decrease, ending in values of 66% and 54% for the hammer and cutting mills, respectively. The hammer mill obtains higher values throughout the series, and the shape of its curve is also slightly more desirable as it has more curvature than the curve of cutting mill. This can be seen by comparing how the grade decreases, clearly slower for the hammer mill when the recovery is increased.

When comparing the PCA grades of both mill types against the share of PCA from the hand dismantling exercise (Section 3.1) which the provides actual amount of PCA in the identical input samples, the so-called total theoretical recoveries can be calculated. For the hammer mill, the total recovery is 73.2%, whereas for the cutting mill the value is 79.5%. Both of the mills reach moderate recoveries, although the higher total recovery for the cutting mill is surprising compared to the liberation results presented earlier. The main reason for this is the sampling carried out for the liberation analysis. Only particles above 4 mm entered the analysis, and PCA under 4 mm is not counted. As the hammer mill produced more fine material under 4 mm than the cutting mill, and comparing the amount of PCA in liberation analysis against the total PCA amount in the sample (Section 3.1), more PCA particles were lost in the fine fraction. These PCA particles lost in the fine fraction were not included in the liberation analysis. Overall, the hammer mill produced better liberated PCA particles compared to the cutting mill, but it loses some of them under 4 mm. Depending on the subsequent separation processes, the actual recoveries of PCA will be affected.

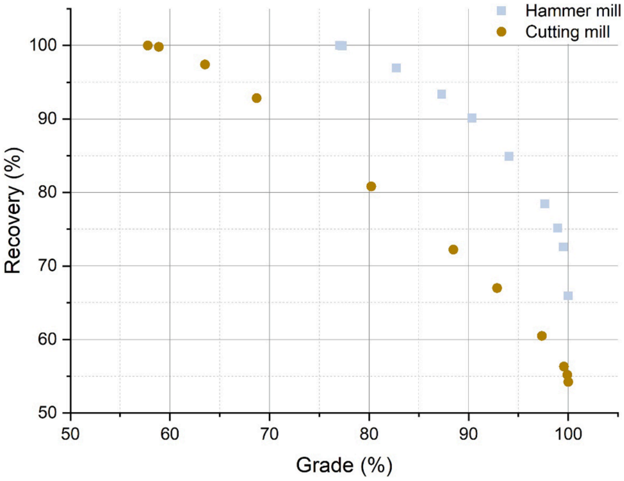

Fig. 12. Recovery grade curve of PCA particles from hammer and cutting mills. The amount of PCA particles analyzed in the liberation characterization was 1 661 for the hammer mill sample and 1 266 for the cutting mill sample.

3.5. Dust and fines composition

The chemical analysis results in Table 3 reveal that the dominant element in the dust (<1 mm) and fines (1–4 mm) fraction is silicon with concentrations of between 13.6 and 16%. Silicon originates mostly from the screens of the mobile phones and the glass fibre in the PCA. In addition, integrated chips such as semiconductors mounted on the surface of the PCB are composed of silicon (Palmieri et al. 2014). Under the impact and attrition stresses in the crusher, they can disengage and even pulverize due to their brittleness. This would also increase the silicon content in the dust fraction. When comparing the silicon contents from different crushers, it can be noticed that they are on the same level. In addition, a notable concentration in specific particle size fraction could not be detected.

The highest metal concentrations in the dust fraction could be found for aluminium, copper, tin, zinc and iron, which ranged from 0.65% for iron from the hammer mill to 4.55% for copper from the cutting mill. Aluminium and iron are used mostly in the structural parts of mobile phones such as frames, whereas copper is used mainly in the printed circuit board as the conductive layers between the resin and fibre glass structures and tin in the soldering and connecting pins (Cordella et al. 2020; Fontana et al. 2019; Holgersson et al. 2018; Manhart et al. 2016; Ueberschaar et al. 2017a; Yang et al. 2017). The hammer mill favours brittle materials in the size reduction leaving more ductile materials larger, while for the cutting mill the shearing stress cuts through the particle regardless of the material type (brittle/ductile) (Koyanaka et al. 1997). This may decrease the selectivity for the cutting mill. To some extent, this can explain the higher iron concentration in the dust from the cutting mill. Concerning the fines fraction (1–4 mm), it seems that aluminium, copper, tin and zinc tend to concentrate slightly to coarser fractions in both crushers. Iron makes an exception by behaving differently between crushers, as in the hammer mill a notably higher concentration can be detected, whereas in the cutting mill the concentration in coarser size fraction is lower. In the hammer mill, steel/iron parts most probably disengage during crushing and break to some extent but do not pulverise, which may occur in the cutting mill.

The content of precious metals was at the same level in the dust fraction for both mill types. Gold concentrations were highest between 401 and 415 mg/kg followed by palladium and platinum with concentrations of between 119 and 129 mg/kg and 1.7 and 4.9 mg/kg, respectively. When looking at the fines fraction, gold content increases whereas palladium and platinum concentrations decrease or stay at the same level. In addition, some variations between the mills can be detected. Especially for gold, the cutting mill has higher concentrations in the fines fraction compared to the hammer mill. However, when calculating the masses of precious metals in the dust and fines fractions from both mills, the hammer mill generates roughly twice as much precious metals compared to the cutting mill. The sources of precious metals in the dusts and fines are manifold. Based on the literature, gold is used especially on connectors as it has good electrical and chemical properties, such as excellent corrosion resistance and high electrical conductivity. In addition, gold can be used in wire bonding between the semiconductor chip and the frame in a semiconductor package. (Charles, 2020; Goodman, 2002) As for palladium, it is often used in various surface-mounted components such as ceramic capacitors, chip resistors, integrated circuits and inductors. In addition, palladium can be used together with gold in connections. (Charles, 2020).

Table 3
Element composition of dust (<1 mm) and fines (1–4 mm) fraction from both crushers.

Element	Unit	Hammer mill		Cutting mill	
		<1 mm	1–4 mm*	<1 mm	1–4 mm*
Ag	%	0.13	0.16	0.14	0.14
Al	%	1.62	2.11	1.55	2.11
As	%	0.06	0.05	0.05	0.06
Cu	%	4.30	8.58	4.55	8.97
Cr	%	0.07	0.52	0.30	0.27
Fe	%	0.65	4.65	2.87	1.91
Pb	%	0.08	0.17	0.09	0.28
Sb	%	0.29	0.60	0.24	0.49
Si	%	14.20	13.61	16.00	15.00
Sn	%	1.77	2.53	1.32	2.39
Zn	%	0.30	1.34	0.69	1.06
Au	mg/kg	415	573	401	845
Pd	mg/kg	119	33.84	129	40.72
Pt	mg/kg	4.87	1.52	1.71	1.36

* The results are computational based on elemental analysis of two size fractions (1–2 mm and 2–4) and their mass distribution.

From the environmental and safety point of view, Wang et al. have reported that the metals in the dust/fines are mainly in simple substance form, and low-melting point metals, such as lead and tin as well as halogen elements, have been found in dust samples (Wang et al., 2015). Taking this into consideration, safety issues need to be addressed when further treatment of fines is planned.

3.6. Fate of metals in dust and fines fraction

In the mechanical treatment of WEEE, dusts represent usually a maximum of a few per cent of the total through-put of the plant. Due to their complex and heterogenic nature and composition, they are often primarily considered through safety and environmental aspects.

As a result, in usual circumstances dusts represent a cost to the plant which arises from their correct manner of disposal. Such methods of disposal of the material are focused, for example, on incineration and seldom on material recovery. Simultaneously, valuable metals are lost through dusts.

When comparing the element composition of dusts and fines in this study with the composition of mobile phones presented in a literature review, it can be identified that for valuable metals such as silver, gold and palladium as well as silicon and tin, the concentration is at the same level or higher in the dust and fines (Ueberschaar et al. 2017a). For the base metals (Al, Cu, Fe), higher concentrations can be detected in mobile phones. Considering the low amount of dust generated in the plant compared to other fractions such as the magnetic metal fraction, non-magnetic metal fraction and plastic/mixed fraction, generally speaking below 10% of metals end up in this fraction. However, when taking into account the fines fraction in the calculations, for some metals such as gold, over 20% can be lost to this fraction in the case of the hammer mill.

From the economic potential viewpoint, one could reflect the concentrations of most valuable metals, such as gold, palladium and silver, in the dusts to the concentrations found in PCAs, which is usually the most valuable component in the EEE. PCAs are generally divided into three economic grade groups, namely high, medium and low (Hagelüken 2006). High-grade PCAs can be found, for example, in mobile phones and notebooks, whereas medium-grade PCAs in televisions and low-grade PCAs in refrigerators and printers (D'Adamo et al. 2019). Especially the gold and palladium concentrations are the greatest economic value in PCAs, and comparing the concentrations in the dusts and fines to these grade groups they would be in the high-grade PCA, which makes them an interesting fraction from the economic viewpoint. Looking more closely at the differences between the hammer and cutting mill, the former generates more dust, and the loss of valuable metals is higher even though the concentrations of the valuable metals are lower. However, as the liberation of PCA is better for the hammer mill, a higher quality PCA fraction is expected to occur, which may in turn affect the total revenue from mobile phone treatment. It should be noted that, if WEEE is treated as a mixed feed or even with the division of the equipment based on the PCA grade, the dust composition will be different and may require consideration of other aspects such as safety and environmental.

4. Conclusions

This study showed that different crusher types have an influence both on the liberation distribution of PCA as well as in the fines generation and their composition. While a fast-rotating hammer mill produced better liberated PCAs with an overall PCA grade of 77% compared to a slow-rotating cutting mill with 58% PCA grade, it produced over two times the amount of fines compared to the cutting mill. Presumably, the breaking stress in the hammer mill is spread over a larger area on the particle, while in the cutting mill it is more exerted and controlled. As a result, more valuables are lost with the hammer mill to the fines. On the other hand, in further mechanical processing, better liberated PCA particles have greater opportunities to end up in the target fractions, which increases the recovery of them.

The dust is composed of the same elements, silicon being the dominant, followed by copper, aluminium, iron, tin and zinc in both cases. As for precious metals, gold had the highest concentrations, followed by palladium and platinum. No great difference between the mill types was detected. However, when extending the consideration to the fines fraction, higher metal concentrations, for example, for gold and copper could be detected for the cutting mill. The shearing type of encounter between the particle and cutting blades in the cutting mill emphasizes ductile materials such as gold plated connectors.

When observing the dust and fines fraction in a boarder context, it is not the most relevant fraction in a technical plant as it generally counts for a few per cent of the throughput. However, as it is composed of both valuable elements relative to medium/high-grade PCA and harmful ones, it should not be discarded. Further treatment options should take both of these aspects into consideration.

Beside future research on dust treatment methods, further investigations with different type of shredders and shears as well as targeting also other parts and components within electrical equipment would complement the study by providing additional information on the effect of machinery design and other parts effect on the topic. In addition, a deeper mineralogical characterization study on the dust fraction may supplement the study with morphology of the small particles.

CRediT authorship contribution statement

J. Bachér: Conceptualization, Methodology, Formal analysis, Investigation, Visualization, Writing – original draft, Writing – review & editing. **L. Rintala:** Visualization, Writing – review & editing, Supervision. **M. Horttanainen:** Writing – review & editing, Supervision.

Declaration of Competing 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.

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