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High yield, low cost, environmentally friendly process to recycle silicon solar panels: Technical, economic and environmental feasibility assessment

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ABSTRACT

The recycling of silicon photovoltaic modules is technically viable, but often not feasible economically due to reasons that vary from high processing cost to low waste volumes that do not justify investment cost. In this study, a novel, simple, cost-effective and environmentally friendly processing method is proposed. The process consists of module deframing, laminate shredding and material concentration using electrostatic separation. The latter outputs two fractions: a valuable mixture of silver, copper, aluminum and silicon, and a mixture of mostly glass, silicon and polymers. The valuable mixture accounts for only 2-3 wt% of the total module, which can be forwarded to the downstream industry for further refinement. This paper evaluates the technical aspects of the process (quantifies material separation, energy and time) while using life cycle assessment and life cycle costing to evaluate the environmental impacts and economic prospects, respectively. The results are compared to a full recovery alternative (FRELP) and to landfilling. Environmentally, a full recovery is preferred, followed by the proposed process, both of which have a net positive impact and are better than landfilling the whole module. Economic assessment shows the process has potential to be more profitable than FRELP i) at lower waste volumes (smaller than 4 kt/y), because of the smaller equipment capital cost, ii) if there is no market for the recovered glass, which is currently the case in many locations or iii) when the end-processing industry is located afar, since only the valuable mixture would require shipping.

1. Introduction

Global installed photovoltaics (PV) is expected to rise 11-fold in the next 30 years; coupled to this rise in infrastructure is the increase of PV waste, which is expected to reach 78 million tons by 2050 [1]. Crystalline silicon cells (c-Si) are the dominating technology with approximately 95% market share; up from 80 to 90% in 2010–2015 [2,3]. PV modules typically have a 25–30 year lifespan [4], so crystalline silicon modules will make up the majority of PV module waste in the foreseable future, and are therefore the main focus of recycling developmentsc. Waste PV volumes could increase faster than expected since the initiation of PV end-of-life is contingent on the module's performance and there is a probability for early loss to occur due to infant, mid-life or wear-out failures [4]. If modules are defective or significantly

degraded and repair is not feasible then end-of-life may occur many years prior to the module's expected lifetime [5]. This increasing PV waste stream raises the question of how to best manage it, recycling being an alternative to landfill disposal.

Benefits of PV recycling arise from the reuse potential of recovered materials, which can offset the economic costs and environmental impacts of raw material production [6]. Crystalline silicon panels contain valuable metals such as aluminum, copper and silver, which have finite reserves that may become depleted in the future [7]. It is well established that the recycling of PV panels generates environmental benefits, including avoidance of global warming potential and ecotoxicity impacts, and water and energy savings [8]. Through an energy-based case study, Corcelli et al. [9] conclude that recycling waste PV is the better alternative for waste management, and that economic motivation cannot be the exclusive motivation for PV waste treatment. Indeed, PV

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List of abbreviations

c-Si Crystalline silicon

ECF Electrostatic conductive fraction ENCF Electrostatic non-conductive fraction

EVA Ethylene-vinyl acetate

FRELP Full Recovery End of Life Photovoltaic ICP-OES Inductively coupled plasma optical emission

spectroscopy

j-box Junction box LCA Life cycle assessment LCC Life cycle costing PV Photovoltaic

RQPA Rietveld Quantitative Phase Analysis

XRD X-ray diffraction

recycling can avoid the emission of about 0.8–1.2 tons of carbon dioxide equivalent emissions for every ton recycled.

Environmental benefit on its own, however, may not be enough to ensure that recycling occurs, since a high financial cost will be a deterrent to consumers and industry. The PV recycling process generally comes at a short term economic loss, and estimated discounted payback periods of initial investment in dedicated PV recycling facilities are always greater than 10 years [10]. There is a need to simultaneously improve the economic viability, practicality, recovery rate and environmental performance of the PV industry regarding the recycling of its products [1]. A recent review study suggested that more complex recycling methods that recover higher value materials could be more financially attractive than simple crushed glass recycling depending on the anticipated selling price of these materials and reduced processing cost from around \$700 to \$450 per tonne for a process that recovers glass sheets, silver, copper and silicon powder and from around \$950 to \$650 per tonne for a process that additionally extracts high purity silicon [11]. From the financial perspective, recycling presents significant revenue opportunities - and predictions show that materials recovered from PV recycling could cumulatively yield revenues of 450 million US dollars by 2030 [4]. However, economic feasibility studies indicate that current costs of recycling often outweigh material revenues and facilities often have long payback times or don't reach profitability [10,11]. Increases in recycling schemes and process efficiency, as well as facility waste throughputs, will likely improve the cost-effectiveness of PV recycling [12].

There are several approaches to PV recycling, with complex processes generally being more expensive but able to achieve precise separation and recover higher quality materials [11]. Initially, the frame and junction box (j-box) are generally mechanically separated [13]. In crystalline modules, solar cells are sandwiched between layers of an ethylene-vinyl acetate (EVA) encapsulant which adheres the cells to the front glass and polymer backsheet. These layers can be separated using thermal or chemical techniques which target the EVA [14]. Thermal treatments involve decomposing the EVA layer at high temperatures of approximately 500 °C using either pyrolysis or combustion. This allows for the separation of glass and solar cells, which, if intact, can potentially be reused in new modules [11,15,16]. However, achieving the high temperatures required for thermal separation can be energy-intensive and mechanical pressure from decomposing gases can cause cells to crack [13]. Separation of the front glass, solar cell and backsheet can also be achieved using chemical solvents to dissolve the encapsulant. Once separated the solar cells can be treated with specific acids or hydroxides to individually remove internal metals such as copper and silver [17,18]. Chemical recycling techniques have the potential to separate high-quality valuable materials. However, they often require the use of toxic chemicals, which, after being used once, must be

appropriately disposed of [13].

The European "Full Recovery End of Life Photovoltaic" (FRELP) project is a targeted recovery process for crystalline modules, able to achieve high-quality material yields using a multi-step approach. Initially, a robotic system separates the aluminum frame, j-box and cabling, which are sent to secondary facilities for dedicated recycling. What is left is the PV laminate, or sandwich, which contains EVA layers, solar cells, glass and other polymers. Glass is separated from the laminate using a high-frequency cutting knife at an elevated temperature. Optical separation is then used to separate glass into similarly sized pieces and remove contaminants. The remaining laminate is cut into small pieces and incinerated to produce energy and ash containing silicon and various metals. The ash is then sieved to separate aluminum connectors originally contained in the laminate. Acid leaching is used to dissolve metals and leave a residue that can be filtered to recover the silicon fraction. Electrolysis is then employed, which yields copper and silver from the metallic oxides within the remaining solution [19-21]. The FRELP process allows for almost complete recovery of material. Over 95% of the glass, aluminum, silver and silicon are recovered from crystalline modules [22]. However, economic analysis of this process showed that throughputs of approximately 7000 tonnes/year are required to maintain profitability [23]. Although recycling technologies are likely to become cheaper in the future [24], reductions in the quantities of valuable materials (such as silver and silicon) used in newer modules [25] may pose an economic challenge for targeted recycling processes such as FRELP [26,27].

Mechanical approaches to recycling waste PV are generally of lower cost and more environmentally friendly when compared to thermal and chemical methods [14]. Recently, high voltage fragmentation was tested to recycle waste PV. The process was able to concentrate metals due to different particle size after the dismantling (crushing) of the module through discharges [28]. Another possibility is to use mechanical shredding followed by sieving, which is also capable of concentrating metals [29]. Mechanical shredding can potentially be used in existing glass recycling plants, providing capital savings on equipment [30]. Mechanical PV recycling yields copper, aluminum and glass cullet amounting to approximately 41, 74 and 92% by mass of these available materials respectively [22]. Recovered glass cullet is often contaminated and valuable materials such as silver and silicon are not recycled [31].

Revenues from PV recycling are dependent on the quality of recovered materials as well as context-specific market demand. This revenue must be balanced against the cost of separating and recovering the materials. Generally, the more complex (and therefore more expensive) thermal and chemical processes allow for the recovery of high-quality glass as well as metallic copper and silver [19]. Prior to the implementation of commercial-scale recycling facilities, it is important to investigate markets for recovered materials and evaluate trade-offs between potential revenues and the economic costs of the recycling process [26]. A Chinese study forecasts that PV components recycling will peak in about two decades in the country, and that the current barriers to increasing the volume of recycled PV include insufficient recycling facilities, supporting policies, and lack of public focus [32].

Improvements in the economic viability, practicality, recovery rate and environmental performance of the PV industry with respect to recycling its products are indispensable. Investigations surrounding the economic costs and environmental impacts of PV end-of-life have often focused on comparisons between simple (e.g. mechanical) processes and more involved complex processes (e.g. FRELP) [6,20].

In the current study, an electrostatic separation process is proposed with the goal of offering an intermediate option that separates glass from a valuable concentrated material mix, which can be further separated via established thermal and chemical methods [33]. Glass makes up between 69 and 75% of the mass of crystalline modules [14,22] and its initial separation can potentially reduce costs and impacts associated with transportation to dedicated recycling facilities able to recover valuable materials. An electrostatic recycling process may also provide a

feasible alternative to more complex processes such as FRELP, especially for countries experiencing PV capacity growth that have yet to implement dedicated PV recycling infrastructure.

Previous research has often examined PV recycling effectiveness by employing either financial or environmental metrics individually [6,10, 24]. The current study simultaneously evaluates both economic costs and environmental impacts, allowing for trade-offs between sustainability and affordability to be considered. A range of scenarios are investigated and are relevant to future PV recycling industry possibilities. High volume and automation of equipment scenarios are investigated to reflect future PV waste projections and economies of scale. Revenues and markets for recovered materials such as glass and the concentrated material mix are likely to vary under different conditions and contexts [26,34]. Previous recycling research has often defined a single generalized scenario for variables such as process automation and waste throughput as well as specific material recovery and revenue [35]. The current study proposes an innovative new process and reflects the transitional nature of the PV recycling industry, exploring how these variables affect the economic and environmental performance of end-of-life options.

2. Material and methods

The current study is based on a previous research [33] that showed electrostatic separation can be effective as a separation step in recycling end-of-life modules and that proposed a methodology to assess the material separation effectiveness of such process. The current study further develops the electrostatic separation process, and evaluates its quality using the same assessment methods. The previously reported process separated waste PV into three fractions. Here the process is simplified to separate into two outputs: a valuable mixture containing

the metals and silicon and a non-valuable mixture containing glass and polymers. Measurements of energy, throughput and labor were taken during processing, from which life cycle assessment (LCA) and life cycle costing (LCC) are used to evaluate the environmental impact and financial cost of the proposed process, respectively. Both LCA and LCC are completed in comparison to an already established recycling process (FRELP, explained in section 1. Introduction) and to the alternative of landfilling the waste PV modules. Fig. 1 shows a summary of the proposed process and the boundaries considered. Any recycling activities outside of the boundaries of this study would be undertaken by a third-party downstream recycler/refiner.

2.1. Experimental

The experimental section presented hereafter describes the processes used to separate and recycle five crystalline silicon photovoltaic modules. The aluminum frames and the j-boxes were manually removed from all modules. The remaining PV sandwich structure (referred to as *laminate* hereafter) were then shredded with a SM300 knives shredder (Retsch, Haan, Germany) with the experimental parameters based on previous work [33,36]. The module was fed through the shredder until the output material went through a screen with 2 mm opening. Subsequently the shredded mix was sampled using a method commonly called "homogenous-quartered-standard-weight": the output was divided into four ("quartered"), and then samples of equal weight (300 g in this case) are taken from each. This standard weighting was chosen based on the suggestion of previously published work [33].

Ten such samples were each separated using the electrostatic process to test for variability. Each was fed into a MMPM-618C (Eriez, Erie, USA) high tension roll separator. Two containers were placed underneath the separator, thus dividing incoming material into two groups: conductive

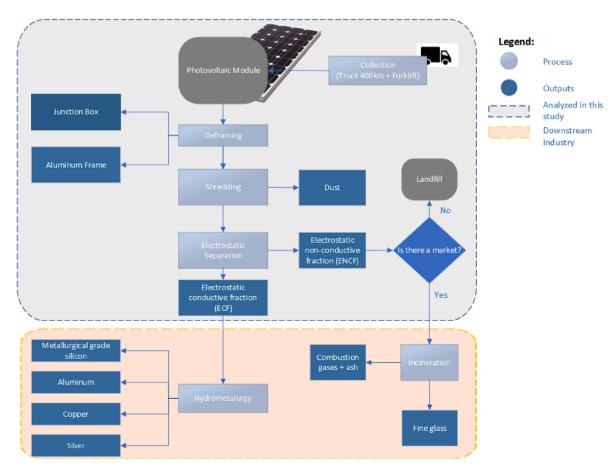


Fig. 1. Schematics of the methodology applied in this study, highlighting boundaries considered.

and non-conductive. The electric potential difference was 24 kV and the rotation speed of the roller was 30 revolutions per minute. All other process variables were kept fixed at values used in previous work [33]. These parameters are given in supplementary table ST1 and Supplementary Fig. S1. Any material that remained adhered to the roller was dislodged by a brush and collected in the non-conductive container. The humidity of the room was measured and kept below 45% using an Arsec250 dehumidifier (Arsec, Sao Paulo, Brazil). An external AK28 New hygrometer (AKSO, Sao Leopoldo, Brazil) was also used to measure the humidity.

The electrostatic separation process had two outputs: the electrostatic conductive fraction (ECF, hereafter) and the electrostatic nonconductive fraction (ENCF, hereafter). The weight of the samples before separation and after four sequential separations was recorded. Additionally, the operators measured the active time (i.e., in which there was human operation), idle time and electricity consumption, the latter using a DDS238-2 single phase electricity meter (Hiking, Yueqing, China).

Among the 10 standardized samples that underwent the electrostatic separation process, 5 were used to measure separation of metals and polymers, and 2 of these were also used to measure the separation of glass and silicon. To evaluate the metal distribution (silver, copper and aluminum) in each fraction, the outputs (i.e., ECF and ENCF) were digested in nitric acid (65% concentration), to leach silver and copper and then hydrochloric acid (38% concentration), to leach aluminum [16]. Each digestion was conducted at room temperature, had a 10:1 liquid-solid ratio (to ensure complete digestion) and was magnetically stirred. After each digestion, the solid fraction was separated by filtration, then rinsed and dried. The solid fraction was reserved. The solutions analyzed by inductively coupled plasma optical emission spectroscopy (ICP-OES) to determine the amount of silver, copper and aluminum in each sample. The equipment used was a 5110 ICP-OES (Agilent Technologies, California, USA).

The solid fraction was then analyzed; the amounts of polymers, glass and silicon were measured using the same experimental procedure described in detail in a previous work [33]. Briefly, the reserved solid was placed in a furnace under atmospheric conditions (500 °C for 5 h). The gravimetric mass difference before and after the furnace was assumed to be the mass of the polymeric fraction contained in each fraction (ECF vs. ENCF). The validity of this assumption was verified in a previous study [37]. As for the glass and silicon, samples were ground and analyzed by X-ray diffraction (XRD) using a Siemens (Bruker AXS, Germany) D-5000 diffractometer. Rietveld Quantitative Phase Analysis (RQPA) was used to measure the crystallinity of the samples by adding an internal standard of hexagonal (P63 mc) ZnO. Detailed parameters including steps, angles, voltage, software, etc. can be found in the previously published work [33]. Any material categorized as amorphous phase or quartz phase were assumed to be glass, while material categorized as crystalline was assumed to be silicon.

To determine the significance of the results obtained from the experiments presented thus so far, a variance analysis was performed for a confidence level of 95% ($\alpha=0.05$). The variance analysis was performed for the silver, copper, aluminum and polymer separation. The cost of the XRD and RQPA made it unfeasible to have the necessary replicates to perform a variance analysis for glass and silicon.

2.2. Industrial scale process

After testing the effectiveness of the laboratory process, an industrial scale process using the same method was envisioned using the experimental results obtained and equipment designed for high throughput production. The process would handle around 13,000 tonnes/year. While the industrial scale process would run as depicted in Fig. 1 (process within the boundaries shown), two alternative scenarios were proposed: one where the deframing occurs manually (B1), and one where it occurs with an automated equipment (B2). All the equipment

for the envisioned processed were available "off the shelf", and quotations from manufacturers were obtained. The description of such equipment (throughput, electricity consumption, etc.) and justification of the cost assumptions are provided in the Supplementary Table ST2 and the accompanying notes.

It is important to establish the possible end-of-life routes to allow the comparison of different alternatives. Table 1 depicts the different alternatives envisioned and which processes are comprised in each. For instance, A1 includes the automated removal and sale of the aluminum frames (deframing) and the landfilling of the remaining laminate.

2.3. Life cycle assessment (LCA)

Life Cycle Assessment (LCA) is an analytical methodology used to assess any product or process from an environmental perspective. The method is based on collecting and analyzing information from the whole product/process life cycle, considering input and output flows such as energy, materials, waste, and emissions [38]. The LCA methodology used here is based on ISO 14040 standards [39].

The goal of this LCA was to analyze the environmental impacts of the innovative recycling process described, using the midpoint impact categories recommended by the ILCD Handbook [40]. The ILCD impact assessment method was chosen in order to compare our results with the "Full Recovery End of Life Photovoltaic – FRELP" process [23], which has a published LCA study containing the used database and the process environmental impact results [21].

The assumptions made to perform the LCA and the database used are detailed in the Supplementary Table ST3. The simplified inventory table is presented in Table 2. The functional unit chosen was 1 tonne of Sibased waste module. The system boundaries are presented in Fig. 1.

To ensure the LCA performed for the proposed process is comparable with that of FRELP, the energy mix used from the Ecoinvent 3 database [41] was the "electricity, production mix IT", which is the electricity mix from Italy (41% natural gas, 23% renewable sources, 15% coal; 6% oil, 15% import from other countries).

The electrostatic process proposed in this study and FRELP have different outputs (e.g., clean glass recovery, energy recovery, etc.). Thus, to ensure a commensurable comparison, the environmental benefits of the material and energy recovery from FRELP were computed and used to calculate the net impact of FRELP (process C, from Table 1). The impacts were obtained directly from [21], and the recovered materials considered are those presented in system boundary (see Fig. 1 in Ref. [21]). However, there was one modification: the increase in the amount of copper that can be recovered from waste c-Si. Latunussa et al. [21] claim that 1.08 kg can be recovered for every tonne of waste PV. This amount seems to be off by a factor of ten based on other sources independent from each other [4,14,42–44]. Thus, it is considered that FRELP can extract 10.8 kg of copper per tonne of PV waste processed.

Table 1End-of-life alternatives and allocated processes.

Α	A1	В	B1	B2	С	Process Name
•	•	•	•	•	•	Transport to recycling facility
•		•	•			Manual Deframing
	•			•		Automatic Deframing
		•				Laboratory shredding
			•	•		Industrial shredding
		•				Laboratory electrostatic
			•	•		Industrial electrostatic
					•	FRELP full process
•	•					Landfill Laminate
		•	•	•		Landfill ENCF
					•	Landfill FRELP Waste
•	•	•	•	•	•	Frame and Cu wire sales
		•	•	•		ECF sale
					•	FRELP Cell Ag, Cu, Si, Al Sales
					•	FRELP Glass Sales

Table 2Life cycle inventory table (comments and additional information in the supplementary material).

Scenario	Process steps	Inputs	Outputs
A: Deframing (Laminate to	Manual	_	-
landfill)	deframing		
B: Lab Process (ENCF to	Manual	_	
landfill)	deframing		
	Shredding	1.07 kWh/kg	0.06 kg/kg
		(electricity)	(dust)
	Electrostatic	1.1 kWh/kg	$0.03\mathrm{kg/kg}$
	process	(electricity)	(dust)
	Transport	400 km	
	Forklift work	1.14 l/tonne	
	(diesel)		
B1: Industry scale process	Manual	-	
with manual deframing	deframing		
(ENCF to landfill)	Shredding	0.045 kWh/kg	0.06 kg/kg
		(electricity)	(dust)
	Electrostatic	0.006 kWh/kg	$0.03\mathrm{kg/kg}$
	process	(electricity)	(dust)
	Transport	400 km	
	Forklift work (diesel)	1.14 l/tonne	
B2: Industry scale process	Automated	0.00094 kWh/kg	
with automated deframing	deframing	(electricity)	
(ENCF to landfill)	Shredding	0.045 kWh/kg	0.06 kg/kg
		(electricity)	(dust)
	Electrostatic	0.006 kWh/kg	$0.03\mathrm{kg/kg}$
	process	(electricity)	(dust)
	Transport	400 km	
	Forklift work	1.14 l/tonne	
	(diesel)		

Other details about the Ecoinvent database the normalization figures used are presented supplementary Table ST3 and ST4).

To further assess the impacts of the proposed process, a sensitivity analysis based on the different electricity sources (considering various percentages of renewable energy sources in the electricity mix – the assumptions are presented in the Supplementary Table ST5) was also conducted for process B2 (detailed in Table 1).

2.4. Life cycle costing (LCC)

To assess the commercial potential of this recycling process, the cost and revenues were assessed using a bottom-up cost model using the same methodology as described in previously published works [11,18, 45,46]. The recycling process flow was proposed as shown in Fig. 1: i) the frames and j-box are removed and sold, ii) the remaining laminate is shredded into fine particles, iii) these particles undergo a 4-stage electrostatic separation, iv) the conductive fraction (ECF) is sold to a downstream processing facility and v) the remaining non-conductive fraction (ENCF) is disposed of in landfill.

A bottom-up cost and uncertainty model was used to assess the recycling process cost, cost of landfill of non-recovered materials, revenue from recovered material and the resultant net cost. The different alternatives shown in Table 1 were all assessed: i) the demonstrated lab process, using equipment cost, throughput, electricity and labor usage data from the experiments (process B), ii) extrapolated high volume processes using price estimates for high volume production equipment combined with estimated improved throughput, electricity and labor use (processes B1 and B2), iii) a process in which the frames are removed, sold and all the rest is landfilled, and (processes A and A1) iv) FRELP process as described in [21,23], with cost, electricity, materials and labor data taken from [22] (process C). For each scenario shown in Table 1, the sales price for recovered material were based on the uncertainty ranges and are detailed in supplementary table ST6 and ST7. The selling price of the ECF was given a large uncertainty range based on informal discussions with metal refining companies.

The cost model used a Monte Carlo approach to consider the impact

of uncertainty in input parameters to the final calculated net cost. For example, the labor cost (in \$/h) is very different among countries [47], so instead of calculating based on one labor cost value, 50,000 cost calculations (or iterations) were completed, each with a different labor rate generated using a random distribution. To determine the distribution for each input parameter, the surprise test described in earlier work [46] is used to identify low and high values which become the 10th and 90th percentile values respectively of a log-normal distribution as used in previous work [11,18]. To ensure the different manufacturing scenarios are commensurable, for each iteration the values of variables such as the cost of electricity (in \$/kWh), labor (in \$/h) and landfill (in \$/kg) as well as the revenue from recovered material (in \$/kg) were kept consistent for each manufacturing scenario. All the cost figures provided in this study are given in USD (U.S. Dollars).

The outputs of this analysis were used to assess and compare the net cost of each manufacturing scenario, identifying the key cost and revenue drivers. Finally, a direct comparison between a high volume electrostatic based recycling sequence and the FRELP sequence was made to identify the key conditions under which the electrostatic process could be cost competitive with FRELP.

3. Results

3.1. Mass distribution, losses, energy and time consumption

The initial shredding of the modules consumed time and energy. Measurements indicated that this phase consumed about 1.07 kWh/kg of energy and 2.2 h/kg of time to process the module laminate. About 23% of this total time was active operation, while the remainder (approximately 77%) was idle time. The processing data for the electrostatic process is shown in Table 3. The total time includes the time of active operation (14.78% \pm 0.06 of the time, on average) and the idle operator time, i.e., while only the equipment was working (85.21% \pm 0.06 of the time, on average). Adding these two processes together, the laboratory scale experiments used about 2.2 kWh/kg of electrical energy and a total processing time of 10 h/kg.

The electrostatic separation yielded material losses of 2.95 wt% on average. Losses may be due to dust during processing or incapability of collecting all the material from the output containers. In addition to mass loss, energy consumed in the separation process and total time consumed are displayed in Table 3.

The average mass distribution after the electrostatic separation had 3.34 wt% (± 0.47) contained in the conductor fraction (ECF), while the remaining 96.66 wt% (± 0.47) in the nonconductor fraction (ENCF). Noting that the laminate represents roughly 82 wt% of the module and accounting for the mass loss, the ECF contained about 2.66 wt% of the total mass of the module.

Table 3Material loss, energy consumed, and time consumed during electrostatic separation process per kilogram of processed material.

Sample	Mass loss (wt.%)	Energy consumed (kWh/kg)	Total time consumed (h/kg)
1	5.86%	0.718	4.90
2	5.20%	1.101	8.37
3	3.52%	1.424	8.78
4	2.54%	1.324	8.28
5	3.35%	1.302	8.68
6	2.54%	1.024	7.80
7	3.35%	1.034	7.70
8	1.65%	1.007	8.17
9	0.04%	1.019	7.41
10	1.40%	1.013	7.21
Average	2.95%	1.097	7.73
Standard Deviation	1.73%	0.204	1.12

3.2. Metal separation – silver, copper and aluminum

The metals contained in the PV modules can concentrate on either one of the two outputs (ECF or ENCF) or be randomly distributed among the two. To assess the separation of metals from the original PV material mix, the metals from both outputs (ECF and ENCF) were digested and the leachate was analyzed via ICP-OES. The results of the chemical analysis were then evaluated by variance analysis to ensure statistical relevance. The results show that the metals concentrate in the ECF fraction (Fig. 2); and that the mass concentration was of 94.7% (± 2.39) for silver, 97.6% (± 2.52) for copper and 74.3% (± 3.99) for aluminum.

3.3. Polymer separation

The polymers contained in PV modules are of little economic value and only partially recyclable [14]. Therefore, these materials would ideally be allocated away from the concentrated fraction of the separation (i.e. would ideally fall into the ENCF). Indeed, this was achieved successfully given only about 2 wt%, on average, of the polymers were contained in the ECF after the proposed process (Table 4). Sample 3 has achieved 100% separation, leaving all polymeric matter in the ENCF, which indicates that further process optimization could improve the effectiveness of the process.

3.4. Silicon and glass separation

Results for the effect of the electrostatic separation on the silicon and glass are measured by analyzing the crystallinity of the remaining sample after removing all metals (3.2) and polymers (3.3). Table 5 shows the crystallinity of the ECF and ENCF, where the glass is considered to be the non-crystalline (amorphous) fraction plus any identified quartz fraction. Under these assumptions, the ECF had only silicon (no glass) in both samples, while the ENCF had both silicon and glass. The distribution of silicon in Sample 9 was 67.54% in the ECF and 32.46% in the ENCF. Sample 10 yielded similar result, with 68.28% of the silicon in the ECF and 31.72% in the ENCF. A visual representation of the silicon distribution taking the average of these two samples is given in Fig. 3.

3.5. Environmental impact (LCA)

Based on the LCA data collection and analysis, and using the midpoint impact categories from the ILCD method, the environmental impacts of the proposed electrostatic process were compared to that of FRELP [21,23]. The normalized [48] results are shown in Fig. 4, where

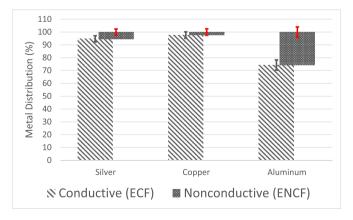


Fig. 2. Results of the acid digestion of metals followed by ICP-OES analysis determining the amount of silver, copper, and aluminum in which of the outputs of the electrostatic separation (ECF versus ENCF). The five sample replicates used were statistically evaluated and the p-value for all the distributions are <0.001; five replicates were used for each metal.

 Table 4

 Polymer mass distribution after electrostatic separation.

Sample	ECF (wt%)	ENCF (wt%)
1	2.40	97.60
2	3.43	96.57
3	0.00	100.00
4	1.14	98.86
5	1.64	98.36
Average	1.72	98.28
Standard Deviation	1.29	1.29

Table 5Crystallinity of materials in the conductive (ECF) and non-conductive fractions (ENCF) after electrostatic separation. Samples are the remainder of the leaching and thermal degradation process done prior.

Sample	Crystallinity (wt%)		
	ECF	ENCF	
9	100	2.08	
10	100	1.76	
Average	100	1.92	

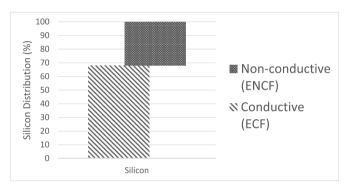


Fig. 3. Silicon distribution after electrostatic separation. Average of the two samples, distribution was $\pm 0.37\%$ around the average.

the impact of the laboratory process (process B) is not shown as it was about 40-fold greater than that of the optimized processes (B1 and B2) and about 6-fold greater than that of FRELP (C). However, it is presented in Supplementary Fig. S2. The difference in impact arises primarily from the difference in the electricity consumption, which was much higher in the laboratory process (B) in respect to the industrial scale ones (B1 and B2). Fig. 4 also shows that FRELP (C) has a greater impact than the proposed electrostatic at scale (B1, B2) before considering the benefits of recovery, which is expected given B1 and B2 are much simpler and not make use of chemicals or thermal treatments.

The LCA considered the environmental benefits of the material recovery for the following materials: aluminum frame, copper, silicon, silver and aluminum from the cell, and the quantities in kilogram per tonne of waste module recovered with B1 or B2 are 180, 10.2, 22.72, 0.46 and 1.9, respectively. More information about the mass distribution in waste c-Si PV modules is given in supplementary table ST8.

Regarding the environmental benefits of material recovery from the ECF, two extremes are considered in Fig. 4: the environmental benefits of recovering "ore quality" or "metal quality" materials from PV modules. The "ore quality" benefits assume the ECF requires similar amounts of processing as concentrated metal ores do before they can be used. The "metal quality" benefits assume the material is as useful as fully separated metals. The true environmental benefits are between these extremes – the amount of processing of the ECF is likely much less than that of reducing metal oxides in ore, but is also more than for pure metals, since some separation and purification (using pyrometallurgical or hydrometallurgical processes) would be required. Most importantly,

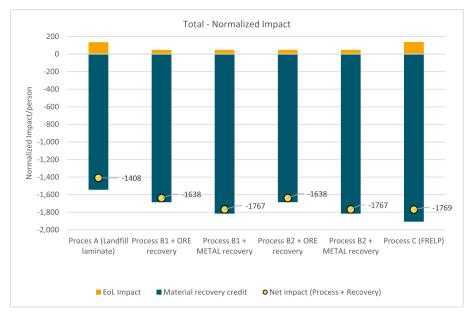


Fig. 4. Total normalized environmental impacts based on the ILCD normalization factors, including the benefits of the materials' recovery from the PV modules. FRELP results were calculated based on [21] but with a 10-fold increase in the amount of copper recovered. B1 is the industrial scale process with manual deframing and B2 is the industrial scale process with automated deframing. All situations consider the impact of transportation (400 km by truck) and of unloading at facility by diesel forklifts. Refer to Table 1 for a complete relation of the steps comprised in each process.

regardless of which impacts are considered the net environmental impact (i.e., the detrimental impacts minus the favorable impacts) is beneficial when implementing the proposed electrostatic process at industrial scale.

The net impact is environmentally beneficial for all the alternative scenarios considered (A, B1, B2, C). Fig. 4 suggests that only removing and recovering the aluminum frames and j-box is already environmentally beneficial, but that the more comprehensive the process is, the better the net impact it will be. Fig. 4 provides the normalized impact for all categories, a detailed list with the individual impact categories is shown in supplementary table ST8, which are on par with the authors that looked into the net impact of FRELP [19].

Transportation is responsible for a significant fraction of the detrimental impact: 68%, 67% and 23% of the impact of B1, B2 and C, respectively. This difference between the electrostatic process (B1 and B2) and FRELP (C) is primarily because the total impacts of the B1 and B2 are significantly smaller than that of FRELP (about 6.7 times smaller), while the impact of transportation is independent of the recycling

process chosen.

Lastly, the sensitivity analysis performed considered the contributions of renewable energies increasing from 23 to 100%, and the normalized results are shown in Fig. 5. The analysis was done for process B2, the most automated electrostatic alternative. The results show a potential decrease of 27% ($\pm 1\%$) in the electricity impact can be obtained when moving from the initial Italian energy mix (which is FRELP's energy mix assumption) to an energy matrix based exclusively on renewable energy (which linearly translates into an 8.6% overall decrease). The normalized impacts are a weighted sum of many different environmental impact measures, and the breakdown of each measure is shown in Supplementary Table ST10. It is noteworthy that only one impact factor is made worse with 100% PV - abiotic depletion. This is because the LCA impacts are calculated on the assumption that the PV modules producing the renewable electricity are not recycled at their end of life, and would thus contribute to the depletion of Ag, Cu, Al, etc. This result emphasizes the need to find sustainable solutions to PV waste management to ensure that PV has a beneficial effect on all

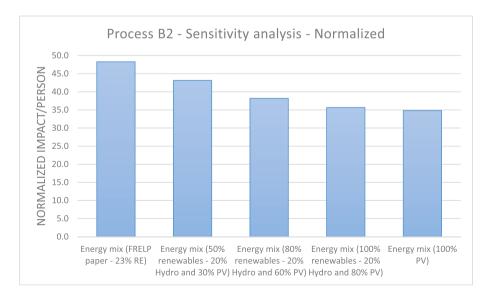


Fig. 5. Sensitivity analysis of the total normalized environmental impacts based on the ILCD normalization factors, considering different electricity mix as the energy input for the recycling process.

environmental impact categories [8]. The recovery of non-renewable materials from solar modules is important for this technology to continue to be sustainable.

3.6. Economic scenarios (LCC)

The cost analysis of the electrostatic process was initially assessed assuming the same equipment, throughput, labor and electricity usage observed from the experimental work (process B). As would be expected, the processing cost was very high (a cost breakdown shown in Fig. S3). The median estimated cost of \$10,000/tonne obtained is clearly too high compared to the \$400–500/tonne proposed by Deng et al. [11] for a cost-effective mechanical recycling processes. The reasons for the high cost can be seen in Fig. S4: i) the high capital cost caused by a small processes throughput, in particular for the electrostatic process and ii) the high labor cost due to the requirement to continuously feed the equipment.

Using these findings, an industrial scale process was envisioned (2.2 Industrial scale process), and the result of this optimization was a much lower production cost (Fig. S3, Process B1 and B2). The industrial scale electrostatic processes (B1 and B2) were then compared to simple landfilling (processes A and A1, where A has manual deframing and A1 has automated deframing) and a more complex FRELP process (C). This comparison is shown in Fig. 6.

Note that frame removal automation significantly reduces the cost primarily by reducing the labor cost (A \rightarrow A1 and B1 \rightarrow B2). The low cost assumes that the automated frame removal equipment is operating at full capacity, so smaller throughputs would incur in higher costs. Comparing B1 and B2 to the more complex process FRELP (C), the electrostatic process will most likely have lower costs than FRELP – an unsurprising result, since the process has fewer steps and recovers material that is less pure. However, this neglects the benefit of revenue from the recovered material, which is much greater for FRELP than the electrostatic process. Fig. 7 gives a more complete picture, including revenue, transportation, and landfill costs. Supplementary Fig. S5 shows the distribution of net cost for A1, B2 and C, indicating that FRELP is likely to be more profitable than either the electrostatic (B2) or frame-only (A1) recycling processes.

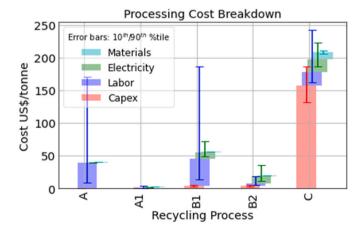


Fig. 6. Processing cost breakdown of deframing followed by landfilling process (A and A1) compared to deframing followed by electrostatic processes (B1 and B2) and FRELP (C). The height of each bar shows the median value, and the error bars show the 10th and 90th percentile values of each cost component. Capex stands for capital expenditure which is depreciated over 10 years, i.e. funds used by to acquire, upgrade, and maintain physical assets such as property, equipment, buildings, etc.

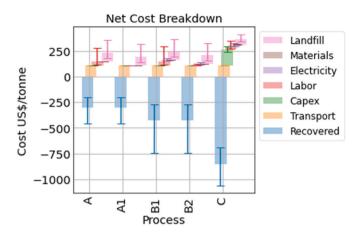


Fig. 7. Breakdown of process costs shown together with the revenue from recovered materials, transportation to the recycling facility and landfill of waste. A and A1 are manual and automated deframing (respectively), followed by landfilling the laminate. B1 and B2 are manual and automated deframing (respectively), followed by shredding and electrostatic separation. C is the FRELP process.

3.6.1. Sensitivities to ECF value and glass recycling revenue

There are significant uncertainties in both the costs and revenues for all the processes, but because the Monte Carlo analysis for each process was done simultaneously, it is possible to do a point-wise comparison between these processes to determine whether there are key variables that determine which provides the optimum net cost. This analysis showed that when comparing FRELP to B2, the most important financial factors were the value at which the ECF could be sold for compared to the FRELP revenue, and the revenue FRELP obtains from the glass (which is landfilled in the case of B2). The relationship between these factors and the difference in net cost between these processes is shown in Fig. 8A. The closer the ECF can be sold for the full value of its constituent materials, the more cost-competitive the electrostatic process becomes.

The question of the economic value of the ECF is difficult to assess until large volumes can be generated and tested by metal refineries. Our expectation is that the ECF will be most highly valued by a recycling plant designed to recover silver, copper and high purity silicon – such as the FRELP metal recovery processes. Unfortunately, such a tailored recovery facility would require large volumes of modules in order to be cost effective. It is surmised that the electrostatic process can be a step towards this, if it can be deployed at low cost in many locations, and each can then ship (at low cost) the relatively low mass of ECF to centralized specialized Ag/Cu/Si recovery facilities.

Another difficulty facing the recycling industry is the ability to sell the recovered material, in particular the recovered glass. This has been confirmed in over five industry enquires and online [49–53]. One of the reasons given was that recycling 100% of recovered glass is not economically viable because the alternative input material (silica sand) is cheap and readily available [53]. Since the electrostatic separation does not attempt to recover glass, but rather separates it and landfills it, this situation would benefit this process from a cost perspective. To understand this current situation, B2 was compared again to C (FRELP), but instead of a selling price range of the glass, the possibility that it costs money to landfill the glass is also explored. The new comparison is shown in Fig. 8B, which helps understanding that if the ECF can be sold for around half the value of the constituent materials in their separated form, this would make economic sense compared to FRELP if the recovered glass had to be landfilled at a cost of \$100/tonne (because there were no buyers for it, for instance).

3.6.2. Sensitivity to production volumes

One important simplification within the cost model is that the capital cost depreciation is calculated assuming that each equipment purchased

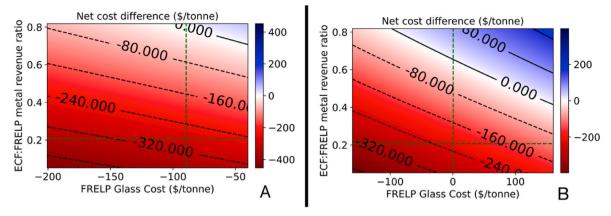


Fig. 8. The impact of the ECF:FRELP metal revenue ratio and the FRELP glass cost on the relative net cost of FRELP and the electrostatic process (Fig. 8A) without and (Fig. 8B) with glass exploration. A negative cost of glass means that the glass can be sold to recyclers, a positive cost indicates that a fee must be paid for disposal. The red region indicates that FRELP (Scenario C) is more profitable than the electrostatic process with automated deframing (Scenario B2), while blue indicates the opposite. The contour lines indicate the net cost difference in \$/tonne.

is run at full capacity over a 10-year depreciation period - there is no adjustment for mismatched throughput or under-utilized equipment. For the optimized electrostatic process (B2), the median values of annual throughput for the de-framer is 15.8 kt/year, the shredder 21.4 kt/year of laminate (26 kt/year of module) and electrostatic separator 10.7 kt/ year of laminate (13 kt/year of module), and for FRELP (C), the cost data from Faircloth et al. [22] assumed a production throughput is 8 kt/year. The cost model used can be adapted to calculate the cost of processing a specific annual capacity of the factory. For example, in early years of recycling, only 1 kt/year of modules may be available, so that equipment may be under-utilized by 90%. The equipment capital cost attributed to this "idle" time can be calculated, while assuming that labor, electricity and material usage per tonne is unaffected. The net cost breakdown is exhibited in Fig. 9, which shows that the industrial scale electrostatic process has minimal additional idle time cost, even at production throughputs as low as 1 kt/year, whereas the high capital cost FRELP factory would be very uneconomical to build and run at such low volumes, and at 4 kt/year, the net cost of both processes are similar. The net cost distributions comparing B2 to C at different production capacities is shown in Supplementary Fig. S5. This suggests that the electrostatic process could be a good interim solution to process modules while waste volumes are low, but that once volumes increase to a critical level, it is possible to economically transition to a full recycling process such as FRELP (C).

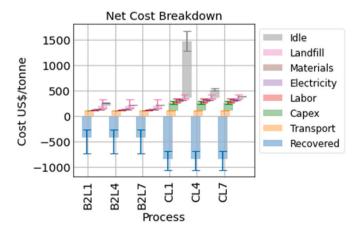


Fig. 9. Net cost breakdown of the optimized electrostatic process B2 and FRELP process C, assuming **low** production throughput of 1 (B2L1, CL1), 4 (B2L4, CL4) and 7 (B2L7, CL7) kt/year.

5. Discussion

5.1. Technical implications

The main feature of the process described is the concentration of the valuable material contained in silicon PV modules in a small proportion of its original mass, i.e., the high concentration of materials of interest. The process was able to separate the original mixture of materials into two groups: the ECF containing 3.34 wt% of the mass and the ENCF containing the remainder. This small mass fraction (ECF) included, on average, 95 wt% of the silver, 98 wt% of the copper, 64 wt% of the silicon, and 74 wt% of the aluminum (the latter is from the photovoltaic cell, given that the frame had been previously removed and was 100% recovered). The other fraction (ENCF) included 100 wt% the glass, 98 wt% of the polymers (encapsulant, backsheet) and 32 wt% of the silicon, on average (Fig. 10). Thus, the output of the process is a high-value concentrated material mix, which can be further separated by known metallurgical processes.

Mechanical processing aims to create a material (ECF, in this study) rich in valuable metals [54], which can be easily handled by downstream recyclers. Ideally, the small fraction of mass should contain most of the valuable materials. Savvilotidou et al. [55] experimentally studied three processes to pre-concentrate silver when recycling polycrystalline silicon solar panels. The mechanical crushing using a blade rotor followed by sieving could concentrate 89% silver in 35 wt% of module mass. A more energy intensive thermal process followed by gravity separation can concentrate 91% silver in 3.7 wt% of the mass. A chemical-thermal process followed by manual sorting could concentrate 95% silver in 3.8 wt% mass. In this work, the shredding and electrostatic process achieve a concentrate of 95% silver in 3.34 wt% of the mass, showing high selectivity compared to the conventional mechanical processing, and comparable concentrating efficacy to more complex, energy-intensive and labor-intensive thermal and chemical processing. Furthermore, if the metals in the ECF fraction can be entirely recycled, the electrostatic process yields similar amount of aluminum and 57 wt% more copper than a conventional mechanical shredding process, for instance [22]. This conventional mechanical process also discards silver and silicon, which can potentially be recovered from the ECF fraction in the electrostatic process [22].

5.2. Environmental implications

The proposed electrostatic process, when applied at scale (B1 or B2), is environmentally beneficial as revealed by the life cycle assessment (LCA). When including the benefits of the material recovery, the

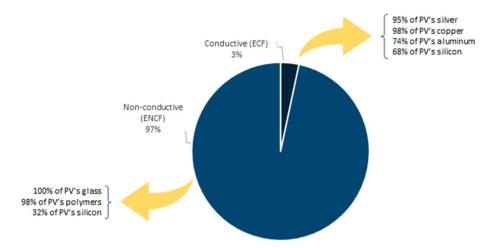


Fig. 10. Summary of separation achieved through the proposed process.

normalized LCA shows that it has a net positive environmental impact, meaning benefits of the material recovered outweigh the negative impacts of processing, which implies that the overall impact is smaller than that of obtaining these materials through the primary source.

When comparing the end-of-life alternatives proposed, the more comprehensive the recycling (i.e., the more materials can be recovered), the more environmentally beneficial it is. Therefore, environmentally, a full recovery process (FRELP, C) should be favored in respect to an intermediate process such as the electrostatic (B1, B2), which should be favored in respect to the process of manual frame removal (process A). This preference order (C > B > A) can be also understood as the reverse order of landfilling, i.e., the more materials are landfilled, the worse the process is from an environmental standpoint.

The environmental impact of "inaction", (a situation where nothing is recovered and the whole module is landfilled), is the most detrimental among the possibilities, as the negative impacts of such landfilling would be greater than that of process A (Fig. 4) without the benefit of material recovery, i.e. it would be a net detrimental impact. This conclusion is on par with other publications in this area [13,56]. The baseline of "inaction", however, is the default situation in several countries regarding waste PV.

Transport is a key contributor to all impact categories. This conclusion had already been drawn [21], and the current results obtained confirmed the significant environmental contributions from transportation in the recycling processes of PV modules. The importance of the transportation may lessen the negative impacts of landfilling the whole module, as landfilling is generally associated with smaller transportation impacts because there are many more landfill sites in respect to recycling facilities.

The impacts of any of the alternatives assessed (A, B1, B2, C) could be minimized by using renewable electricity sources instead of the Italian energy mixed originally used. The analysis done for process B2 shows that the electricity impact for proposed process can be decreased by about 27% if renewables are used to power the process. Given the normalized impact of electricity represents about 32% of the total, the total potential reduction would be of about 8.6%. From this sensitivity analysis, it can be inferred that the more fossil fuel dependent a region is, the more environmentally favored the proposed electrostatic process should be in respect to FRELP. This is because the former consumes less than half the electrical energy the latter consumes (and electricity represents about a third of the impact of process B2, for instance).

5.3. Economic implications

The economic analysis of the proposed process shows that if all the

material separated using a full recovery process like FRELP are sold, then this is the best end-of-life alternative among the ones compared (A, A1, B1, B2, C). However, if the glass cannot be sold due to limited market availability, then the proposed electrostatic process may be economically more profitable depending on the valuation of the ECF and the cost of landfilling.

The proposed electrostatic process also benefits from situations in which the incoming waste volumes are not sufficient to operate at full capacity because the capital expenditure of a full recovery process such as FRELP is much higher than that of the electrostatic process (Fig. 9).

The results show that the transportation is a significant contributor to the cost (Fig. 7). This also favors a less capital-intensive process such as the proposed electrostatic because it is more feasible to have it installed in more locations, hence reducing collection distances. Equally important is the consideration of the location of the infrastructure required to deal with the material output from each process. If the downstream industry capable of processing the output materials is located far away from the waste source (e.g., if the waste is in Australia, but the downstream industry is located abroad [57]), then the electrostatic method is preferred, given only a small fraction of the total mass has to travel long distances to be processed.

5.4. The importance of glass recycling

The analysis of Fig. 8 makes it evident that the revenue obtained from the glass waste PV is significant in determining which recycling process to choose from a financial standpoint. This is mainly because the glass represents most of the PV panel in weight (roughly 70–75% [14]). If it can be sold, then it will be a source of revenue (even if small in absolute value), but if no market exists for it, it must be disposed of or stockpiled - becoming an economic liability. The literature generally assumes that glass can be resold to the manufacturing market, and, more often than not, at a profit [11,22,58]. However, as discussed previously (refer to 3.6 Economic scenarios (LCC)) the finding of suitable companies willing to buy recovered glass was challenging and, to date, unsuccessful.

Environmentally, the decision to recycle or landfill the glass is minor, as it represents only about 1% of the normalized benefits of material recovery (case of process C) and only about 4% of the negative impacts of landfilling (case of processes A, A1, B1 and B2). Among individual impact categories, the landfilling of glass had the greatest representation in particulate matter (16%), photochemical ozone formation and terrestrial eutrophication (both 14%), ozone depletion and acidification (both 12%).

5.5. Limitations

The proposed methodology was based on a previous framework [33] that was enhanced in this paper. Further work would be necessary to optimize the process to increase the yield of silver, copper and silicon (e. g. determine the optimum particle size to favor electrostatic separation). This was not done for this paper because the objective was to evaluate the process comprehensively (technically, environmentally and economically), and it would be overwhelming to also optimize the process alongside the rest of the analyses. As is, the proposed process already offers an alternative route for material recycling while large volumes are not sufficient to justify full recovery processes. Further work should not only try to optimize the process, but also try to do it at pilot scale.

The amount of material lost during processing (in the form of dust and inaccessible material in the output container) is not negligible (roughly 3% on average) and can skew the results if the amount lost does not contain the materials in the same proportion as the remainder of the sample (e.g., if silicon is more prone to be lost as dust in respect to glass). Losses can be reduced with a dust collector. Studies at pilot scale should observe less material loss and would be able to collect and observe the composition of the dust and/or the inaccessible material after shredding and electrostatic separation. Likewise, a pilot scale study should be carried out to confirm the results obtained in this study, given the results are based on bench scale samples of 300 g.

Lastly, this study has simplified the impacts of transportation by pinning the distance from the collection point to the PV recycling site at 400 km. Given transportation has significant influence in both the environmental and economic considerations, additional studies evaluating different means of transport and distances would be important to better understand this dynamic relationship and to allow for an optimization in the placing of recycling facilities.

5.6. Main conclusions

The proposed process is capable of processing waste silicon photovoltaic panels and concentrating most of its valuable components in a small fraction of the original mass (2-3 wt%). The final separation includes i) the aluminum frames and j-box, ii) a mixture of mostly silver, copper, aluminum and silicon (ECF), and iii) a mixture of mostly glass, polymers (backsheet and encapsulant) and silicon (ENCF).

From an economic perspective, this process is recommended in situations where there is not enough waste volume to justify the full recovery of material contained in end-of-life PVs, in situations where the infrastructure required for end-processing is not geographically close to the waste source, and may be desirable when there is no market to buy the glass extracted from end of life PVs. In other situations, a full recovery of materials (such as the one proposed by FRELP) is superior.

Environmentally, the less material is landfilled, the more beneficial the process is. Thus, among the explored end-of-life scenarios, a full recovery (FRELP) is preferred, followed by the proposed electrostatic process, followed by exclusively recovering the aluminum frames, followed by the landfilling of the whole module. All recovery options have a beneficial net environmental impact, while landfilling the whole module has a detrimental net impact. These conclusions are based on the normalized result, which removes important nuances from the LCA. All the individual impact categories are presented in the supplementary information section for a more in-depth analysis.

Glass is an important economic component to be considered when evaluating the prospects of end-of-life PV recycling. Mainly because it represents most of the weight of the panel, and thus can be a source of revenue or a significant economic liability if there is no market for it. Future research and analyses ought to consider the possibility of there being no downstream market for glass when evaluating end-of-life prospects.

CRediT authorship contribution statement

Pablo R. Dias: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Writing – original draft, Writing – review & editing, Visualization, Supervision. Lucas Schmidt: Conceptualization, Investigation. Nathan L. Chang: Methodology, Validation, Formal analysis, Writing – review & editing, Visualization. Marina Monteiro Lunardi: Methodology, Validation, Formal analysis, Writing – review & editing, Visualization. Rong Deng: Validation, Writing – review & editing, Visualization. Blair Trigger: Methodology, Validation, Formal analysis, Writing – review & editing, Visualization. Lucas Bonan Gomes: Methodology, Formal analysis, Investigation. Renate Egan: Resources, Writing – review & editing. Hugo Veit: Validation, Resources, Writing – review & editing, Supervision.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: The main author of this work (Pablo R Dias) has recently (2022) started a photovoltaic panel recycling business. The methodology, results and analysis reported in this study, however, were done prior to the creation or contemplation of creation of such business and were not influenced by the new venture. As such, 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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.rser.2022.112900.

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