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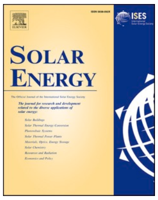

Electrohydraulic fragmentation processing enabling separation and recovery of all components in end-of-life silicon photovoltaic panels

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ABSTRACT

The exponential increased use of PV panels for energy production would also lead to enormous volumes of PV waste that need to be dealt with in an environmentally responsible manner. In this work we present experimental results for recycling crystalline silicon (c-Si) PV panels using recently developed electrohydraulic shock wave-based fragmentation of PV panels. The electrohydraulic fragmentation process allows for the efficient delamination of the modules and subsequent recovery of almost all valuable materials used in the manufacturing of PV panels, without thermally decomposing the polymers and eliminates creation of any toxic or hazardous waste during the process. We study the impact of the type of panel, size of the feed material and process duration on the quantity and quality of material recovered after the process.

1. Introduction

1.1. *Background succinct literature review*

There has been a rapid growth of photovoltaic (PV) installations in the past decade. Global installed capacity of PV reached 1625 GW at the end of 2023 (Gaëtan [22], and is expected to reach 8520 GW by 2050IRENA [26]. The exponential growth of PV installations is an important and desirable element in the global response to climate change. A typical PV panel has a lifetime of 20–25 years. However, some will be damaged during production, transportation, handling, or installation, and some will even stop functioning after a shorter time, while some would be discarded during testing called as factory reject panels. There is an urgent need to establish efficient measures to deal with the upcoming enormous volumes of PV waste in an environmentally responsible manner. According to the predictions published by International Renewable Energy Agency (IRENA), the cumulative PV panel waste could reach 1.7——8 million tonnes in 2030 and 60 – 78 million tonnes by 2050 for installed capacity of 4500 GW [66]. Given an average panel life of 25 years and the increased production and deployment of PV panels, large amounts of PV wastes are anticipated by early 2030 s. It is expected that by 2040, the weight of waste PV panels generated would match that of the new installations [66].

The growing amounts of PV panel waste presents new environmental and waste management challenges. However, with proper waste management procedures in place, this challenge could present us with unprecedented opportunities to pursue new technological and economical avenues which could increase from US$ 450 million in 2030 to US$ 15 Billion in 2050 [66]. Recovery of raw materials from the PV panels could lead to the establishment of new solar PV end-of-life industries. In fact, solar PV recycling would be of paramount importance for the transition into a sustainable and economically viable renewables-based energy future. To unlock the mechanisms and advantages of recycling PV panels, it is important to research different avenues of reusing the end-of-life PV panels in time to meet the expected surge in the PV panel waste. In many countries, stringent guidelines are already in place to deal with Waste Electrical and Electronic Equipment (WEEE) [11,5]. Especially the EU has published one of the most detailed regulations related to recycling of PV panels. These regulations stipulate a minimum of 80–85 % recovery or reuse of materials [59,7,47]. Hence, it is important to consider the pathways in which the components could be recycled or reused most effectively, minimizing the generation of the pollutants, unusable waste and contaminated by-products of the panel recycling process.

End-of-life PV panels could provide valuable metals such as aluminium, copper and silver. Additionally crystalline silicon, and glass can also be recovered and recycled [33,54,2]. While the economic impacts of recycling PV panels was discussed earlier, efficient recycling of PV panels can further improve the sustainability of PV industry by safeguarding against the depletion of expensive metals such as silver and

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copper and avoiding toxic emissions into the environment. Further they could also prevent future investments and efforts to produce virgin materials, thereby further protecting the environment from the ill-effects of mining [56]. Various recycling processes have been developed to recover these raw materials from panels, exhibiting different processing efficiencies, recycling yields, costs, and environmental impacts [56]. In some cases, complete Si ingots and solar cells have been manufactured using recycled Si [17,49]. A comparison of the life cycle assessments for different recycling processes have shown the considered recycling scenarios can reduce environmental impacts, boost ecosystem, promote human health and make resources more resilient compared to landfill and incineration scenarios[39].

End-of-life PV panels are usually crushed and used for low value applications, essentially downcycling the materials [23,36,37,24]. However, to recover the valuable materials from the end-of-life PV panels, they should be viewed as a resource rather than waste. To extract valuable materials from the end-of-life PV panels, usually four steps are required – disassembly, delamination, material sorting and material extraction. Each of these steps could further comprise single or multiple process steps [15,8]. The external wiring (copper) and the aluminium frames are easily extracted and recycled or in some cases reused entirely [18]. In some cases, commercial recycling of PV panels could only include these steps, while discarding the remaining unframed panels or PV laminates [8].

Delamination is among the most complicated steps which decides the further recyclability of the PV panels. Since PV panels are designed to withstand environmental impacts an ingress of moisture and air, delamination of the panels is not easily achieved. Delamination of the PV panels could be achieved by mechanical, chemical or thermal processes, or a combination of them. Thermal delamination involves heating the PV panels to temperatures in the range of 550°C which decomposes the cross-linked polymers such as Ethyl Vinyl Acetate (EVA), Polyvinylidene fluoride (PVDF) or polyethylene terephthalate (PET) [16]. This separates the glass, the interconnecting metallic ribbons and the solar cells, and allows subsequent recovery of high-quality materials [34],Fiandra et al. [20,51],Fiandra et al. [21]. However, this process is energy intensive, generates toxic emissions and is expensive to implement[7,39,63],Tammaro et al. [62]. Chemical delamination of modules is achieved by immersing the modules or crushed pieces of modules in organic or inorganic chemicals which could dissolve the cross-linked polymers. Usually, they involve extremely hazardous chemicals and long process times, sometimes extending more than 10 days [8,15,39]. Researchers have combined chemical treatment with microwaves[50], ultrasonic waves [28]or supercritical carbon dioxide [38] to shorten the process durations. However, complete separation of EVA and PET/PVDF was not always achieved, and in some cases a secondary treatment such as pyrolysis or glycolysis could be necessary (X. [70]. Furthermore, disposal of such large scale of organic chemicals could be environmentally challenging.

Mechanical fragmentation techniques are the most widely reported and used approach for delamination (Keiichi [31,36]. Shredding, crushing, milling and grinding have been used for fragmentation of PV panels [48,13]. Recently delamination of end-of-life PV panels using high voltage crushing or electrohydraulic fragmentation (EHF) has gathered attention of the researchers as an excellent alternative to the existing delaminating processes for achieving higher recycling efficiency and material selectivity than conventional crushing [1,61,75,52,44]. It has been used to recycle fiber based composites earlier, showing the versatility of the process.[68,67]; 2015a). Its principle of operation includes applying a high voltage pulse to a solid immersed in a fluid to generate a shockwave. The shockwave travels through the fluid and interacts with the solids, causing fractures at the weak points between the interfaces of dissimilar materials, along with crushing of the material [1]. The material recovered after the fragmentation usually consists of fine powder containing metals and silicon from the solar cells, along with glass powder [61,75]. EHF has been shown to be better suited for recovery of metals (Al, Ag) and Si from the PV panels [45]as well as extremely energy efficient [61]. While the researchers have optimized the process parameters, they have been limited to the parameters of the process chamber. Data regarding the relationship of feed material and their impact on the process is not sufficient in the published literature.

1.2. State of the art in EOL Si PV modules recycling

EOL PV module recycling is in still a nascent stage in terms of industrial applications and is at early research stage (K [30,43,40]. The state-of-the art in EOL SI PV recycling has been reviewed by researchers [53,41,42,9,72,6,64],Yuan and Zhenming [74,55]. Most of these reviews highlight the current trends towards developing research options for effective and complete usage of EOL Si PV modules, to prevent them from being dumped into the landfills [8], which could lead to serious pollution issues [42]. In past decade, up to 2023, researchers have focused their efforts on purification and recovery of metals such as Cu and Ag and silicon from the EOL PV modules, as maximum number of publications in the same period were related to this topic [53,41,42]. Further, thermal delamination was found to be mostly used by researchers followed by chemical delamination. Almost universally, mechanical pretreatment was used to prepare the modules for the delamination process. Chemical processes for purification and extraction of the valuable materials such as Cu, Ag and Si are most reported [53,41,42,10]. This is followed by electrical, thermal and plasma methods respectively.

Policies and guidelines pertaining to recycling EOL PV modules exist in several countries (Y. [71,58,25]). However, instances of commercial scale adoption of PV module recycling are relatively low. Among the various initiatives by industries, First Solar could be considered the leader in PV recycling [58]. However, they produce Cd-Te thin film panels. Initiatives for recycling c-Si based PV modules were established in Europe by solar PV manufacturing industries; however, their long-term commercial viability has not been established [58]; Y. [71]. According to a current report by IEA PVPS on status of global PV recycling, a total of seven commercial entities are engaged in recycling PV modules out of which six have their operations in EuropeWambach et al. [65].

1.3. Scope of the present work

In this work we present the impact of the feed material on the EHF process used for delaminating the PV panels. We use two types of panels – glass-glass (GG) and Glass-backsheet (GB) panels. It is observed that both panels could be recycled using EHF process. However, GG panels were more amenable to this process and tool less time for delamination than the GB panels. Further, the quality of the delamination is also observed to be better for the GG panels for similar process parameters. We also study the impact of the size of the feed material, and the process duration on the process yield, both quantitatively and qualitatively. The feed material is prepared by cutting with high pressure water jet into square shaped pieces of assorted sizes. It is observed that smaller were more easily delaminated, and resulted in higher quality of delaminated products. Additionally, the PV panels are also mechanically crushed, and the resulting material is then used as a feed for the EHF process. This considerably improves the throughput of the process and makes the process suitable for scalability. Glass, polymer and silicon with metals could be separated using simple mechanical sieving. The materials recovered can be further processed individually to recover materials such as silicon, glass, metals, especially Ag as well as polymer. We have demonstrated a simple and effective process to recover high quality material including metals, silicon and polymer from the end-of-life c-Si PV modules.

2. Experimental details

2.1. Fabrication of solar cells and panels

The PV panels used in this work along with metallized wafers used for fabricating the panels were fabricated in the laboratory. Metallized crystalline silicon (c-Si) large area (M2) boron doped (p-type) Cz wafers were used for fabricating the panels. Fig. 1 shows the process flow used in the production of metallized Si samples used in the fabrication of the modules. The wafers were doped with Phosphorus to allow for the contact formation during the metallization process [57]. The wafers, after being coated with anti-reflection coating (ARC) using plasma enhanced chemical vapour deposition process (PECVD) were metallized with H-pattern on both front and rear sides using an inline industrial screen-printer. The samples were fired at high temperature (750℃) to allow the metal to form contacts to the Si substrate, as in actual solar cells. After fabrication each solar cells contained ≈1g of Al and ≈80 mg of Ag. The weight of the metallized wafer was measured using an inbuilt weighing scale on the screen-printer system.

Two types of panels were prepared for determining the effectiveness of the electrohydraulic delamination process in separating the components of the PV panels. Glass-Glass (GG) panels had layer of glass on both sides, with ethylene–vinyl-acetate (EVA) copolymer-based layer as the encapsulant. Glass-Backsheet (GB) panels had glass on the front and polyethylene terephthalate (PET) based backsheet on the rear, with EVA as the encapsulant. Every panel was prepared with 60 metallized samples on an industrial inline stringer (TT1800 Stringer) machine. The samples were stringed using 0.9 mm Cu ribbons to form a string of 10 samples. Six such strings were cross connected (soldered) using 5 mm tin-coated copper connector. The thickness of the glass panel used in this work was 3 mm. The panels laminated at a temperature of 150℃ with an application of 800 mBar of pressure in the industrial Burkle Multistep Laminator M−LAPV 1222–5 HKV. The total time for lamination process was ≈20 min. Similar process was used for glass-backsheet panel where the second layer of glass was replaced by the white PET based backsheet (0.275 mm). The physical properties of the fabricated modules along with the materials used in their preparation are given in Table 1. The schematic cross section of the modules are shown in Fig. 1.

Depending on the experiment design, the panels were cut into desired size using a high-pressure water jet or crushed using an industrial crusher to prepare the feed for the EHF batch process chamber. The materials were fed into the chamber according to the experimental plan and treated using EHF process. After the process was completed, the water was filtered, and the material was recovered in the retaining sieve of the equipment. The material was recovered and dried by placing under an IR lamp. After drying the material was sorted by mechanical sieving. The experimental steps are shown in the form of a flowchart in Figs. 2-4.

Table 1
Dimensions, weight and the materials used in preparing the modules used in this work.

	Glass-Glass (GG)	Glass-backsheet (GB)
Dimensions (L, W, Th) [mm]	1670, 990, 7.5	1670, 990, 4.5
Weight [Kg]	27.5 ± 0.33	15.5 ± 0.45
Bill of Material	Front glass, rear glass, stringed solar cells, copper cross connectors, EVA encapsulant	Front glass, rear PET white backsheet, stringed solar cells, copper cross connectors, EVA encapsulant

L = module length, W = module width, Th = module thickness

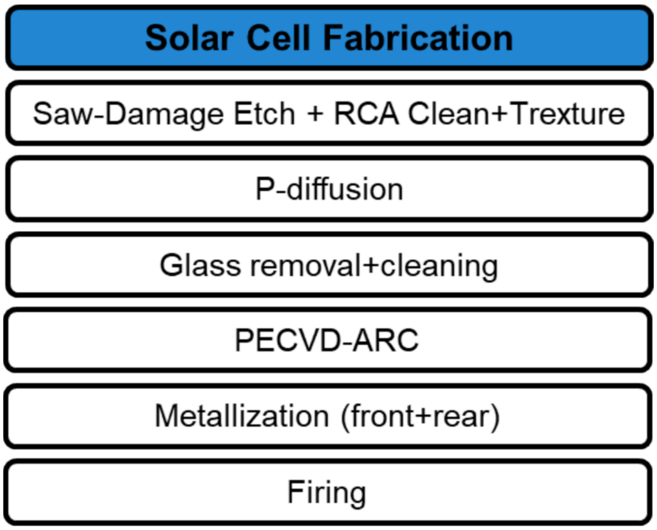

Fig. 2. Process flowchart showing the steps involved in the preparation of the solar cells.

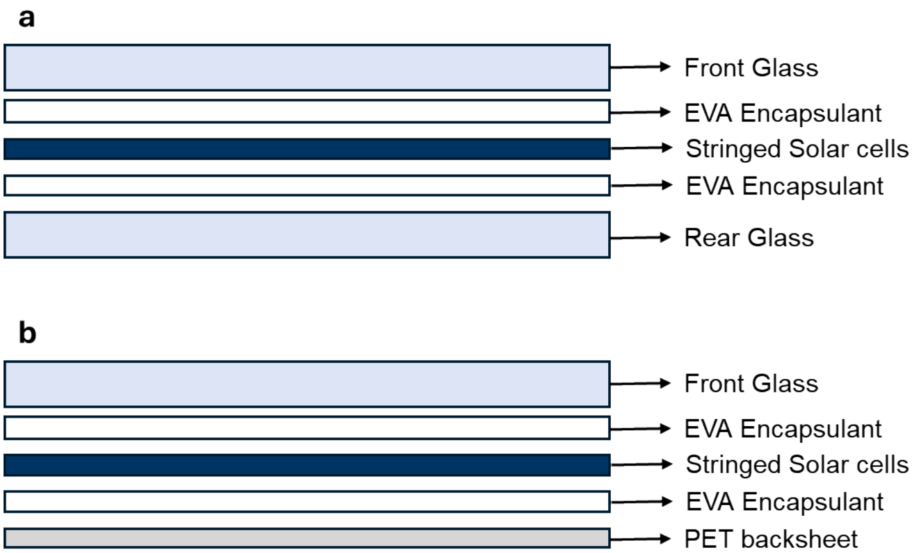

Fig. 1. The schematic cross section of the (a) Glass-Glass (GG) and (b) Glass backsheet (GB) modules prepared in the lab used in this work. The images are not to scale, and the colour scheme is used for identification of the different layers.

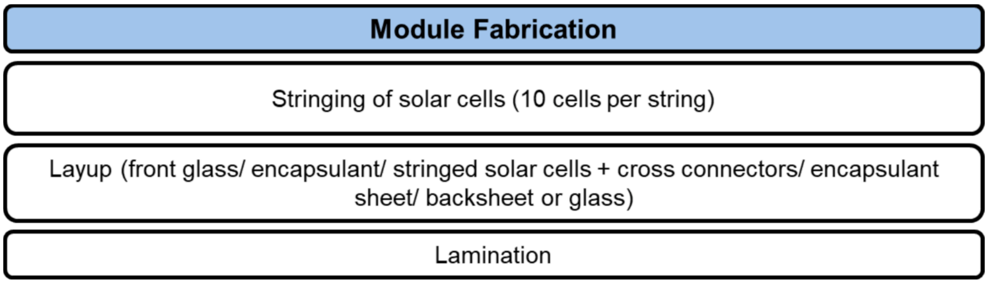

Fig. 3. Process flowchart showing the steps involved in the preparation of the PV panels.

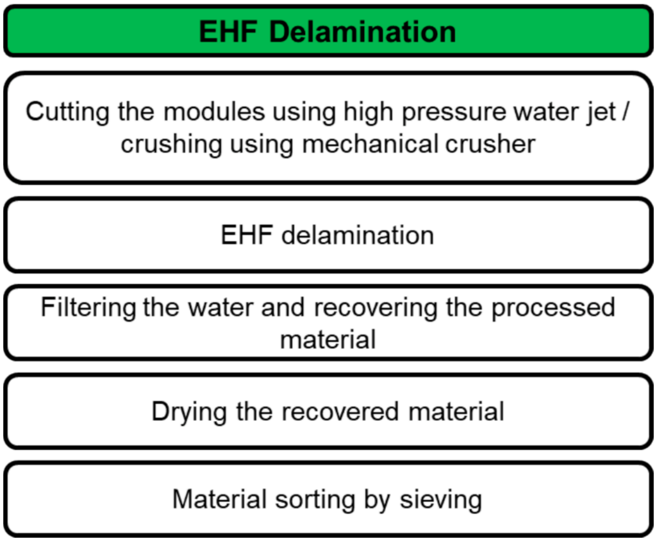

Fig. 4. Process flowchart showing the steps involved in the delamination of modules using EHF process.

2.2. Material characterization

The panel dimensions and weight were measured with the integrated system within the laminator. The material obtained after the EHF delamination was weighed on an electronic weighing scale with least count of 0.001 g (1 mg). The scale was calibrated with reference weight of 5 mg, 1 g, 10 g and 100 g after every 10 measurements. The particles were observed under an optical light microscope (Axio Imager M1m Ziess). Further characterization of the materials was also carried out with the help of a scanning electron microscope (SEM) (Auriga, Carl Zeiss) coupled with Energy Dispersive X-ray spectroscopy (EDS) (Quantax, Bruker).

3. Results and discussion

3.1. Impact of the panel type on the delamination process

The GG and GB PV panels were cut into pieces of size 2.5x2.5 cm^2 and 5x5 cm^2 using a high-pressure water jet. The samples after cutting from the water jet are shown in Fig. 5.

For each experimental run, a measured quantity of PV panel pieces was fed into the batch processing chamber. In the first experiment the process voltage input (50 kV) and duration (≈240 s) were kept constant. Earlier work published have used higher voltage in the range of 90–200 kV [1,61]. In this work, the operating voltage was kept fixed at 50 kV. The material recovered after the process was dried and manually sieved to sort the constituents based on their size. The different materials recovered after processing are shown in Fig. 6. While most of the silicon and metal present in the form of solar cells were separated from the EVA layers, a fraction of the solar cells was still found attached to the front EVA, in the glass-glass and glass-backsheet PV panels. In the case of glass-backsheet PV panels, the front EVA was separated from the solar cells while for most pieces, the rear EVA and PET backsheet remained stuck together, along with some remnants of the silicon solar cells. The front EVA separated from the front glass layer was also found to be cleaner (having less silicon particles sticking to it) than in the case of EVA + backsheet layers. Recycled glass was mainly obtained as pieces (≈1-5 mm) and coarse powder (≈ 0.25–1 mm) while finer glass, silicon and metal particles were obtained as fine powder (≈ <0.25 mm). Similar distribution of particle size has been observed in earlier work when using glass-backsheet panels [1,61,44,75]. However, results of the process with glass-glass panels have not been shown earlier.

Fig. 7 shows the pieces of the glass, coarse powder and fine powder as seen under an optical microscope. Copper ribbons and cross connectors used for interconnection were also obtained their whole form. The weight of the materials (dried) recovered from all types of PV panels exceeded 99 %, signifying the high material recovery efficiency of the process. Fig. 8 shows the relative percentage of different materials recovered after the electrohydraulic processing of the PV panels using a

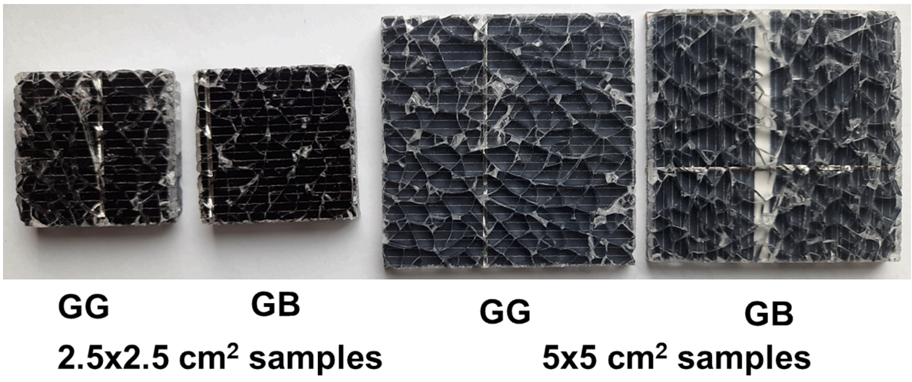

Fig. 5. GG and GB panel samples after being cut into pieces using a high-pressure water jet. The cracks on the glass visible here appeared during the cutting process.

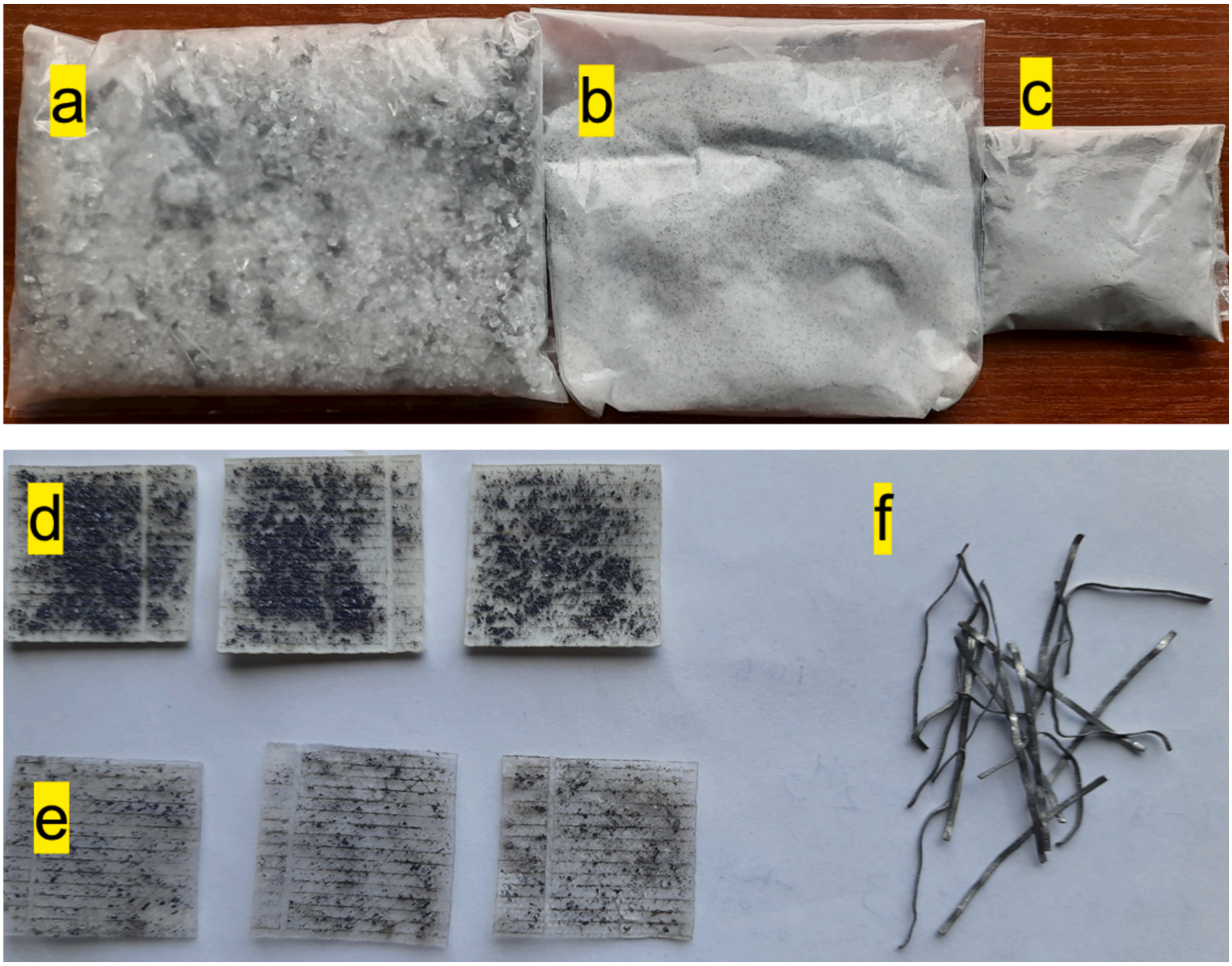

Fig. 6. The materials recovered after the electrohydraulic treatment of the PV panels a) glass pieces (1–5 mm) b) coarse powder (0.25–1 mm) c) fine powder (<0.25 mm) d) EVA + backsheet e) only EVA (from the front encapsulation layer) f) conducting strips.

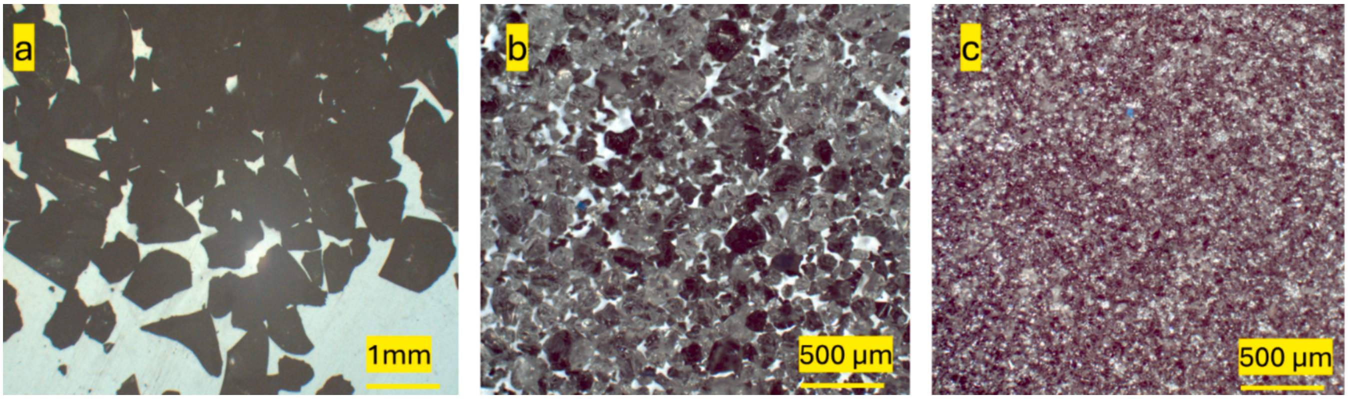

Fig. 7. Optical microscopic image of the a) glass pieces (1–5 mm) b) coarse and b) fine powders obtained after the electrohydraulic fragmentation of the glass-glass panels.

feed size of 2.5 cm x 2.5 cm^2 and processing duration of 240 s. For glass-backsheet panels, ≈20 % wt. of the polymer obtained was only EVA, while the remaining was EVA + PET. The fraction of coarse powder obtained from the glass-glass panel was slightly higher than that from the Glass-backsheet panel. This is expected as the glass-glass panel has more glass as compared to the glass-backsheet panel. Further, the polymer layers contribute a lower percentage of the wight in the glass-glass panel, which is also reflected in the materials recovered after the process. In this work, only a single crushing process has been employed. It is also possible to have a multi-stage crushing process for primary and secondary crushing [1].

From the experiments it was found that the glass –glass panels were

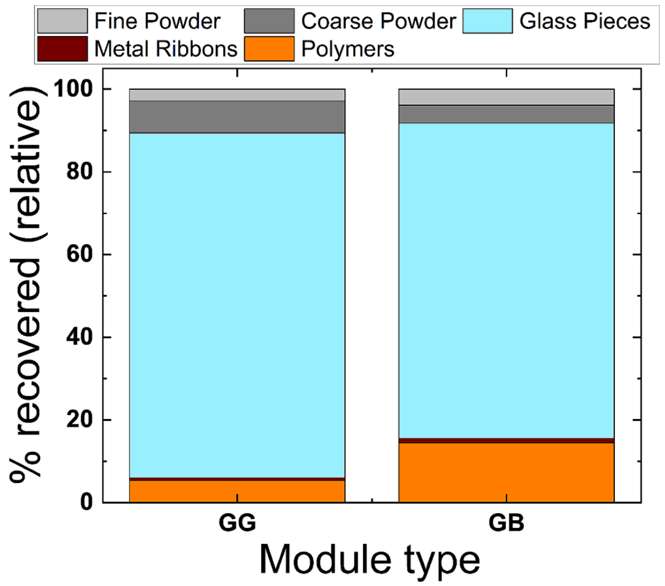

Fig. 8. Material recovered for different types of panels for a feed size of 2.5 × 2.5 cm^2 and processing time of 240 s.

easily and quickly separated and yielded better quality of recovered materials post processing. For glass-glass panels more than 90 % of the constituents were separated and recovered as single material (glass, ribbons, EVA). The pressure pulse generated in the electrohydraulic process is most effective in separating materials with different mechanical properties [1]. In the glass-glass panels, the encapsulant (EVA) is attached to the glass on both front and rear sides, while in glass-backsheet panels, EVA is attached to glass on the front and to the PET backsheet on the rear. Hence, the dissimilarity of the mechanical properties of the material at the interface is higher in allowed for the efficient separation and subsequent recovery of the EVA in the glass−glass panels during the recycling process. In glass-backsheet panels, the EVA on the front was separated and recovered as a single layer. The EVA bonded with the backsheet could not be separated and all pieces of backsheet has some EVA stuck to them. It was also observed that in glass-backsheet panels, the only EVA pieces were relatively cleaner and had less silicon pieces sticking to them than the EVA-backsheet combined pieces. This could be explained as the EVA pieces alone are lighter, they can move around the chamber more easily than the EVA + PET pieces, and hence, have higher probability of interacting with the high-pressure pulse.

3.2. Impact of the feed material size on the delamination process

To study the impact of the size of the feed material on the effectiveness of delamination using the electrohydraulic process, glass-glass panels was selected first. Due to the difference in the material properties at each interface, the glass-glass panels are ideal for electro-hydraulic delamination process. The glass-glass panels were cut in three different sizes; 2.5x2.5 cm^2, 5x5 cm^2 and 7x7 cm^2. The cut samples were fed into the batch process chamber (one size at a time) and processed for 240 s. After the process, the materials were recovered, dried and analysed. Each process run was repeated three times, so a total of 9 experiments were carried out. Table 2 shows the experimental results of the individual process runs. Immediately it can be seen that almost all experimental runs resulted in high yield exceeding 98 %. The average yield of EVA and the metal ribbons remained similar for all sizes of feed material. However, there were slight differences in the ratio of glass, coarse powder and fine powder with the change in the size of the feed material.

Fig. 9 shows the mean of relative percentage of the glass, coarse powder and fine powder for each feed size, relative to the sum of yield of glass, coarse powder and fine powder.

It was observed that as the mean relative yield of glass particles increased with the feed size. This meant when bigger pieces were fed into the chamber, a higher fraction of yield comprised glass pieces in the range of 1–5 mm. When the feed size was smaller, the glass pieces were crushed more, resulting in increased percentage of coarse and fine powders. It was also observed for smaller size of feed material, the EVA was cleaner with less silicon particles sticking to the surface. This could be due to the fact that smaller particles are easily suspended in the water and move around more easily than the heavy particles. Therefore, they have higher probability of getting impacted by the shock wave generated in the medium. Hence, from this experiment it was concluded that

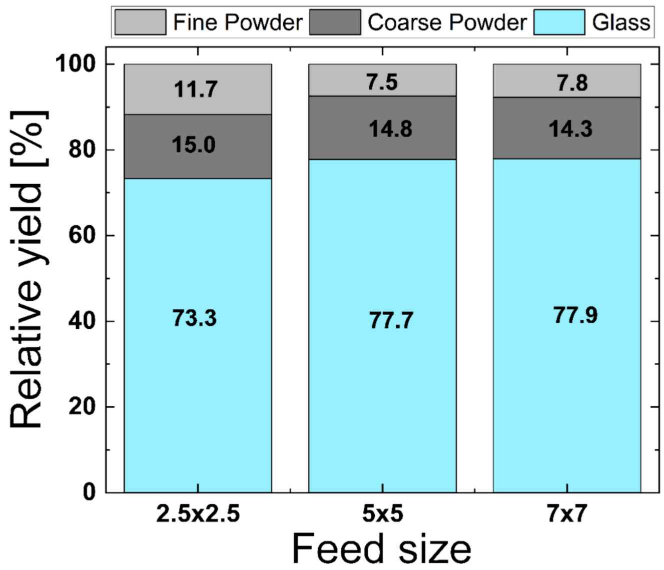

Fig. 9. Stacked column plot showing the mean relative yield in percentage for glass pieces, coarse powder and fine powder obtained for different feed sizes of glass-glass panels.

Table 2
Experimental results of the effect of the feed size on the recovery of materials using electrohydraulic fragmentation process. Only glass-glass panel was used for this experiment.

Size [cm^2]	Run #	Input wt [g]	EVA [g]	Ribbons [g]	Glass pieces [g]	Coarse Powder [g]	Fine Powder [g]	Total yield [g]	Yield % [%]
2.5x2.5	1	403.0	21.5	2.4	273.3	56.0	45.2	398.4	98.9
	2	400.8	20.7	2.9	272.6	55.8	44.4	396.4	98.9
	3	400.0	21.1	3.3	273.7	56.0	41.8	395.9	99.0
5x5	1	411.0	21.1	2.7	293.7	56.0	29.9	403.4	98.2
	2	410.9	20.8	3.4	290.4	55.3	30.6	400.5	97.5
	3	419.8	22.1	3.7	305.4	58.2	24.6	414.0	98.6
7x7	1	409.8	21.6	2.9	291.5	53.6	31.8	401.3	97.9
	2	409.7	22.3	2.2	293.5	53.8	29.8	401.6	98.0
	3	413.2	22.2	2.8	301.7	55.1	26.8	408.6	98.9

smaller size feed material leads to efficient delamination between the panels.

The experiment was repeated with the glass-backsheet panels. It was observed that while pieces up to 5x5 cm^2 were completely separated, all pieces of 7x7 cm^2 were not completely delaminated and for some pieces glass pieces were still sticking to the EVA + PET layer. Furthermore, the EVA and EVA + PET pieces obtained had significantly more silicon particles (from the solar cells sticking to them). This could be explained as due to the higher amount of tough flexible polymer, the large pieces of the glass-backsheet panels could withstand the pressure pulse, without having the same impact as that in the pieces of brittle glass-glass panels. Insufficient processing time could also be a reason, implying longer processing durations for glass-backsheet panels to achieve similar results.

3.3. *Impact of the process duration on the delamination process*

To test the impact of the process duration on the effectiveness of delamination of the PV panels, glass-backsheet panels were used. The panels were cut in two sizes, 2.5x2.5 cm^2 and 5x5 cm^2 using high pressure water jet. Similar operating times of 300–500 s have been reported earlier [44]. Three processing durations were experimented in this work – 240 s, 420 s and 600 s. Table 3 shows the results obtained from the experiments.

The relative proportion of the EVA and EVA + PET recovered in % wt. is shown in Fig. 10. For this plot, the proportion of EVA pieces and EVA + PET pieces are expressed as a fraction of the sum of both. Hence, % (wt.) EVA + % (wt.) of EVA + PET is always 100. A similar plot is shown in Fig. 11, which charts the relative proportions of glass pieces, coarse powder and fine powder for different processing time and feed size.

Here two clear observations could be made. First, the relative amount of EVA recovered increases with the process duration for both feed sizes. However, the increase in the proportion of EVA recovered is more significant in case of larger feed size. Whren the feed size is 2.5x2.5 cm^2, the proportion of polymer recovered as only EVA increases from 6.9 % for 240 s process duration to 7.6 % for 600 s process duration. The corresponding values when the feed size is changed to 5x5 cm^2 increases from 2.4 % for 240 s process duration to 7.8 % for 600 s process duration. Hence, smaller feed size can achieve optimal delamination quicker. This can be explained as the smaller pieces are expected to move around the chamber more easily and have higher probability of getting impacted by the shockwaves and getting separated.

Fig. 7 shows the relative wt. percentages of glass pieces, fine powder and coarse powder obtained for different process duration and different feed sizes. In this case, for both sizes of feed, the proportion of glass particles decreased while those of coarse and fine powder increased with the increased process duration. For longer process duration more shock waves impact the material, hence breaking the larger pieces into smaller pieces. The increase in the relative proportions of coarse and fine powder could mainly result from the increased fraction of glass particles. However, since the polymer pieces were cleaner for longer process durations, increased amount of Si and metal in the fine powder could also be expected, albeit their proportional contribution would be miniscule compared to that from the increased glass particles.

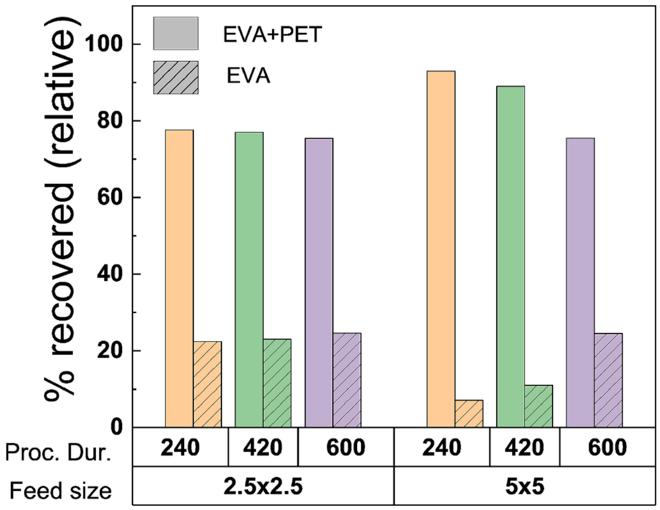

Fig. 10. Column plot showing the relative % (wt.) of EVA + PET and EVA layer recovered after the electrohydraulic treatment of the glass-backsheet panels.

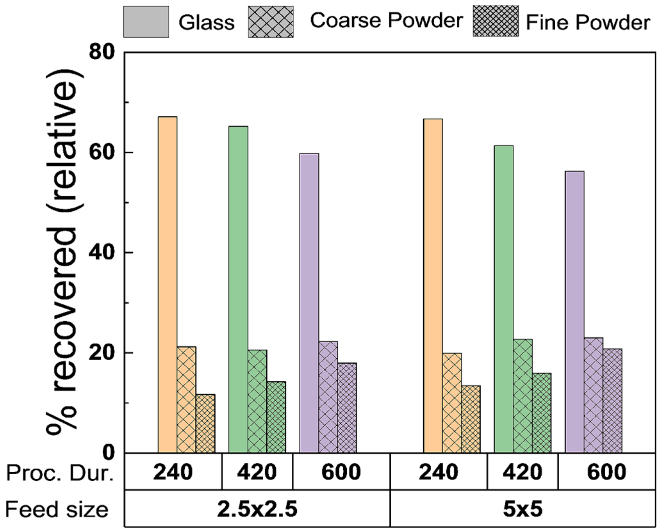

Fig. 11. Column plot showing the relative % (wt.) of glass pieces, coarse and fine powders recovered after the electrohydraulic treatment of the glass-backsheet panels.

Similar experiments were carried out with the glass-glass panels as well. The results were similar to those obtained with glass-backsheet panels. The EVA pieces were cleaner for the longer process duration for both smaller and larger feed size. Like glass-backsheet panels, the increase in the quality of recovered EVA and the increase in the proportion of fine powder in the recovered material changed more significantly for the larger feed size (5x5 cm^2) than for the smaller feed size (2.5x2.5 cm^2). The proportion of coarse and fine powder recovered increased with increased process duration at the expense of proportion

Table 3
Experimental results of effect of process duration on the material recovery for glass-backsheet panels using electrohydraulic delamination process.

Size [cm^2]	t [s]	Input wt [g]	EVA + PET [g]	EVA [g]	Ribbons [g]	Glass pieces [g]	Coarse Powder [g]	Fine Powder [g]	Total yield [g]	Yield % [%]
2.5x2.5	240	236.8	23.9	6.9	2.9	135.4	42.7	23.6	235.4	99.4
	420	238.4	23.8	7.1	3.1	130.5	41.2	28.5	234.2	98.2
	600	233.8	23.3	7.6	2.3	119.1	44.3	35.8	232.4	99.4
5x5	240	238.2	31.6	2.4	2.6	133.7	39.9	26.9	237.1	99.5
	420	239.8	28.4	3.5	3.0	122.9	45.4	31.9	235.1	98.0
	600	237.8	24.0	7.8	3.5	113.1	44.6	40.3	233.3	98.1

of glass pieces.

3.4. Improving the process throughput using mechanical crushing as pretreatment

With the help of experiments, it was established that the smaller feed particles resulted in higher throughput and quality of the electrohydraulic delamination process. However, cutting from water jet is a time intensive process and reduces the throughput considerably, as the feed size is reduced. To improve the throughput, a process which reduces the feed size of the panels in short duration was needed. This could be done with the help of mechanical crushing of the panels. In this work a rotary shaft crusher was used to crush the panels into smaller size. Previous works have compared the mechanical crushing with the EHF process to demonstrate the superiority of the EHF process in crushing the panels [44,61]. However, in this work we shave shown that both processes could be used together to improve the process throughput. After mechanical crushing the panels, the collected material was sieved manually and sorted according to the size of the particles into three size groups. Pieces larger than 5 mm were laminated foils with or without glass and pieces of solar cells sticking to them. Pieces of sizes ranging from 1 mm-5 mm mostly comprised glass cullets while a mixture of glass and solar cell powder was recovered as particles with size $<$ 1 mm. Due to the rotary shaft force, the glass cullets were knocked off from the panels during the crushing stage. The laminated pieces (with or without glass) with size exceeding 5 mm, required shock wave fragmentation for further separation into individual components. In contrast, the other two categories comprised mainly glass cullets and glass + solar cell powder, as ascertained by optical microscope. Since the pieces ranging in size from 1-5 mm and the powder with particles $<$ 1 mm particles are already obtained in a form similar to that obtained by electrohydraulic delamination process, they didn't need to be processed again. Hence, only pieces of size $\geq$ 5 mm were used as feed material for the electrohydraulic delamination process. Thus, the advantage of crushing the panels prior to the electrohydraulic treatment is that it reduces the feed material requiring separation, increasing the throughput. The pieces ($\geq$ 5 mm) were introduced in the electrohydraulic delamination process chamber and processed for 600 s. The material recovered after the processing were similar to earlier experiments and comprised separated polymer pieces, glass cullets (1–5 mm), coarse powder (0.25–1 mm) and fine powder ($<$0.25 mm). The process steps for the increased throughput process incorporating mechanical crushing process are shown in Fig. 12.

While crushing the panels could increase the overall process throughput, it has some disadvantages as well. When the panels were cut using water jet, the pieces obtained as feed material were of uniform size. Hence, the polymer pieces and connecting ribbons recovered after the process were also of uniform sizes. This resulted in easier separation of the material using a sieve. When the panels were crushed, the size of the pieces in the feed material was not uniform. For example, the feed material after sorting contained pieces ranging in size from 3.5 mm to 15 mm. This made the sorting of materials after the electrohydraulic delamination processing more difficult. For example, when the panels were crushed, more polymer pieces were obtained in the glass pieces (1–5 mm). While these can be separated by using a gravitational/mass-based separation process, it adds another process step making the entire process more complicated and could impact the gain in throughput obtained from crushing the panels.

4. Characterization of the coarse and fine powder recovered after electrohydraulic delamination process

The coarse and the fine powders obtained after the electrohydraulic delamination process were further characterised by using scanning electron microscopy (SEM) and energy dispersive x-ray spectroscopy (EDS). For the characterization, coarse and fine powders obtained from the process where water jet was used for preparing the feed material were used. Coarse powder and fine powders from glass-glass and glass-backsheet panels were respectively mixed in the ratio 1:1 by mass. This provides another advantage of the recycling process using electrohydraulic delamination as the output stream from different panel types could be mixed. For characterization, the powders were mounted on adhesive conductive carbon tape. Fig. 13 shows the elemental maps of the coarse powder. The coarse particles were mainly composed of glass particles as Si, O, Na and Mg were observed in the same regions. The carbon seen in the image corresponds to the carbon tape used for mounting the powder. Na and Mg are commonly added to glass used in

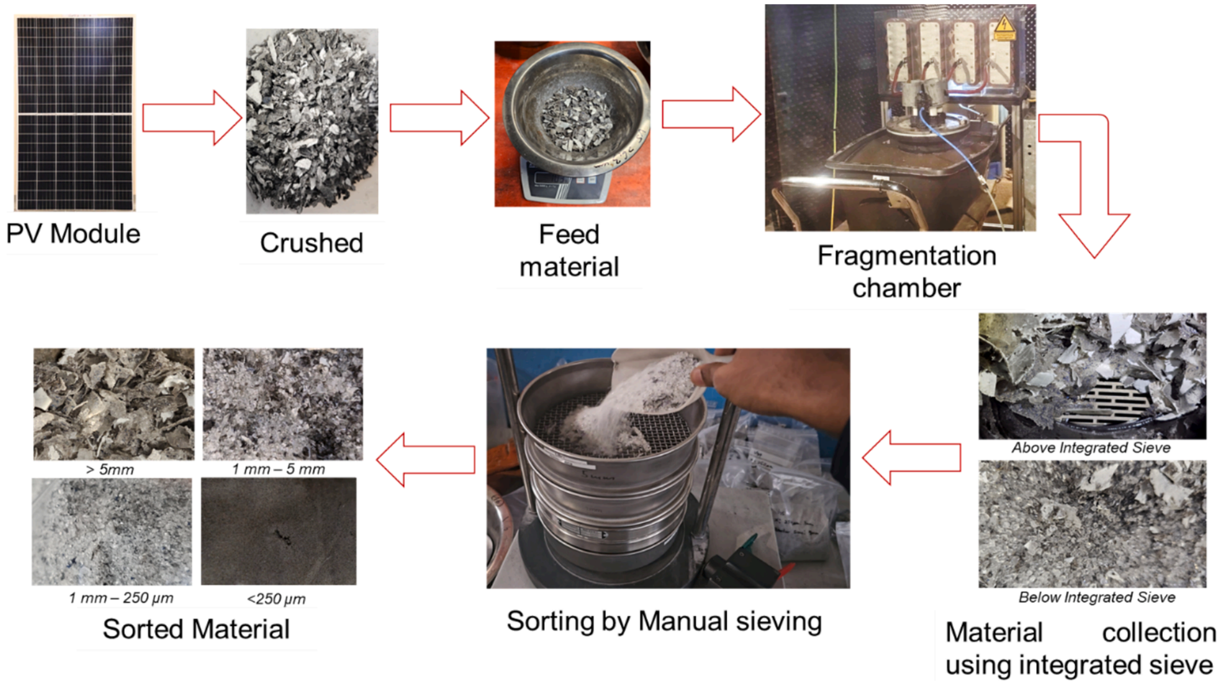

Fig. 12. Process steps of the modified process for increased throughput using mechanical crushing of the panels before electrohydraulic processing.

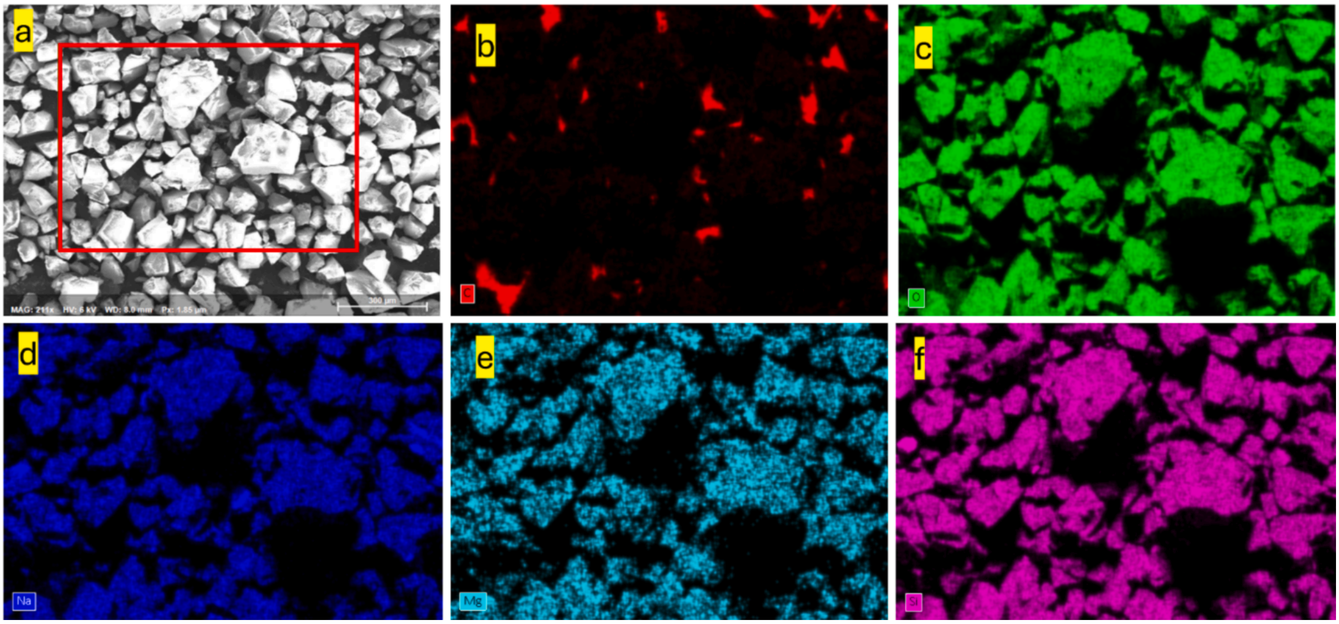

Fig. 13. Elemental maps of coarse powder obtained by EDS showing (a) the area of analysis (b) carbon (c) Oxygen (d) Na (e) Mg (f) Si.

manufacturing PV panels [1].

The presence of metal and Si pieces from the solar cells were confirmed in the fine powder by EDS analysis. As shown in Fig. 14b and d, the brightly lit areas of Si correspond to the c-Si pieces from the solar cells, as O is not prominent at those places in the corresponding Si−oxygen combined map in Fig. 14-d. Al agglomerates were also confirmed to be present in the fine powder as shown in Fig. 14-b and c. The presence of Ag was difficult to ascertain due to the fine particle size and small amount. However, certain agglomerated particles of Ag were identified due to the strong signal associated with them. One such

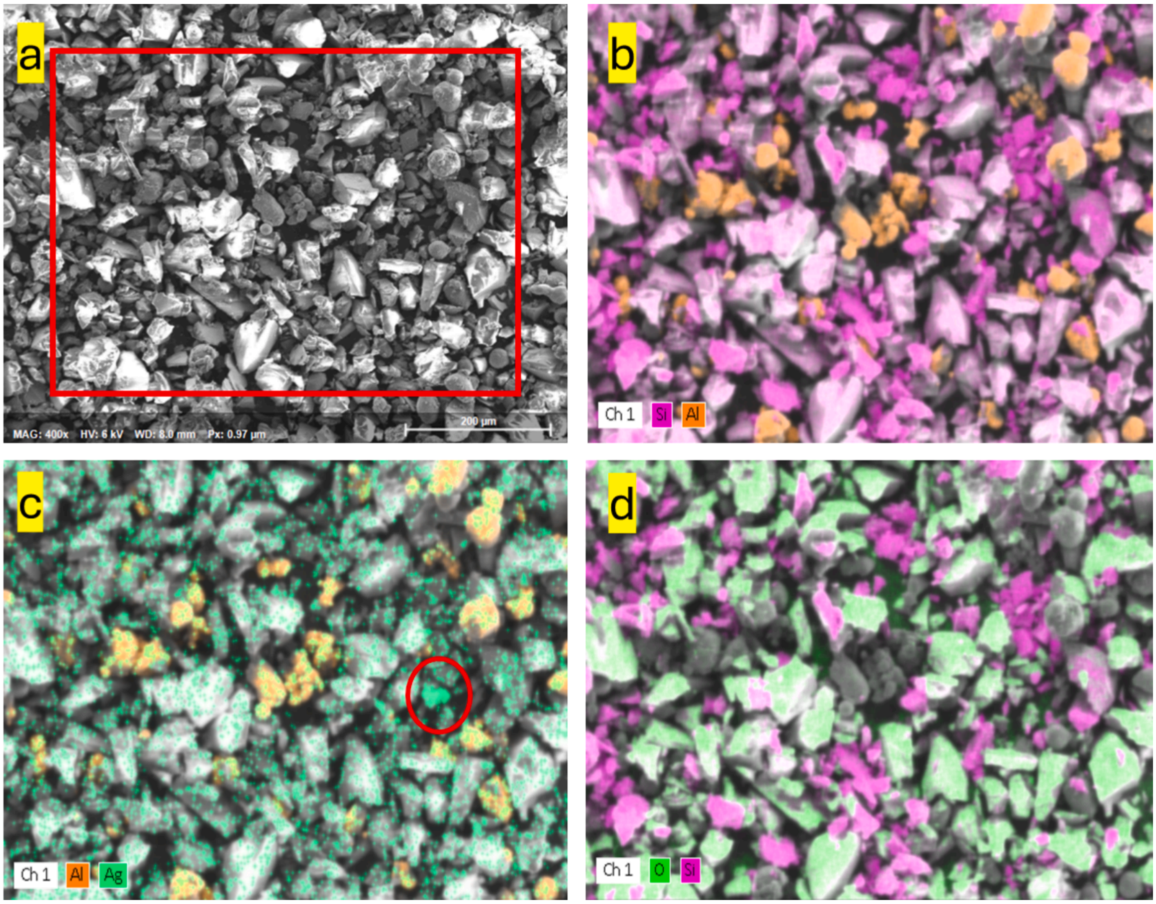

Fig. 14. Elemental maps of coarse powder obtained by EDS showing (a) the area of analysis (b) Si&Al (c) Al&Ag (d) O&Si.

agglomerated Ag particle in highlighted within a circular region in Fig. 14-c. Hence, this proves the capability of the electrohydraulic separation process to separate glass and metal + silicon particles by employing a simple mechanical sieving process.

5. Separation of individual materials and their recovery

Delamination of the modules is usually followed by purification steps to recover the raw materials in a form that could be reinserted, ideally in the supply chain to manufacture new PV modules. In the context of this work, the materials extractable after the EHF treatment include Ag, Si, glass and polymers. Recovery of each of these materials requires extensive experimental set-up which is beyond the scope of this work and could be a future research direction. However, here we provide most probable methods under consideration to extract and purify the material mix obtained after the EFH processing of c-Si PV modules.

The glass pieces obtained after the EHF processing might have some polymer and Si pieces which could be undesirable. While the polymer pieces could be easily removed by washing with water, removal of Si particles could be difficult. However, the percentage of Si in the glass cullets is estimated to be less than 0.001 % by wt. To remove Si from these glass cullets, they can be treated with sodium hypochlorite (NaOCl) or potassium hydroxide (KOH), both of which would preferentially react with Si while leaving the glass particles intact [3,14,69]. For fine silicon and glass powder the material targeted for extraction would depend on the composition of the powder. In the context of this work, the fine powder was found to contain more Si than glass. To retrieve Si, the glass needs to be dissolved which can be done by treating the powder with hydrofluoric acid (HF). The HF would preferentially dissolve glass, while not etching the Si appreciably. Another advantage of this process is the removal of the ARC coating on the solar cells [4,27], Knotter and Dee Denteneer [29]. Finally, the fine powder mixture of Si, Ag, Al and some glass is among the most valuable material to be recovered, especially due to the presence of Ag in this mixture. The Ag in the mixture can be recovered by treating it with Nitric Acid (HNO_3), which will dissolve the silver. The dissolved Ag can be recovered from the acid solution by treating it with other chemicals or using electrolysis (de [46,12,73,35]. Further, the remaining glass and Si can be separated as mentioned earlier by dissolving the glass in HF.

One of the advantages of EHF recycling is that the polymer used in the PV module could be recycled as well. The polymer pieces were delaminated from the module due to EHF treatment but still contained some Si sticking to the surface. While the amount of Si sticking to the surface could be minimized by a careful optimization of the process parameters, treating the polymer pieces with KOH and HNO_3 would further dissolve any remaining silicon and metal pieces sticking to the surface. The cleaned polymer pieces could then be recycled using the specialized process developed for recycling them [32,60].

6. Strengths and limitations of the proposed method

EHF defragmentation of the EOL c-Si PV modules provides a way to effectively delaminate the PV modules, without the use of thermal or chemical decomposition. The electrohydraulic delamination process for recycling c-Si PV panels provides the easiest and quickest method of delaminating c-Si PV panels, leading to near total material recovery of different components of the PV panel. This agrees with the current standards of EU WEEE directive [7,47]; European Standardization Organisation [19,59]. This delamination process enables recovery of all the components of the PV module. The process is adaptable and can be tailored to suit different module types and can be combined different pretreatment processes such as mechanical crushing to further improve the throughput. The EHF process requires only water (which need not be potable) and electricity to operate. It does not create any hazardous gasses or toxic effluents/wastes. EHF process is scalable and can be integrated with the existing module recycling facilities to improve the material recovery.

The material revered after the EHF process can be segregated by simple mechanical filtering. Although the EHF process appears to be and clean and quick process of delaminating PV modules, the separation of the individual components for recovery of high value materials may need some post processing treatments to recover pure material for closed loop recycling. While interconnectors and ribbons are easily recovered and separated as whole pieces, the silicon glass and metal are crushed and intermixed. In our experiments, it was easy to separate polymer pieces from the glass and silicon-metal powder using simple sieve filtration. However, inclusion of some fine polymer fibres (< 1 mm in diameter), especially in the fraction of glass cullets could not be ruled out. It was possible to separate the large glass pieces from the finer particles using simple filtration, which could be easily recycled. However, the coarse glass particles included some pieces of Si solar cells. Similarly, the fine powder, while mostly composed of Si and metal powder, contained some fine glass powder too. One way to separate the different materials could be density based medium separation [1]. Further research efforts would be focussed on optimization of process parameters to minimize the mixing of the individual components to facilitate their separation and recovery using simple mechanical sieving process. Additionally, efforts would be made to develop processes for efficient recovery and purification of high-quality raw materials using EFH process for delaminating c-Si solar PV panels.

7. Conclusion

Development of processes enabling complete recycling of silicon PV panels is essential to improve the sustainability of silicon PV panels. In this work we have presented the electrohydraulic fragmentation process as an alternative to the popular thermal process of delamination of c-Si PV panels. We have demonstrated the compatibility of the EHF process with different panel types. While similar experiments with GB panels have been reported earlier, we have reported the results with the GG panels for the first time, to the best of our belief. The electrohydraulic fragmentation process was most effective for glass-glass panels, since the EHF process relies on the difference in the mechanical properties of different layers to respond to the propagation of shock waves. As the difference in the mechanical properties of the intermediate layers is larger in the case of GG panels as compared to GB panels, similar process parameters facilitated more effective separation for GG panels. For GB panels, while the EVA layer sticking to the front glass was easily separated, the rear EVA + PET layer could not be separated easily. For glass-glass panels, even larger pieces of the panel could be separated with ease. This emphasizes the fact that different types of PV panels may have different response to the same EHF process. Hence, for commercial integration, similar panel types could be grouped together for improved process efficiency. One of the main advantages of the EHF process lies in the recovery of the polymer without generating any toxic effluents or hazardous gases. With proper process optimization is was possible to separate the polymer pieces from the glass and the silicon solar cells. The polymer pieces recovered were clean with lower amount of silicon sticking to their surface, which implies more critical material recovery in the terms of Si and the precious metals. Two different pretreatment methods were used to prepare the feed material – mechanical crushing and water jet cutting. Cutting the panels into regular sized pieces for processing using a high-pressure water jet, albeit slower than crushing, provides for the cleanest method of preparing the feed material. Furthermore, regular and uniform size of feed material could be treated more efficiently and simplify the sorting process post processing. To test the integration of the EHF process with the already existing crushing process, the panels were crushed as a pretreatment process, rather than being cut by water jet. Due to crushing and following separation processes, some of the metal, glass pieces and powder (glass + silicon + metal) were already separated and only the non-separated, laminated pieces were fed into the electrohydraulic chamber. While crushing could improve the throughput of the overall process, it could also deteriorate the quality of the separation of material obtained after processing. The non-uniformity of the feed material made it difficult to separate the materials such as glass pieces and polymer with the help of simple mechanical sieving, and hence longer and complex separating process may be needed. This could counteract the gain in process throughput obtained earlier. Nevertheless, the combination of the various pretreatment process provides the flexibility of designing the process to suit different end results, for example, amount of material to handle, energy costs, labour costs and other commercially important factors. Development of a pilot line using such process might be needed to direct the further research. With the longer process durations, the proportion of fine powder in the final recovered material increased at the expense of glass pieces, due to more interaction with the pressure waves. It was also observed that the smaller pieces in the feed material were separated more effectively. This could be related to their increased mobility in the chamber and hence, more interaction with the pressure waves.

Through this work we have demonstrated the flexibility of the electrohydraulic delamination process in the processing of the end-of-life c-Si PV panels. The EHF process is a clean, environment friendly process which does not generate any toxic effluents or gases during its operation. We have shown that the process can be carried out efficiently at lower voltages (50KV) than reported earlier. EHF process can be used for different panel types and is compatible with different pre-processing options such as cutting or crushing. However, different panel types should be processed separately to maintain the uniformity of the process. Further the process can be customised for high throughput or high-quality process. Furthermore, we have shown that recycling end-of-life PV panels using electrohydraulic fragmentation can lead to a high yield and high quality of materials enabling almost complete recovery of components from end-of-life crystalline silicon PV panels. We demonstrated that it is possible to recover and separate clean polymer pieces with process optimization. While most of the materials could be segregated with simple mechanical sieving, intermixing of the materials could be completely avoided. Future research could further optimize the process, for example, by including a multistage processing sequence involving continuous separation of the materials, investigation of the methods of separation of materials leveraging the differences in their densities, and recovering and recycling high-quality silicon, metals and polymers revered in this process.

CRediT authorship contribution statement

Pradeep Padhamnath: Writing – original draft, Visualization, Validation, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization. **Srinath Nalluri:** Writing – original draft, Visualization, Validation, Methodology, Investigation, Formal analysis, Data curation. **Filip Kuśmierczyk:** Validation, Methodology, Investigation, Formal analysis. **Mateusz Kopyściański:** Methodology, Investigation, Formal analysis. **Joanna Karbowniczek:** Visualization, Methodology, Investigation, Formal analysis. **Leow Shin Woei:** Validation, Supervision, Resources, Project administration, Methodology, Investigation, Conceptualization. **Thomas Reindl:** Writing – review & editing, Supervision, Resources, Project administration, Funding acquisition.

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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