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Electro-Hydraulic Fragmentation vs Conventional Crushing of Photovoltaic Panels – Impact on Recycling

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Abstract

Currently, the first generation of solar panels are reaching their end-of-life, however so far, there is no best available technology (BAT) to deal with solar panel waste in terms of the optimized circular economy of metals. In this brief communication, electro-hydraulic fragmentation (EHF) is explored as an initial conditioning stage of photovoltaic (PV) modules to facilitate the recovery of valuable metals with the main goal to produce liberated fractions that are suitable for the retrieval of materials like Si, Ag, Cu, Sn, Pb, and Al. When compared to traditional crushing, the results suggest that dismantling of PV panels using EHF shows more selectivity by concentrating metals among well-defined particle size fractions. Using this method, the subsequent recovery of metals from PV panels can be achieved in a straightforward manner by simple means like sieving. The fragmentation achievable with EHF technology allowed approximately 99% Cu, 60% Ag, 80% of Pb, Sn and Al total elemental weight within the solar panels to be concentrated solely within the > 4 mm size range, whereas high purity (> 99%) Si could be found in the fractions between > 0.50 mm and < 2 mm. To the best of the authors' knowledge, this paper presents for the first time a comparative analysis on the use of EHF technique and conventional crushing for the processing of PV solar panel waste.

Keywords: Solar panel waste; Secondary raw materials; Metal recovery; Circular economy; Closed loop; Mechanical processing.

1. Introduction

By the end of 2016 it was estimated that photovoltaic (PV)-based energy provided an estimated 303 GW of energy globally, nearly 100% increase from the previous year as shown in Figure 1 (Werner et al., 2014; Xu et al., 2018). This already significant contribution to the world's energy grid is projected to continue to rise in the coming years, strongly driven by the need to reduce the level of greenhouse gas emissions from fossil fuels related to electricity production (Xu et al., 2018). In Europe alone as of 2016, PV energy was responsible for nearly 70% of the global renewable generation making Europe the front-runner in utilizing PV based renewable energy (Latunussa et al., 2016).

At present, the amount of EoL PV panels is relatively low when compared to other WEEE or batteries, which is one of the main reasons why bespoke recycling processes have not yet been fully realised (Bogacka et al., 2017). On the other hand, it is predicted that the amount of EoL PV panels will increase markedly over the next twenty years with predictions that will be over a million tons of waste PV panels by 2030 (Rocchetti and Beolchini, 2015), rising to about 9.6 million tons by 2050 (Monier and Hestin, 2011). With this increase in PV waste, steps are being taken to reduce the environmental impacts as exemplified by inclusion of solar panel related materials in the latest EU Waste Electrical and Electronic Equipment (WEEE) directive (Shin et al, 2017; Xu et al., 2018). This legislation obliges those who are involved in PV manufacture, supply and sales to ensure the collection and safe recycling at end of life, although this only applies within the EU. Nevertheless, currently the effective management of PV waste remains an issue with only low rates of environmentally responsible recycling performed worldwide as significant amounts are still illegally disposed of in developing countries resulting in numerous pollution incidents (Bakhiyi et al., 2018).

Globally, there are currently no fully dedicated PV module recycling plants in operation and at present, EoL PV modules are typically processed with laminated glass or other WEEE at general recycling facilities (Pennington et al., 2016; Wambach and Sander, 2015). On the other hand, end-of-life solar panels, contain significant amounts of valuable (Ag, Cu, Ga, In), desirable (Al and Si) and even some environmentally toxic (Pb, Cd) elements. In addition, the presence of critical metals like Te and Se (in copper indium gallium diselenide (CIGS) based PV modules) make the recycling of solar panel waste at EoL of crucial importance (Graedel et al., 2015). For example, Ag content in EoL c-Si based PV panels is approximately 600 g/t, a competitive amount when compared with primary Ag production where a minimum of 700 g/t is deemed necessary in order to be economically viable (U.S. Geological Survey, 2018). Furthermore, based on the current rates of solar energy growth, it is projected that by 2050 the total amount of Ag utilized in PV cells will be equivalent to ~10% of the world's total reserves (Dias et al., 2016).

Nonetheless, it is imperative that specific sustainable methods for PV recycling are established in order to recover the valuable metals present without contamination of the environment. At EoL when PV modules are uninstalled, the Al frame and the junction box are typically removed before the crushing process to facilitate recycling (Tao and Yu, 2015). One of the main challenges of recycling PV panels is that the PV cells and associated metals are laminated between two-ethylene vinyl acetate (EVA) sheets to protect them from air, water and other impurities, which enables a long life span (Xu et al., 2018). However, this assemblage hinders separation efficiency at EoL as valuable metals like Ag remain trapped between EVA laminates (when using typical crushing technologies). This may be overcome by exposure of separated panel modules to acids, organic solvent (Doi et al. 2001; Kim and Lee, 2012) or heat treatments (Dias et al., 2017; Fiandra et al., 2019). However, a recent study on PV panel demanufacturing techniques has shown that although chemical and thermal techniques result in improved recoveries of glass, Al, Fe, Cu and Ag when compared with current industrial practice, selective separation allowed over 90% of Cu and Ag to be recovered against < 85% for thermal/chemical methodologies (Duflou et al. 2018).

At the same time, new fragmenting technologies such as Electro-Hydraulic Fragmentation (EHF) – also referred to as High-Voltage Pulse Crushing - are emerging, offering new liberation possibilities for higher separation efficiency. Shockwave technology or EHF processes use mechanical shockwaves, generated in a fluid medium such as water, to fragment different materials (Figure 2). These vibrations are produced between electrodes via an electro-hydraulic mechanism that leads to brief, but intensive shockwave impulses that propagate through fluid, impinge on the material resulting in breakage at mechanically weak points between different material interfaces such as at joints. Although EHF has long been used for mining, construction or geological applications like rock fragmentation (Bluhm et al., 2000; Pijaudier et al., 2016), the technique is being increasingly applied in the area of waste electronics recycling to crush liquid crystal displays (LCD) (Dodbiba et al., 2012) and printed circuit boards (PCBs) (Duan et al., 2015; Zhao et al., 2015). Among the reported advantages offered by EHF compared to traditional crushing are: i) high selectivity of fragments of materials; ii) minimal contamination due to the process being contactless; and iii) absence of fine dust released to the environment during processing, all of which make EHF a more environmentally cleaner process *cf.* crushing (Andres, 1985).

A recent investigation by Akimoto et al. (2018) has previously demonstrated that EHF also offers an interesting alternative for the comminution and liberation of polycrystalline silicon photovoltaic panels. Consequently, this study compares EHF with industrially crushed samples in order to determine the effect of dismantling technique on the partitioning of different component parts after sieve fractionation. As the primary focus of this research is on the potential of PV waste as a valuable secondary raw material (SRM), the resultant fractions are analysed to ascertain the content of materials like Si, Ag, Cu, Sn, Pb, Al, to deduce their suitability for subsequent recovery processes.

2. Materials and methods

2.1. Materials

The material used in this research comprised of a commercial c-Si based PV panel (Salosolar, Areva Solar Oy, Finland) from which the Al frames and junction box had been removed prior to crushing or EHF treatment. While the exact composition of a c-Si PV depends on the model and manufacturer, content typically comprises of 76% glass from the panel surface, 10% polymer (including encapsulant and backsheet foil), 8% Al from the frame, 5% Si semiconductor, 1% Cu and < 0.1% Ag from connectors, whilst the remainder is other metals like Pb and Sn (Weckend et al., 2016). The quantities of the metallic/inorganic components within the panel were verified by using small sections (10 - 20 g) that were cut and treated with 100 ml of Aqua Regia. The resultant solutions were analysed by AAS (atomic absorption spectroscopy, Varian AA240, USA) and ICP-OES (inductively coupled plasma, optical emission spectroscopy, Perkin Elmer Optima 7100 DV, USA). The total amount of organic matter within the panel matrix was found by measuring the weight loss of the material after treatment in a muffle furnace and whilst there is the possibility for weight gain due to metal oxidation at such temperatures, this was considered to be negligible. The overall weight composition of the PV panels (without the aluminium frame) as wt. % is presented in Table 1.

2.2. Industrially Crushed c-Si based PV panel (Method 1)

Following removal of the Al frame and junction box, the remainder of the PV panel was cut into smaller pieces (approximately 20 cm × 30 cm). These sections of PV panel were then subjected to an industrial crushing process at Kuusakoski Recycling, Heinola using a rapid granulator with a multi-blade, triple knife rotor (AB Rapid GK300, Maskin, Sweden) to a target particle size of 15 mm.

2.3. Electro-Hydraulic Fragmentation (Method 2)

As an alternative to conventional crushing, two sets of similar PV panel samples were fragmented using EHF technology with an IMPULSTEC EHF 400 apparatus (ImpulsTec GmbH, Germany). PV panel material was cut into pieces with an approximate size of 12 cm x 8 cm and weight of 85 g. A maximum of six samples were then placed in a 2L capacity EHF reactor and the vessel filled with water. All experiments were performed using 600 Joules of energy to generate each shockwave impulse and two different impulse durations of 300 and 500 at a rate of 1 impulse per second - equivalent to experimental times of 300 and 500 seconds, respectively - were used to assess their influence on separation. After the experiments were complete, the fragmented material was collected and dried for 24 hours at 60 °C in a muffle furnace.

2.4. Particle Size Classification

Following the different comminution processes, the industrially crushed and electro-hydraulic fragmented PV panel materials were sieved separately with nominal mesh sizes of 8 mm, 4 mm, 2 mm, 1 mm, 500 μm and 250 μm. Separation was performed with a bench scale-vibrating sieve (AS 300, Retsch, Germany) for 20 minutes at an amplitude of 5.0 mm to facilitate complete screening. The mass of sieved material was of 202 g for industrially crushed raw material, and of 354 and 507 g for EHF treated material with 300 and 500 impulses. Additionally, the fractions > 1 mm and > 2 mm from the EHF raw materials were sieved for a second time, for 10 seconds with an amplitude of 5.0 mm, to separate the copper wire from the rest of the material. Fractions from both methods were then subjected chemical analysis to determine the relative quantities of organic and inorganic components.

2.5. Chemical Analyses

In order to determine quantities of the metallic/inorganic components in each fraction, between 10-20 grams of the crushed material – from either the industrial or EHF method - was subjected to total leaching in 100 ml of Aqua Regia. The resultant solutions were analysed by AAS (atomic absorption spectroscopy, Varian AA240, USA) and ICP-OES (inductively coupled plasma, optical emission spectroscopy, Perkin Elmer Optima 7100 DV, USA). Similar portions of each fraction were heat treated at 650 °C in a muffle furnace for 12 hours such that the organic content could be established via weight loss, assuming any additional weight due to metal oxidation was minimal under these conditions.

3. Results and Discussion

3.1. Crushing of c-Si Based PV Panels (Method 1)

During crushing of the c-Si PV panels, seperation of the EVA bonded to the glass and PV was found to be challenging due to the very strong bonding between the materials, consequently, several crushing runs had to be conducted to attain the target particle size of 15 mm. Previous work by Dias et al. (2016) has tried to address this issue by manual removal of the front glass and adhesive layer prior to the knife milling of solar panel samples, nevertheless, their results showed that EVA remained attached to the PV cells, which made the separation of metal-rich components challenging. In the current work, the crushing process resulted in a heterogeneous material with various particle size fractions. This is illustrated in Figure 3, which shows the particle size fractions obtained for the industrially crushed PV material after screening with various mesh-opening sizes.

The distribution of different materials in the six different particle size fractions is presented in Figure 4. It can be observed that almost 90% of organic matter from industrially crushed PVs is present in the fractions with a size > 2 mm. In contrast, a majority of Cu is present within the > 1 mm fractions, with an almost even distribution (ca. 40% each) found in the 2 - 4 and > 4 mm portions, whereas a further 18% present at 1 - 2 mm. A similar pattern is also found for Pb and Sn as nearly 80% of these metals are found in the > 2 mm region, with a near equal distribution among 2 - 4 and > 4 mm particle sizes. This similarity in observed dispersion of Cu, Pb and Sn results from the use of PbSn as the soldering material for the Cu current collectors and the results suggest that most of the PbSn solder remain attached to the Cu wires. Nonetheless, in the case of Pb and Sn, ca. 10% is also found in the fines (< 0.25 mm) indicating that a small portion of this material is separated and ground down during crushing. In the case of the remaining materials, Al can be seen to dominate at both smaller (< 0.25 mm) and larger (> 4 mm) particle sizes, both having ca 40% share, whereas approx. 70% of Si is the fractions of > 1 mm with the biggest fraction (over 50%) of this at sizes > 2 mm. These results demonstrate that the industrial crushing of solar panel waste leads to a lack of selectivity in terms of material distribution among the screened fractions, which in turn would complicate the next recovery steps in any future recycling process.

3.2. Dismantling by EHF Technology (Method 2)

EHF for PV panel dismantling was investigated using shockwaves generated by a 600J discharge, at a rate of one impulse per second, for two different durations of 300 and 500 impulses, respectively. The various fractions resulting from fragmentation of PV material with 300 impulses – after collection, drying and sieving into six fractions (> 4 mm, 2 - 4 mm, 1 - 2 mm, 1 – 0.5 mm, 0.25 - 0.5 mm and < 0.25 mm) – are shown in Figure 5. As can be seen, when compared to the analogous fractions from industrial crushing, the material from EHF is more homogenous in appearance, particularly at sizes < 2 mm.

Figure 6a and b depicts the distribution of different materials among the different particle sizes after EHF treatment of 300 and 500 pulses, respectively. In both cases, it can be seen that the elemental distribution across the different fractions differs remarkably from those observed for the crushed panels (Figure 4), for example, the Si was found to be more evenly spread throughout the different size portions *cf.* crushing. In contrast with the 300-impulse treatment, over 95% of organic matter and Cu were to be found in largest (> 4 mm) fraction along with ~80% Pb, Sn, Al and 60% of Ag. Interestingly, a majority of the remaining Pb, Sn, Al, Ag was determined to be in the fractions < 0.5 mm showing that there is a clear partitioning of these metal materials between the highest and lowest screen sizes with EHF.

A similar type of distribution is also seen for the PV panel material from the 500-impulse treatment (Figure 6b). Once again, most of the Cu and organic matter are concentrated within > 4mm particle size, although in this case the levels of Pb, Sn, Al and Ag while still relatively high, they are proportionally smaller compared to those obtained with 300 impulses - approx. 60% vs. 80% for Pb, Sn, Al and ~40% vs. 60% for Ag. Similarly, to the 300-impulse results, the 500-impulse data also shows that the lower fractions (< 0.5 mm) contain a significant portion of remaining metals (except Cu) with over 30% Pb, Sn, 20% Al and 40% of Ag present. It is also worth noting that in both EHF cases, there is a relatively low percentage of metals in the fractions between 1 and 4 mm.

3.3. Electro-Hydro Fragmentation Technology vs. Conventional Industrial Crushing

When EHF technology for the dismantling of PV panels is compared with traditional crushing methodology, it can be observed that there are distinct differences in the distribution of metals among the specific fractions with EHF treatment leading to a concentration metals in either the highest (> 4 mm) or two lowest sizes (< 0.5 mm). This observed behaviour of EHF that makes it different from conventional crushing could be explained by the absence of discrete contact points of impact in the former. Consequently, comminution forces can be assumed to be more evenly distributed across the entire sample and thus, the degree of particle size reduction is more strongly dependent on the brittleness of the materials only. In addition, the two EHF durations employed in this study also produced some change in the distributions of components, which suggests that a variation in the number of impulses used could provide an opportunity to selectivity concentrate metals to specific size fractions. This potential for selectivity with the EHF method observed in this research has also been indicated by recent studies on recycling of WEEE materials (Dodbiba et al., 2012; Zhao et al., 2015; Akimoto et al., 2018).

A more detailed analysis of the individual fractions in terms weight percentage of the various different components are shown in Figure 7 (a-c). From the results shown (Figure 7a-c) irrespective of the method of dismantling the majority of all the fractions is composed of Si and that the organic components are generally found in the highest fraction (> 4 mm). A comparison of the metal only distributions with crushing and EHF (Figure 8a-c) indicates that there are distinct differences between the two methods. In particular, although in all cases - crushed, EHF with 300 and 500 impulses - the highest amounts of metals are present in the > 4 mm fraction only the EHF method leads to an effective partitioning of metal components between the > 4 mm and < 0.5 mm portions, respectively. Industrially crushed PV-panel fractions in contrast, were found to have relatively high levels of metals present in every fraction with, for example, Cu elemental wt. % of between 20 and 40% detected in the fragments > 1 mm.

A comparison of the results from EHF with 300 and 500 impulses - both with the same 600 J shockwave and impulse rate of one per second - demonstrates that a difference in the total number of impulses results in distinct differences in the separation of the metals present. For example, use of the > 4 mm fraction from the 300-impulse treatments in a subsequent recycling process would potentially allow up to 99% Cu, 60% Ag and 80% of Pb, Sn and Al respectively from within a PV-panel to be recovered (Figure 8b). The same fraction from the 500-impulse treatment contains lower amounts of metal (Figure 8c, 98% Cu, 40% Ag and 60% of Pb, Sn, Al) although when compared to industrially crush material (Figure 8a, 38% Cu, 30% Ag and 35% of Pb, Sn, Al) the levels are still enhanced. Moreover, if the > 4 mm and two fractions < 0.5 mm from the 300-impulse tests were combined prior to further processing, this new material would contain almost 100% of the Cu, Pb, Sn, Al and ~90% Ag available from the PV-panel.

In addition, this effective separation of metals to the highest and lowest fractions also provides high purity (> 99%, Figure 7b and c) Si containing fractions (from > 0.50 to < 4 mm), which themselves can be amalgamated to be recycled for re-use in new solar panel arrays or other related applications. It should be noted that the Si fraction might comprise of both semiconductor and panel glass though we make no clear distinction here. Nevertheless, this would still indicate that the various size fractions produced using EHF with a size < 4 mm, have only two components to be subsequently separated, as the organic and metallic fractions are only present in trace amounts. Moreover, as this study only used two EHF impulse durations (300 and 500 impulses) with the same shockwave energy and impulse rate, further investigations are needed in order to better understand the distribution patterns and reproducibility of the technique with different of waste solar panel configurations in order to optimize the metal partitioning.

4. Conclusions

In an effort to develop a suitable recycling method for PV solar panels, the EHF technology method has been employed for the first time to facilitate the separation and recovery of metals from PV modules. In particular, the dismantling of PV panels using shockwave technology provides more selectivity by concentrating target metals on specific particle size fractions. This enrichment of metals within certain fractions enhances the prospects for metal recovery as the higher concentration materials facilitates their subsequent processing and extraction via current industrial processes e.g. black copper smelting. Furthermore, the EHF process also gives rise to high purity Si containing fractions that can also be more readily recycled, thus making the whole approach more economically attractive further enhancing the possibility for a closed loop approach to solar panel recycling.

Overall, use of EHF on photovoltaic solar panel waste offers a straightforward alternative solution for the combined rapid dismantling-fractionation of valuable metal and metalloid components from EoL panels.

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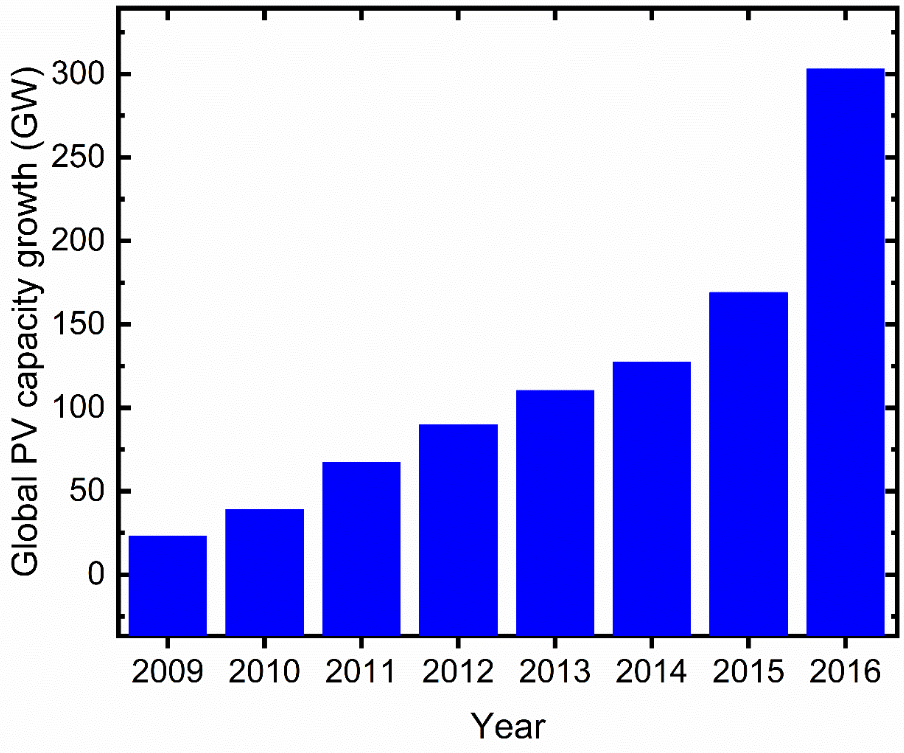

Figure 1. Trend in global PV installed capacity (Werner et al., 2014; Xu et al., 2018).

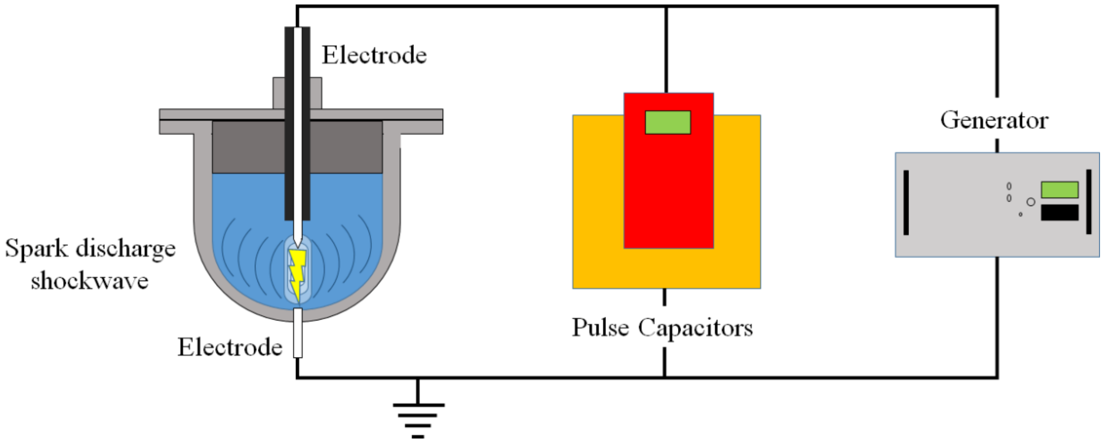

Figure 2. Schematic diagram of an EHF set-up discharge circuit for shockwave generation.

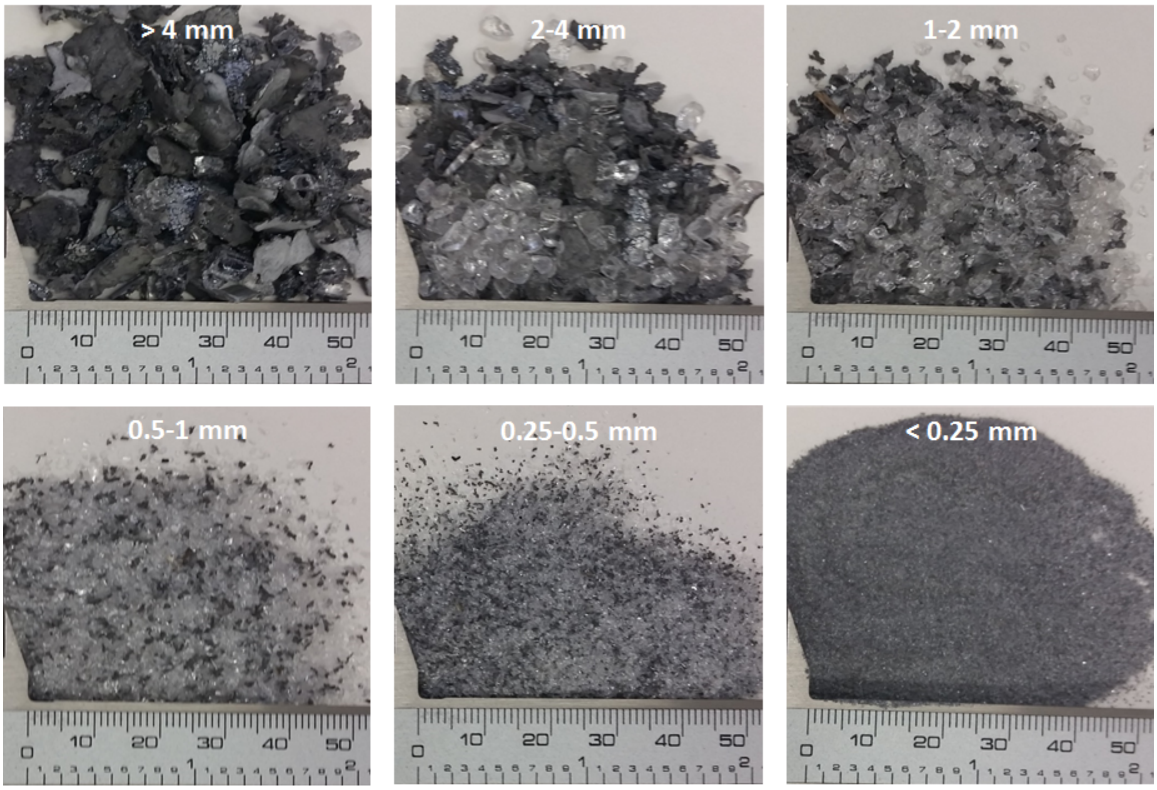

Figure 3. Images of sieved fractions of crushed photovoltaic panel at different particle size fractions (> 4 mm, 2 - 4 mm, 1 - 2 mm, 1 – 0.5 mm, 0.25- 0.5 mm and < 0.25 mm).

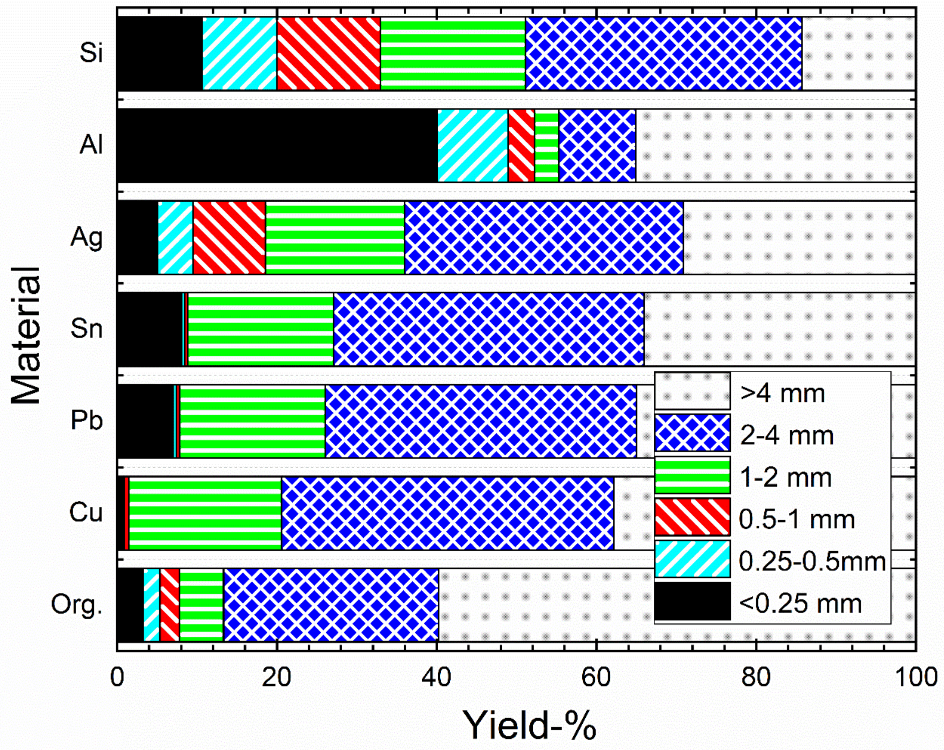

Figure 4. Distribution of Al, Ag, Sn, Pb, Cu, and organic matter in wt. % among the sieving (> 4 mm, 2 - 4 mm, 1 - 2 mm, 1 – 0.5 mm, 0.25- 0.5 mm and < 0.25 mm) for industrially crushed PV panel.

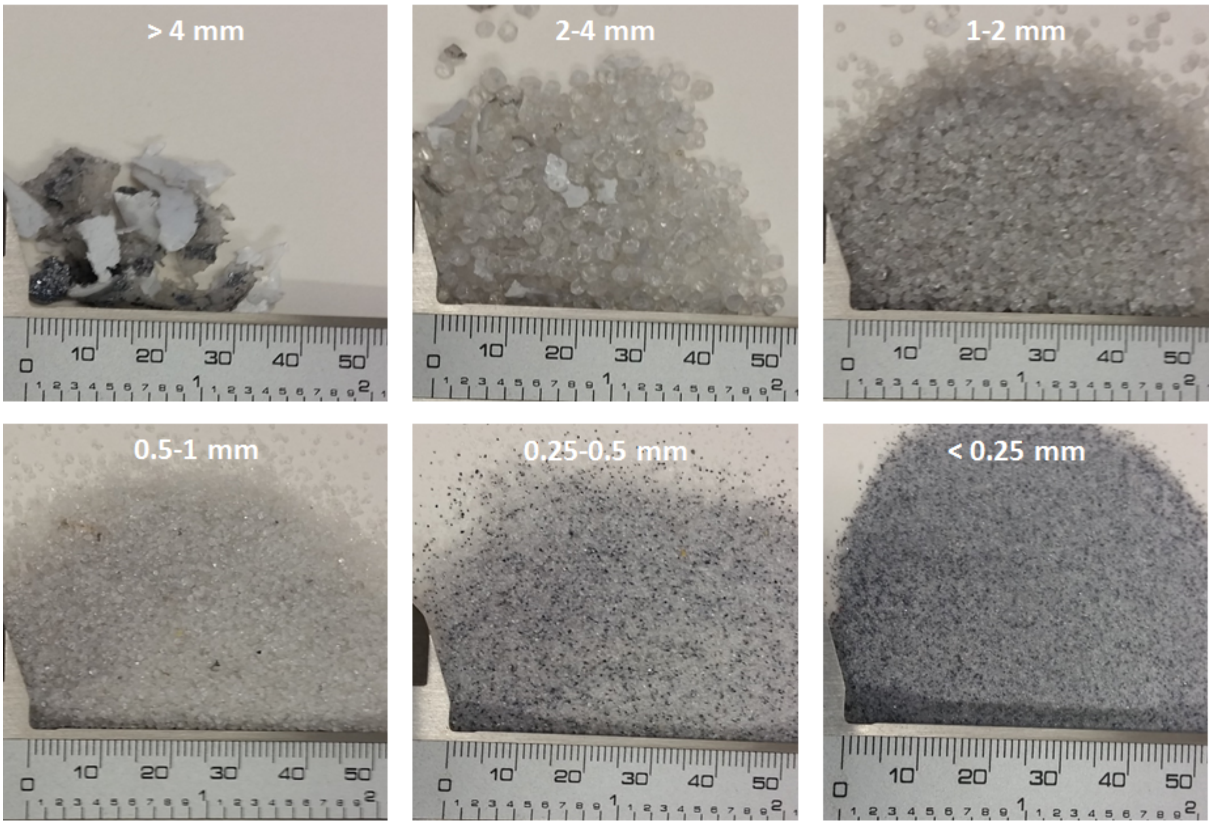

Figure 5. Images of fractions of sieved dismantled PV panel by shock wave technology at 300 impulse, (> 4 mm, 2 - 4 mm, 1 - 2 mm, 1 – 0.5 mm, 0.25- 0.5 mm and < 0.25 mm).

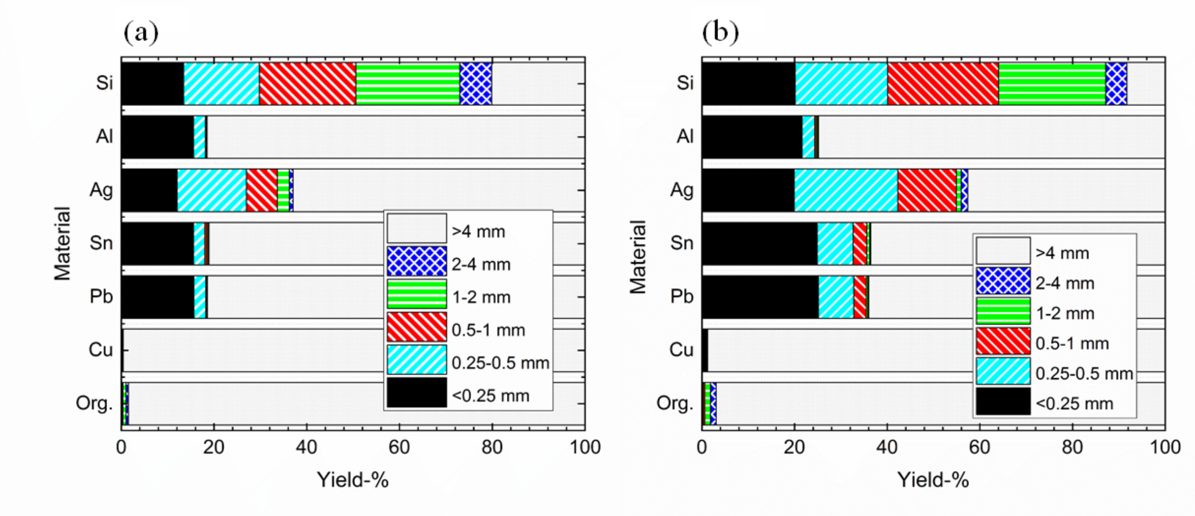

Figure 6. Distribution of Al, Ag, Sn, Pb, Cu, and organic matter percentage among the sieving openings (> 4 mm, 2 - 4 mm, 1 - 2 mm, 1 - 0.5 mm, 0.25 - 0.5 mm and < 0.25 mm) after dismantling with EHF treatments comprising of (a) 300 and (b) 500 impulses, respectively.

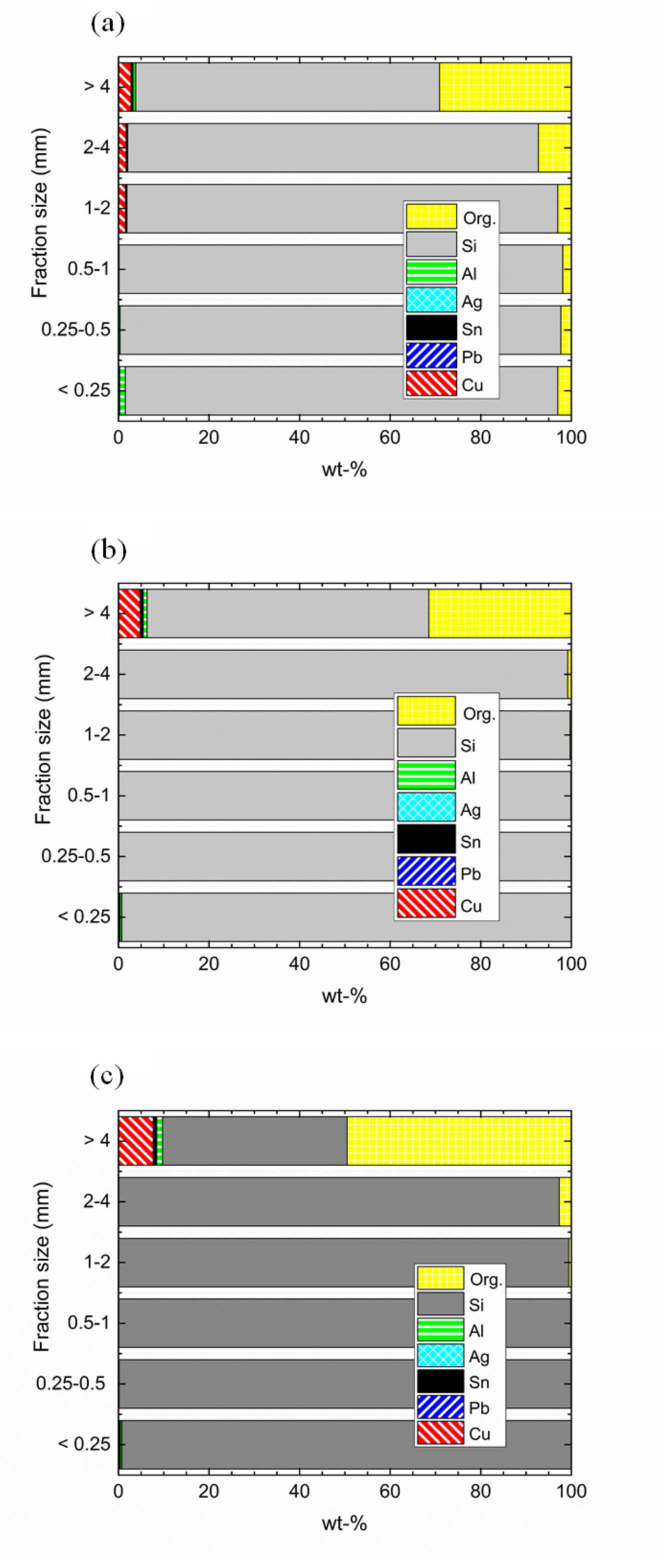

Figure 7. Weight percentage distributions of all components among the different fractions after dismantling with (a) industrial crushing, (b) 300-impulse and (c) 500-impulse EHF treatments.

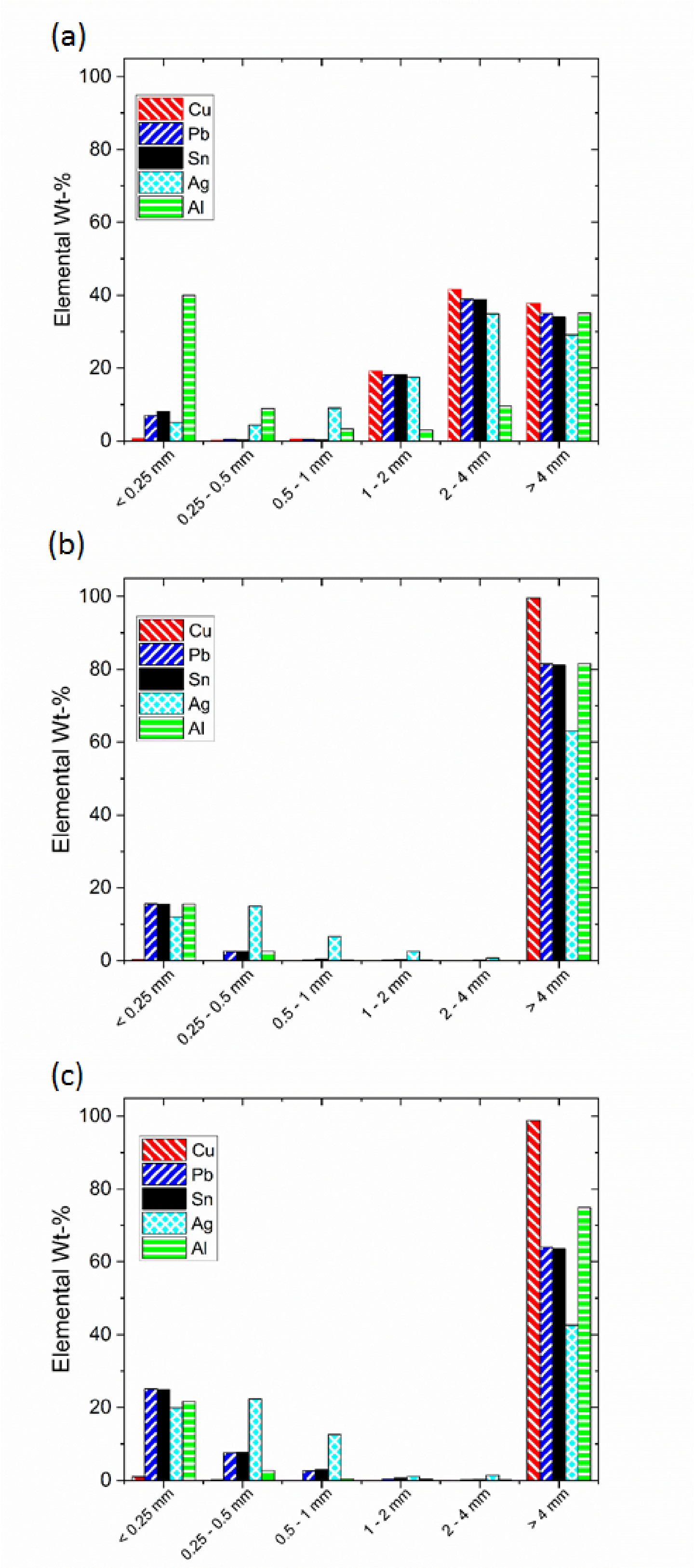

Figure 8. Weight distributions among the different fractions of all metal components - as total individual elemental content of the PV-panel - after dismantling with (a) industrial crushing, (b) 300-impulse and (c) 500-impulse EHF treatments.

Table 1. Composition of the industrially crushed EoL raw material of c-Si based PV panel with Al frame removed.

Material	**Organic matter**	**Cu**	**Pb**	**Sn**	**Ag**	**Al**	**Si**	**Others**
Amount wt. %	9.2	1.4	0.08	0.1	0.004	0.3	88.9	0.016