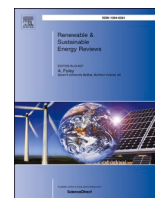


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Technical challenges and opportunities in realising a circular economy for waste photovoltaic modules

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ABSTRACT

The global exponential increase in annual photovoltaic (PV) installations and the resultant levels of PV waste is an increasing concern. It is estimated by 2050 there will be between 60 and 78 million tonnes of PV waste in circulation. This review will investigate and establish the most efficient routes to recycle end-of-life modules. It will consider current design constraints, focusing on the maximum recovery of constituents from the module, reporting on some of the latest advancements in recycling methodology at both industrial and laboratory scale. Circular challenges, opportunities, models and arguments are presented for critical analysis of closed-loop recycling alongside alternative open-loop cascading options. Adopting circular economy principles will help offset environmental factors such as emissions associated with the manufacturing stages and increase recycling & recovery rates. First-generation crystalline silicon (c-Si) modules have had an 80–90% market share over the last 40 years and will constitute the majority of the impending PV waste stream. These PV modules are composed of several material types such as glass, metal, semiconductor and polymer layers in a strongly bound laminate. This design makes reusing and maintaining these modules difficult and limits potential recycling options. Here we provide guidance for understanding the c-Si PV module manufacturing process and how to best approach the challenge of recycling this vast and inevitable waste stream. In conclusion, pyrolysis offers the best potential for the optimum recovery of material and energy found in first-generation c-Si modules to help promote a truly circular economy within the well-established PV industry.

1. Introduction

Worldwide, as the transition from conventional fossil fuel sources to renewable energy technologies occur, the public's opinion towards renewable energy is ever-changing. Of this technology, the solar PV industry has experienced on average 50% growth per year from 2006 to 2016 and a 32% growth rate in 2017 [1,2]. At the end of 2018, the cumulative global installation of PV was 515 gigawatt (GW) [3], crossing half a terawatt (TW) in that particular year [4], and is estimated that by the end of 2019, will reach the mark of 540 GW [5]. The industry

will prove invaluable, not only from an energy perspective but also an environmental one to help achieve Paris climate targets in the transition to a low-carbon future [6–8]. Renewable energy technologies such as solar, wind, hydro, tidal, geothermal and biomass must be developed and installed sustainably in order to mitigate any potential adverse environmental impacts and help achieve the Sustainable Development Goals (SDGs) outlined by the United Nations [9–16]. One of the key aspects of the transition to sustainable energy systems, along with long term environmental impacts, is energy security on the national, regional or local level [17]. One such example is solar taking the lead for renewable capacity additions in 2018 at 55%, next to wind and

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Nomenclature			
<i>Abbreviations</i>		TPE	Tedlar-Polyethylene terephalate-Ethylene vinyl acetate
Al-BSF	aluminium-back surface field	TPT	Tedlar-Polyethylene terephalate-Tedlar
[Al _n (OH) _m Cl _{3n-m}]	poly-aluminium-hydroxide-chloride	UN	United Nations
ARC	Anti-reflective coating	WEEE	Waste electrical and electronic equipment
a-Si	Amorphous silicon	<i>Units</i>	
BIPV	Building integrated photovoltaics	GJ	Gigajoules
CIGS	Copper indium gallium selenide	kWh.kg ⁻¹	kilowatt-hour per kilogram
c-Si	Crystalline silicon	MJ.m ⁻²	Megajoules per square metre
EoL,	End-of-life	µm	micrometre
EVA	Ethylene vinyl acetate	MW	Megawatt
ICP-MS	Inductively coupled plasma mass spectrometry	<i>Symbols</i>	
IEA	International energy agency	Ag	Silver
IRENA	International renewable energy association	Al	Aluminium
ITRPV	International technology roadmap for photovoltaics	Cu	Copper
PERC	Passivated emitter rear contact	POCl ₃	Phosphoryl chloride
SDGs	Sustainable development goals	Pb	Lead
SiN _x	Silicon nitride	Sn	Tin

hydropower at 28 and 11%, respectively [18]. In our local demographic of Europe, the EU has developed a plan to increase the share of renewable technologies in the production of energy to a minimum of 30% by 2030, with this eventually rising to 100% by 2050 [19,20].

This global expansion of the PV industry is due to different demographic regions throughout the world capitalising on the fact that PV technology arguably has the highest potential of all renewable energy technologies to replace fossil fuels [21,22]. One advantage is that PV modules have the ability to be installed at numerous scales (from small scale decentralised, off the grid to large scale PV farms) due to their modular nature and the ability to add more modules to any existing PV system [23]. Additionally, one way in which the solar photovoltaic industry has the advantage to become the most popular form of renewable energy is its potential to produce electricity without producing any emissions, noise or any form of air pollution whilst in operational use [24–26]. At the end of their design life, PV modules are now considered electronic waste (e-waste) and fall under the category of large equipment [27].

E-waste conventionally ends up on landfill sites along with other municipal solid waste (MSW) or is incinerated with little gas emission control, releasing toxic and carcinogenic materials into the atmosphere [28]. Most End-of-life (EoL) PV modules globally are sent to landfill. Landfilling is not an environmentally friendly option and is not sustainable long-term. With the heavy metals present in crystalline silicon (c-Si) PV modules such as lead and tin, these materials can leach or contaminate the soil or groundwater, resulting in significant environmental pollution issues.

It is estimated that by 2050, the global PV waste category could exceed and account for 10% of the total Waste Electrical and Electronic Equipment (WEEE) generation. Additionally, it could represent the most significant e-waste stream if there is not a correct and preventative measure about how to manage and recycle future wastes of this type [29, 30]. As reported by Czajczyńska et al. [31], the traditional waste management methods of landfilling and incineration are outdated at this point in time and need to be replaced by modern, effective and user-friendly solutions. Alternative approaches to traditional disposal now consider the life cycles of raw materials and products. The current outdated approach is to take raw materials, make a product (i.e. PV module) and at the end of its design life, dispose of it without recognising the value or potential value in the product or its materials. This model is known as the linear economy [32].

First-generation PV modules contain finite rare and precious metals that are mined from the earth such as silver, copper, aluminium, lead

and tin. Of these finite materials, Hunt et al. [33] have shown there are 5–50, 50–100, and 100–500 years until depletion of silver and tin, copper and lead, and aluminium, respectively. As a result of worldwide lists of critical raw materials, manufacturing industries are beginning to realise the value that is often locked within EoL products. The approach to unlock and recover value from these EoL products, and ultimately to design new products so that value can easily be recovered and these products re-used is known as the “circular economy” [32]. Adopting circular economy principles could generate 4.5 trillion USD of additional economic output by 2030 whilst dissociating economic growth and natural resource consumption [34].

Currently, there is a need for a framework that gives appropriate signals to PV manufacturers and consumers to promote finite resource-efficiency in PV modules and the circular economy [35]. To date, limited research on the recycling methodologies of PV technology exists considering the impact it will undoubtedly have in the future. Despite this, the first studies on the technical and economic feasibility behind the recycling of c-Si modules and the idea of pyrolysis were published in the 1990s [36–38]. Since then, there have been significant changes and improvements in the manufacturing process to minimise the number of constituents used in a PV module. Also, advancements at a cellular level have been made by researchers to maximise the efficiencies of the available PV modules on the market. However, the majority of the research concerning photovoltaics focuses on laboratory-scale third-generation PV, with limited attention given to circular EoL approaches for legacy c-Si PV modules.

Considering the increase in PV installation and commercial setups of PV farms or renewable energy generation centres globally, the recycling, re-use and recovery of materials will become an issue in the future. It is advisable to start researching and employing these solutions now so that a deficit of some of the rare and precious metals in these panels do not occur in the future. This would have a significant impact on the supply chain for the solar industry but also other industries using these materials.

This work summarises and provides an up-to-date review of the state-of-the-art advances in PV module recycling. More specifically, first-generation c-Si PV modules, that constitute the dominant market share of photovoltaic technology in all demographic regions for the last 30–40 years. It outlines efforts from academic and industrial sources in the areas of thermal, chemical and mechanical recycling. The objective of this review is to provide implications on the most efficient recycling method for c-Si PV modules to date in the form of delamination and removal of degraded polymeric material as well as report on ways to

utilise the waste polymers. The polymers are known to be a main and significant contributor to PV modules EoL status. Once they are removed, this allows recovery of the leftover constituents. Additionally, the review also suggests steps & future work from the evident gaps in the knowledge. The conclusions of the study will help support researchers in the field, the policies on end-of-life management and will focus on themes such as the circular economy and environmental impacts.

2. Methodology

In constructing this literature review, databases such as Web of Science and Scencedirect was primarily used to obtain literature. As the recycling of PV modules is an issue concerning academia, industry and policymakers alike, there was some grey literature used in the form of governmental and industry reports (such as EU commission, Fraunhofer, IEA, IRENA, ITRPV and PV CYCLE), that was required for essential data to assess the most efficient recycling strategy for these modules.

With regards to Web of Science, their core collection database was used and a 25-year period from 1994 to 2019 was selected for this particular study. This was due to prior knowledge that the first report which had considered reuse of materials in PV modules using pyrolysis or chemical methods was published in 1994 by Bruton [37]. As this study does not consider second-generation PV modules, it was decided that boolean operators to not include thin-film or include multiple search criteria would be utilised in specific cases shown in Table 1. Boolean operators are shown in uppercase.

The keywords that were compiled for the search were as follows: PV module, PV recycling, recycling, crystalline silicon, thin-film, circular economy, pyrolysis, Sustainable development goals. The articles were then further screened for results referring to terms including but not limited to: delamination, deconstruction and EVA removal.

From the search results, it is evident that there have been significant increases in both the number of citations and total publications over the last ten years and more notably so, the latter half of the ten-year period. This is likely due to the announcement of PV modules being added to the WEEE directive and active as of the year 2014. For example, the most basic of search criteria case 1, “PV module AND recycling” produced 179 results and an increase of 29 times the number of citations over the 2009–2019 period. When employing further search criteria to remove any mention of thin-film technology, this still equated to 23 times the number of citations over the ten-year period. It is worth noting that when using this search logic that any literature facilitating both c-Si and thin-film technology would have been left out if only this case was considered.

Furthermore, case 4 produced only one result, which happened to be our previous work, indicating a direct gap in this research area. Case 5 was interesting in that the term circular economy in relation to PV recycling has only been mentioned in literature over the last five years. This has seen the most significant rise in citations of any of the other search criteria over a relatively small timeframe of 4 years. This search would also happen to include general research papers and reviews on e-

waste. The number of publications and citations are expected to increase as the issue of EoL PV modules becomes more of a concern in the near future.

Interestingly, case 6 showed that although there was outlying literature dating back from 1994 on the potential of pyrolysis for the recycling of PV modules, there has been a resurgence of research papers in the last three years despite evident gaps. Finally, case 7 yielded a valuable observation in that there was a clear gap of literature relating to PV recycling and the UN’s sustainable development goals. These results provided a clear insight into gaps and focus areas for this review.

However, some methodology limitations should be noted. One such reason is that the technologies and concepts that relate to the topic of PV recycling are multi-disciplinary in nature and span many different trains of thought and skill sets. As academic and industrial efforts currently exist in different sectors and areas, it is much more difficult to review, and screen literature widely compared to one particular recycling methodology. Another is that the number of databases was limited for this study and may not include some outlying literature in searches that could be deemed as potentially eligible for the study.

3. Drivers for circular approaches to recycling PV modules

3.1. Governmental legislation

The main legislative drivers behind industry and recycling bodies approach to PV modules and their recycling are the EU’s revised Waste Framework (rWF), Restriction of Hazardous Substances (RoHS) and Waste of Electrical and Electronic Equipment (WEEE) directives. The revised waste framework directive introduces into law in EU member states circular concepts like the waste hierarchy, which is shown in Fig. 1. Here landfilling of material is the worst option, then energy recovery, recycling, re-use and minimisation, then prevention of waste through efficient design. To fully engage with, and travel up the waste hierarchy, the concept of designing products for ease of recycling,

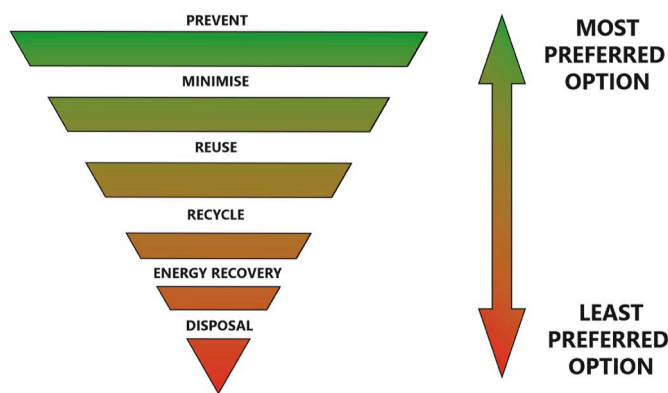


Fig. 1. The waste management hierarchy adopted in Europe.

Table 1
Cases of Web of Science criteria and results used to obtain literature for the review.

Case number	Keywords and boolean operators used	Number of results	Citation percentage increase from 2009 to 2019 (%)	Citation percentage increase from 2015 to 2019 (%)	Citation percentage increase from 2017 to 2019 (%)
1	PV module AND recycling	179	2903	–	–
2	PV module AND recycling NOT thin-film	142	2345	–	–
3	PV recycling AND crystalline silicon	54	1623	–	–
4	PV recycling AND crystalline silicon AND circular economy	1	–	–	–
5	PV recycling AND circular economy	18	–	5100	–
6	PV recycling AND pyrolysis	5	–	–	700
7	PV recycling AND sustainable development goals	4	–	875	–

recovery of materials and minimisation/prevention of hazardous materials must be considered. The RoHS directive has had a positive effect on PV design by lowering the amounts and types of materials used in PV construction regardless of their exemption from the current RoHS 2 directive [39].

PV modules were added to the WEEE directive in 2012. Making it law as of 2014, that the management of PV waste and their EoL treatment was to become the responsibility of manufacturers and suppliers [40, 41]. This decision was made by the European Union Commission and was designed as an effort to limit the negative impacts PV waste was having and the overall waste that would be produced in the future. This would also have a positive impact on e-waste figures and governmental targets if the waste was handled in the correct manner. It also was likely due to the rapid exponential increase in annual installations and data was starting to be drawn on the waste forecast of PV modules.

To be able to pay for PV module recycling, the EU imposes a fee on the module manufacturers that is subsequently passed onto consumers [42]. Annex V of the directive provides minimum recovery targets by category and timeframe. From August 15, 2018, 85% shall be recovered and 75% shall be prepared for re-use and recycling. This is a slight increase from the 2015–2018 target of 80 and 70%, respectively [43].

Currently, Europe is the only jurisdiction that has a correct, clear and strong regulatory framework. However, this is imperfect, as modules that are collected for recycling are then distributed to one of the ~350 sites in Europe [44]. With this, each particular site may not have the same recycling expertise or specification, and so only certain constituents are considered for recycling at certain centres. Some of the other material may be landfilled or transported to other recycling facilities for specialist recycling. This method of shipping certain constituents is not environmentally friendly when consideration is given to the emissions that are produced to transport the material in question from site to site [45].

PV markets that are growing rapidly outside of Europe, such as Japan, China, Australia, India, USA still lack specific PV EoL management regulations [46–50]. As of 2018, the top ten countries for installed capacity of solar PV were China, Japan, USA, Germany, India, Italy, United Kingdom, Australia, France and the Republic of Korea. For example, China has become the country with the highest installed capacity of solar PV, having approximately three times higher the installed capacity (175,016 MW) than that of its closest competitor of Japan at 55,500 MW [51]. In these regions, PV modules are included in a general recycling regulatory framework for hazardous and non-hazardous solid waste and are considered as generic e-waste [52]. The lack of regulations, poor environmental legislation and infrastructure can be dangerous as these countries could be potentially targeted for PV waste exportation, which should be considered worse than landfilling.

This shows that Europe is at the front of the PV module waste issue and is committed to adhering to the Sustainable Development Goals (SDGs). However, there is still significant research and development to be undertaken to determine the optimum way to recycle EoL PV

modules as currently, approximately only 10% of the total PV modules worldwide are recycled [52].

3.2. Scale of the PV waste problem

As shown in Fig. 2, the 2016 International Renewable Energy Agency (IRENA) solar PV end-of-life management report estimated that by 2030, there would be between 1.7 and 8 million tonnes of PV module waste in circulation on the recycling market, with an increase to 60–78 million tonnes by 2050 [46]. This near ten-fold increase over the space of a 20 year period is largely down to the exponential increase in annual installations, production of solar farms and the integration of solar modules into existing or new building architecture (BIPV). With this, a proportional exponential increase in future PV waste is apparent even if such waste appears after a long design life of 20–25 years [53].

One such problem that will occur in the future with the handling and characterisation of PV module waste is that over time, the industry has developed PV design through continual technological developments and innovations. First and second-generation PV are readily available to consumers. Third-generation PV is very much still in its infancy and has yet to reach the market with a product.

As PV design changes then PV waste streams will also change in terms of quantities of constituents over time. This will only affect the ratio or amounts of the constituents and not necessarily the constituents themselves, as most of the material that makes up a c-Si PV module have remained similar over their entire technology lifetime.

First-generation PV which is subdivided between monocrystalline silicon and multicrystalline silicon has averaged a market share of between 80 and 90% over a 36 year period from 1980 to 2016 as shown in Fig. 3 [47,54]. This means that of the PV waste that will occur in the future, the majority of this particular waste stream will be comprised of c-Si PV technology and the constituents associated with this type of module. There was a decline in the market share of the first-generation technology in the late 1980s with the introduction and increased affordability of thin-film technology. However, c-Si PV modules rose again in market share and have never lost their dominance, currently holding greater than 90% of the PV market.

4. Design of crystalline silicon (c-Si) PV

4.1. First generation makeup

Although monocrystalline and polycrystalline PV modules are considered as two separate types of PV modules, the manufacturing process is the same and the only constituent of the module that differs between the two is the silicon substrate that makes up the wafer. Silicon accounts for well over 90% of all semiconductors and solar cell wafer production worldwide [55]. The two types of silicon are separated according to crystallinity and crystal size that is present in the ingot prior to the wafering process [56].

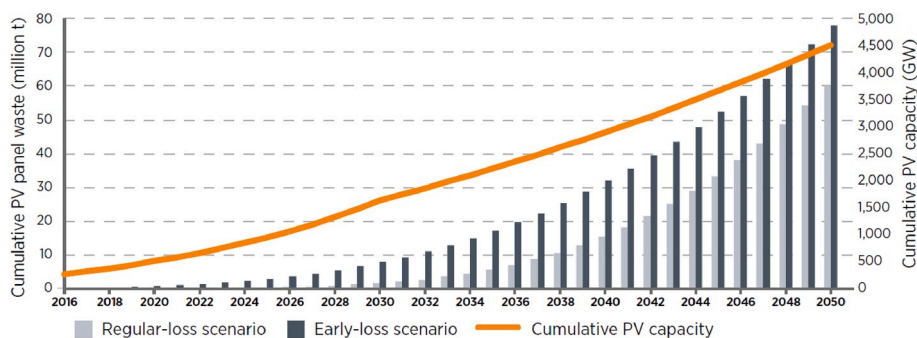


Fig. 2. Estimated PV module waste reprinted from Ref. [27] with permission from A. Kumar et al., “E-waste: An overview on generation, collection, legislation and recycling practices”, Resources, Conservation and Recycling, 122, 32–42, Elsevier, 2017.

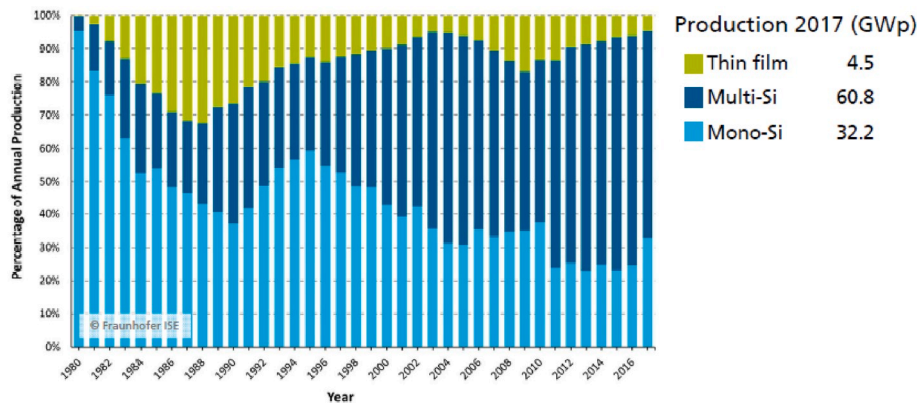


Fig. 3. Market share of first and second-generation PV modules over a 36 year period from 1980 to 2016. Reprinted from Ref. [3] with permission from M. Zentgraf et al., “©Fraunhofer ISE: Photovoltaics Report, updated: March 14, 2019”.

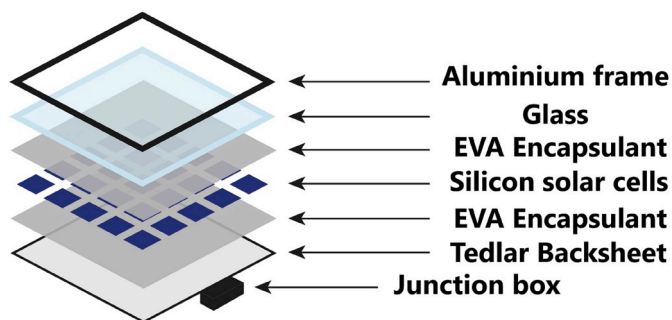


Fig. 4. An exploded diagram of a conventional c-Si PV module.

The layers that make up a c-Si PV module in order of mass are as follows: glass, an exterior aluminium frame, two layers of Ethylene-vinyl acetate (EVA) both, top and bottom of the silicon solar cells that encapsulate the cells, a junction box and PV backsheet (usually made from Tedlar) located at the rear of the module [57]. Standard module construction utilising EVA aims to enhance impregnability by protecting the module from outside sources such as moisture ingress, foreign impurities and physical damage, whilst also acting as an electrical pottant for the cells [58]. Additionally, the use of EVA aims to boost the longevity of the module, hence the 20–25 year manufacturer’s guarantee/warranty [59].

Typically, PV backsheets appear white in appearance due to the diffusion and reflectivity of light with these structures. This helps guide the light that happens to reach inactive areas back to the solar cell to increase the electrical efficiency and power output of the module [60].

There are two conventional types of Tedlar backsheets; TPE and TPT, that constitute a market share of approximately 80% of the c-Si PV modules on the market [61]. Farrell et al. [62] have recently reported that Tedlar backsheets will represent the biggest share of the decommissioned modules until 25 years after a new industry standard is potentially established. This further reiterates the need for policy-makers, industry and researchers to focus on this composition for maximum recovery from what will be an impending waste stream.

PV modules consist of these subsequent layers laminated into a very thin structure approximately 4 mm in thickness. This does not include the aluminium frame, as when this is attached to the core laminate, it makes the overall thickness in the range of 25–38 mm. For more information, an exploded diagram on a c-Si PV module’s construction is shown in Fig. 4 [63]. A conventional household PV module contains 60 individual solar cells compared to the commercial counterpart of 72 cells, and these are connected in series using flattened copper wires dipped in molten solder material [64].

EVA is used in a wide range of industrial applications such as cable insulation as well as the most common PV module encapsulation material [65]. It has been the industry standard of PV encapsulants since the 1980s [66,67] and according to the current 2019 ITRPV report [68], it is unlikely to lose its dominance as the standard encapsulant. Additionally, due to the excellent adhesion properties that EVA possesses towards the glass and backsheet layers, it has been reported as the most challenging step in the recycling of c-Si PV modules [44]. For the delamination process, Dias et al. [63] have reported that the removal of the EVA encapsulant should be the first step.

In order for a solar cell to be fabricated, the first step is the manufacture of the silicon substrate from the ingots. Silicon is rarely found in the pure free element form and is usually found in sands and dust [69]. The mining of silicon for the PV industry and the requirement of high-purity silicon is of high energy consumption and creates heavy pollution (usually in the form of fossil fuels) [70,71]. Moen et al. [69] have reported that approximately 16% of the world’s supply of silicon is used for PV and that the energy consumption for solar and electronic-grade silicon (6 N) from metallurgical grade is between 50 and 100 kWh.kg⁻¹. Additionally, PV Cycle has claimed in a previous study that recycling one tonne of PV waste equates to an 800 kg of CO₂ equivalent and savings of 8.5 GJ of energy from non-renewable energy sources [72].

Conventionally, the silicon substrate is p-type by default. A typical silicon PV cell is a thin wafer consisting of a very thin layer of phosphorus-doped (n-type) silicon on top of a thicker layer of boron-doped (p-type) silicon [73]. The silicon is initially doped by group III elements (such as boron) to create the p-type substrate. The three valence electrons on the outer shell of boron accept a fourth electron from silicon which creates a hole where the electron was originally located in silicon without producing a free electron. After the acceptance of the electron, boron becomes negatively ionised and cannot receive any more electrons [74].

After the doping of silicon, the ingots are created and sawed to manufacture the substrate. During this period, there are significant losses due to the sawing process. Only 45–50% of the silicon feedstock ends up in a wafer. The remaining fraction is lost in the ingot cutting process (tops, tails, and slabs) and the biggest portion of the material is lost as sawing slurry. It was reported by Dong et al. [75] that at least 100 MW h of electrical energy were required to produce 1 tonne of all silicon ingots.

The n-type doping occurs when a group V element (such as phosphorous) occupies a site in silicon and only four of the five electrons in the outer shell are needed to satisfy the bonding criteria. The energy required to release this free electron is very small. After the phosphorous has donated this electron, it cannot lose any more electrons and thus becomes a positive ion. As the ionisation has occurred, a conduction

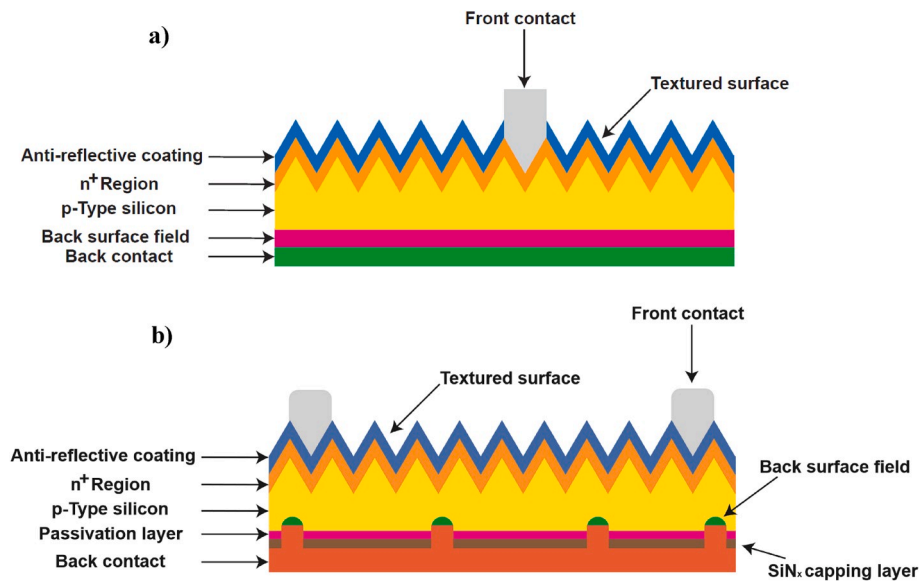


Fig. 5. Cell structure of a conventional a) Al BSF cell and b) PERC cell.

electron is made without creating a hole. After the substrate is made, the electrical field (also known as the p-n junction) has to be created by doping [73,76] the p-type substrate with a thin layer of n-type material from a source of phosphorous such as POCl_3 [77]. The emitter diffusion using POCl_3 requires a high-temperature budget as POCl_3 is used in a diffusion furnace at approximately 975°C for up to an hour [78]. The p-n junction is required to allow the cell to emit an electron by obtaining a photon from a light source and allow an electrical current to flow in order to produce power. After the emitter diffusion has occurred and prior to the anti-reflective coating being added to the cell, phosphosilicate glass is formed all around the surface of the silicon substrate [79]. The purpose of the anti-reflective coating is to increase optical absorption by allowing more incident light to reach the cell and not be reflected from the surface of the silicon wafer and thus increase efficiency [79].

Aluminium back surface field (Al-BSF) cells are considered to be the most commercially produced cell technology [80]. However, in recent years there have been other cell technologies that have started to gain popularity. The back surface field is screen printed to apply back surface passivation. This method is the most widely used technology due to its simplicity, low-cost and high throughput capability for PV manufacturers [81].

Contact metallization and anti-reflective coatings are applied at a later stage of the fabrication phase to allow the manufacture of a conventional c-Si Al-BSF cell type which is shown in Fig. 5a [82].

Although Al-BSF is the current industry standard now, it is estimated

that the passivated emitter and rear cell (PERC) market share will increase significantly in the next decade [68] and will eventually become the new industry standard. According to the international technology roadmap for photovoltaics, Al-BSF in 2017 represented approximately 75% of the market share with PERC representing 20%. In 2022, it is expected that PERC will become the dominant cell type with approximately 50% of the market share, as opposed to 33% that is predicted for Al-BSF [61]. This indicates that these two cell types should take priority when being considered for circular opportunities and recycling processes as they will constitute the majority of this waste stream now and in the future. The PERC cell allows the rear contact to the substrate through holes in the rear passivating oxide. The substrate has to be heavily doped for contact resistance between the silicon and metal. PERC will likely be the ideal solution if wafer thickness continues to drop to under $150\ \mu\text{m}$ [83,84]. The PERC cell type is shown in Fig. 5b.

4.2. Weight percentages of constituents in c-Si PV module

From Table 2, it can be clearly seen that of all the constituents used in PV modules, most have remained relatively consistent in terms of mass percentage regardless of year or source. The constituents that have experienced the most significant change in mass is the aluminium frame and the glass fraction of the module. This is interesting because majority of the rare and precious metals involve the solar cell and the contacts used. Industry, researchers, and policymakers can use these relatively

Table 2
Weight percentages of each constituent in c-Si PV module.

Components	Ref [85] wt. %	Ref [86] wt. %	Ref [87] wt. %	Ref [59] wt. %	Ref [88] wt. %	Ref [89] wt. %	Ref [90] wt. %
Glass	70.00	74.16	74.16	56.65	80.10	74.00	67.40
Frame	18.00	10.30	10.30	18.50	9.80	10.00	15.80
Encapsulant	5.10	6.55	6.55	14.45	Not specified	–	7.40
Silicon cell	3.65	3.48	3.48	9.83	4.70	3.00	3.10
Backsheet	1.50	3.60	3.60	0.58	4.30	–	3.70
Cables	1.00	–	–	–	–	–	–
Aluminium conductor	0.53	–	–	–	–	–	–
Copper	0.11	–	–	–	–	–	–
Silver	0.05	–	–	–	–	–	–
Tin & lead	0.05	–	–	–	–	–	–
Contacts	–	0.75	–	–	–	–	–
Adhesives	–	1.16	–	–	–	–	–
Polymers	–	–	–	–	–	6.50	–
Ribbon	–	–	–	–	–	–	1.00
Junction box	–	–	–	–	–	–	1.60

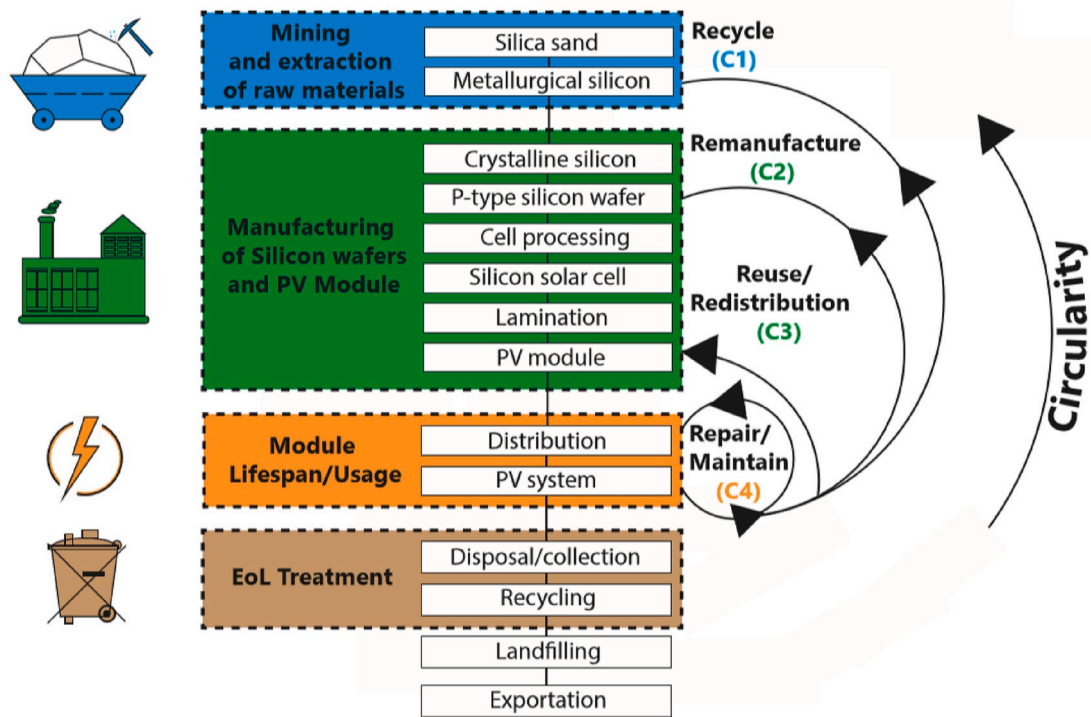


Fig. 6. Butterfly diagram showing the life cycle of a c-Si PV module from the extraction of raw materials to EoL management.

static values of the laminate (EVA, backsheet, silicon) to model, predict, calculate and simulate the outputs and the weight of the constituents that may be recovered in any given recycling process.

When considering the smallest reported constituent, silver at 0.05 wt % [85], it is important to consider this against the waste forecast of 60–78 million tonnes in 2050, as this equates to a potential source of silver at 3–3.9 million tonnes. This can also be extended to the other constituents in PV modules.

Liu et al. [91] have reported that of two separate PV systems in two separate demographic regions 74–75% of the lifecycle energy use of a PV system is in the module manufacturing stage. This further supports the fact that if silicon wafers are able to be recycled from PV modules, then every effort must be made as the manufacturing process has high energy requirements and produces emissions that could be offset with a change in recycling methodology. Furthermore, Huang et al. [92] have reported five main manufacturing steps which include: industrial silicon smelting, polysilicon production, ingot casting & wafer slicing, cell processing and module assembly. Of these steps, polysilicon production, cell processing, and module assembly had the highest environmental impact of all the processes, with climate change caused by carbon dioxide in polysilicon production and cell processing being the significant contributor.

5. PV recycling processes

Outlined in this section is a mixture of recycling methodologies for PV module waste. The methods are further explained using Fig. 6, a “butterfly diagram” of circular opportunities that arise within the PV industry and its lifecycle from the extraction of raw materials up to EoL treatment. The outer circles focus on recycling secondary raw materials with the inner circles focusing on re-use and repair of products. The cascades are labelled C1–C4 ranging from the least circular to most circular process. Due to the PV design method of lamination and encapsulation, opportunities for repair and maintenance, re-use and remanufacturing of PV cells and associated secondary materials without aggressive processing is very limited. This integrated design means circular economy opportunities for current PV cells generally begins

within the centre of the waste hierarchy at the recycling/remanufacturing stage (Fig. 1). In order to gain maximum value from any recycling process, it is necessary to produce a high-quality recyclate which is or can become a high quality and valuable product. Closed-loop recycling attempts to recover products and secondary materials that can be fed into the supply chain of the original product. In this case, materials and products such as the silicon wafers being recovered and re-used in the manufacture of new PV cells. Open-loop recycling happens when the quality of materials isn't good enough for the original product but may still have a lesser value and use within other industries. This loss of value but still retaining a use as a product post-processing can be considered a recycling “cascade”, where a material may have several design lives and opportunities for recovery in sequentially less valuable products. At present, there are three families of processes that are used either alone or in conjunction with one another to recycle PV cells. These are physical, chemical and thermal processes. How these processes are applied (and the order they are applied) will have an effect on the quality and value of materials recovered.

5.1. Physical processes

Conventionally, PV modules are recycled via physical treatment such as crushing and then milling. This is represented by cascade C1 in Fig. 6. This mechanical method is ideal for high throughput and manageable with current PV waste flows but disregards the energy-intensive process requirements needed to form the silicon ingot, wafer, cell, and module, respectively. Additionally, the emissions that are produced in these manufacturing steps are not considered. An example of physical processing of PV cells is that of Maltha glass recycling based in Belgium and the steps are as follows:

- Manual removal of the aluminium frame and junction box.
- Shred the module.
- Recycle the glass layer.
- Pre-sort the shredded material.
- Crush the laminate layers.
- Separation and extraction.

- Recovered materials sent to respective recyclers.

This aggressive physical and mechanical process has drawbacks. It produces products that are of lower value and use compared to the function of the original PV module. It disregards the fact of a large amount of energy is required to arrange the silicon and wafer in the original module. Crushing this constituent down means that the energy that was required at initial fabrication is again required if an attempt is to be made to refashion the material into a PV cell. It also makes the crushed silicon less valuable as silicon is more valuable in the wafer form than in the form of the raw material [61]. The outputs of this process are secondary raw materials and not final products.

Recently, Zhang et al. [93] published work relating to recycling WEEE. They stressed a well organised and dedicated process chain for WEEE is crucial and stated the recycling efficiency for WEEE is a product of the collection, dismantling, pre-processing and recovery percentages. In order to achieve high efficiency, the recycling rate of these four processes all has to be high.

The German recycler Impulstec GmbH, allows selective separation and disassembly of PV modules using a continuous shock wave recycling system they manufacture called EHF 400. The module is mechanically pre-treated and cut to smaller sizes prior to the shock wave treatment. This can treat between 75 and 200 kg.h⁻¹, which corresponds to approximately 4–10 standard 60 cell c-Si PV modules [94]. The separation occurs at the interface between the front glass, EVA and silicon layers when a voltage of between 30 and 50 kV is used. The high yield recovery of the selected constituents is based on figures quoted shown below [95]:

- Glass (73/74 wt%)
- EVA (7/8 wt%)
- Silicon (2.9/3 wt%)
- Copper (1/1 wt%)

Of the constituent materials (glass, EVA, silicon, copper), silicon requires the highest amount of pulse treatments (>250) in order to achieve over 90% recovery. Again, when considering this recycling method, there is a high yield for the constituents recovered. However, in order to achieve such high recovery, there is substantial energy required for the cutting and shockwave processes.

For example, Hahne et al. [89] reported the ecological efficiency of different PV recycling processes varies significantly. The shredding type of recycling that is still currently the industrial norm is relatively low cost, but is not suitable long term and has an environmental impact when considering the re-processing requirements in order to remelt silicon, wafering and cell production.

This effectively downcycles the selected fractions recovered from the module. Additionally, in order for silicon to be used in a closed-loop process to produce new wafers and cells, there is further refinement and smelting required to remove impurities and allow the construction of a silicon ingot once again.

5.2. Physical and thermal process

Granata et al. [96] reported the recovery of multiple types of PV modules (c-Si, a-Si and CdTe) using physical methods. They tested two routes where one involved two-blade rotors crushing followed by hammer crushing and the other involved two-blade rotors crushing followed by thermal treatment of which the atmosphere was unspecified. They found that the two-blade rotors crushing, followed by hammer crushing were the ideal method for the highest mass recovery. However, for the three types to be processed concurrently, the following process would be suitable:

- Crushing by two-blade rotors crusher
- Hammer crushing

- Thermal treatment (650 °C) of fractions larger than 1 mm
- Sieving by a d = 0.08 mm sieve

This would then allow around 85% of the weight of the total panel to be recovered as glass from fractions of d > 0.08 mm.

5.3. Thermo-chemical process

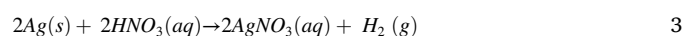
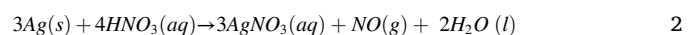
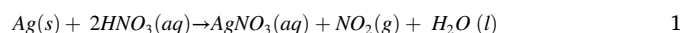
One particular industrial-scale recycling system was developed by Deutsche solar [97]. The recycling plant focuses on the recovery of the wafer substrate from c-Si modules. The process initially starts with the burning of the EVA encapsulant. This will promote chemical oxidation of the other constituents found inside the module and so an etching phase is required shortly after for the removal of the metallization, anti-reflective coating and pn-junction as shown in Fig. 5. The clean p-type substrate is the final product which can be integrated back into the standard solar cell production line again and placed into a new PV module which is represented by cascade C2 in Fig. 6. The direct incineration of EVA is problematic in that it will promote the production of CO₂. A superior thermochemical conversion method to decompose EVA is pyrolysis in that it does not produce as much CO₂ as combustion and that it allows the clean recovery of the module constituents without promoting any chemical oxidation.

6. Chemical recovery of c-Si PV secondary raw materials

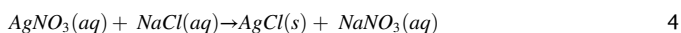
None of the processes reviewed in the previous section are perfect with many producing low-quality recyclate that could be destined for disposal in a landfill. Even processes that produce high-quality products and materials will not be 100% efficient, and rejected materials will have to be managed properly. Opportunities to increase the value of these secondary raw materials through upcycling of low-quality recyclate are reviewed.

Herein, potential chemical recycling routes of various valuable materials with the PV panels will be evaluated, such as silver (Ag), copper (Cu), aluminium (Al) and silicon (Si). The valuable elements by quantities are the first four metals (Si, Al, Ag, and Cu). Al in the form of the exterior aluminium frame is usually separated using mechanical means from the PV module while Cu, Ag and Si either thermally or chemically [98], along with the release of hazardous heavy metals such as lead (Pb). The relative mass fractions for the valuable metals in a typical standard c-Si PV module are 10.3, 3.48, 0.57, 0.01 and 0.12 wt% for Al frame, Si wafer, Cu, Ag, and Sn, respectively [98]. It was reported that a typical 60-cell Si module could make a total revenue of \$16–17, which is more than enough to cover the recycling cost and keep it a profitable recycling business without any support from the government [42,98]. Usually, the extraction of the metal is performed via three main routes; precipitation, electrolysis and metal replacements methods [42,99–101]. Ag production peak will be reached by 2030, while by 2075, the future of Ag production will be at risk [102]. Dias et al. [102] studied the Ag extraction from PV modules with and without pyrolysis and found out that the extraction method without pyrolysis showed a slightly better yield than that with pyrolysis (approximately 2%, which could be due to experimental error). They milled, sieved and leached the module in HNO₃ (64%) then precipitated the solution using NaCl (99%) which led to 94% of Ag yield (630 g t⁻¹ of Ag per module) as shown in Equations (1)–(4).

The leaching process leads to the generation of toxic nitrogen oxides as shown in Equations (1) and (2).



The precipitation reaction occurs as in Equation 4



While in the pyrolysis method, the module was pyrolysed first at 500 °C, then leached and precipitated exactly as shown in the first method, wherein the Ag yield was 92%. Łażewska et al. [103] showed that Ag can be recovered from the etching solution from acidic and basic etching of metal contacts that showed 1600 g t⁻¹ of Ag per broken solar cells. Lee et al. [100] reported a leaching process using various leaching agents (HNO₃, H₂SO₄, HCl and NaOH) to extract Ag and Al from scrap silicon solar battery cell. They found out that Ag was totally leached with 6 N HNO₃, while Al was totally leached with 18 N H₂SO₄ acid at 70 °C. Al was recovered using a crystallisation method, while Ag was recovered using precipitation, electrolysis and replacement methods, with a Ag yield of 98–100% [100]. However, it is doubtful that the chemical treatment becomes a viable industrial process because of the evolved hazardous emissions during the recycling process along with the large quantities of chemicals needed herein. Alongside this, the chemical treatment method would require the collection and disposal of hazardous chemical wastes. This directly contributes to emissions and can not be seen as a circular or as a sustainable process.

Yang et al. [104] recently tried to promote a more circular approach for the recovery of Ag using methanesulfonic acid, a more environmentally friendly option than using inorganic acids as it does not generate the same waste acid solutions and toxic fumes. They determined that an oxidising agent was required to dissolve the Ag from the solar cells and increasing the amount of this oxidising agent would increase the decomposition. In their case they used H₂O₂, however, this led to another problem in that water was produced as a by-product and kept diluting their mixture and thus, rate. The purity of Ag was quite low at 99.8% but could be improved up to 99.995% using electrorefining methods; decreasing impurities like tin and lead.

Lee et al. [105] considered the recovery of copper and lead-tin solder alloy from spent PV modules. They conducted the experiment at 700, 800 and 900 °C under reductive conditions using CH₄ gas and hold times of 30 and 60 min, respectively. At higher temperatures, the fluidity of the lead-tin coating increased, which made the separation of copper easier. ICP-MS analysis of the recovered copper fraction showed copper content of 97%. The recovered coating layer showed 69, 27.9 and 2.9% of copper, tin and lead, respectively. With further refinement of the copper fraction, it is the author's belief this would suit open-loop cascading methods in which copper can be used as a precursor for a value-added product. However, the high temperatures required for this process are not circular when you consider it is only recovering partial constituents of the module and not intended for re-use in newer modules.

Basic solutions such as KOH and NaOH were used to remove the Al layer, where the optimum condition was obtained by using 30% aqueous KOH at a temperature range of 60–80 °C within 2–3 min. On the other hand, a mixture of acidic solutions was used to recover Ag along with the anti-reflective layer (HNO₃ (65%), HF (40%), CH₃COOH (99.5%) and Br₂) [57,106]. Palitzsch and Loser proposed the preparation method of poly-aluminium-hydroxide-chloride [Al_n(OH)_mCl_{3n-m}] which is used in wastewater treatment and the paper industry from the de-metallization method using AlCl₃ solution [107]. Jung et al. [101] proposed a detailed process to recover Si, Cu, Ag, and Pb metals. Firstly, they recovered the Al via two steps; thermally using detachment step then precipitate the Al using KOH solution as gibbsite Al (OH)₃ (Equation (5)) and finally the calcination at 1200 °C for 3 h to prepare aluminium oxide Al₂O₃ (Equation (6)) with a recovery rate of 94%.



After removing the Al electrode and Si_nx layer, the Si was recovered with 80% recovery rate [101]. Afterwards, to the leaching solution containing Cu, Ag, and Pb, 2-hydroxy-5-nonylacetophenone oxime was

added to extract Cu, which is then stripped using H₂SO₄ solution as CuSO₄ solution. Finally, an electrowinning method was used to recover Cu with a recovery rate of 79%. Then HCl was added to precipitate Ag from the leaching solution containing Ag and Pb as AgCl precipitate which is then purified with a recovery rate of 90%. Finally, Pb was recovered using NaOH solution by precipitation in the form of Pb (OH)₂ with a recovery rate of 93% [101].

Kang et al. [108] investigated the recovery of glass and silicon from PV modules using three separate methods. First, the recovery of glass using organic solvents which caused swelling similar to the work conducted by Doi et al. [36]. The second step involved thermal treatment to remove all of the encapsulant fraction. The final step involved chemical etching with the inclusion of 20 wt% surfactant for 20 min and the recovery of 86% silicon. The yield of pure silicon was 99.999% which corresponds to five-nine purity (5 N).

Park et al. [109] obtained crystalline solar cells from EoL modules that were delaminated via means of an undisclosed thermal process, likely to be combustion due to the usage of etchants in the study. They tested two separate etching mixes on the cells to remove metal electrodes, ARC, emitter layer and p-n junctions. The first etchant mix of HNO₃ + HF and KOH resulted in deep grooves of approximately 36 μm due to different etching rates of the Ag electrodes and the Si_nx ARC on the front surface of the recycled wafers. This is not ideal as in this case, they used 180 μm thick silicon wafers. This would be unusable in the construction of new cells due to imminent cracking. The second etching process, however, resulted in minor grooves of approximately 7 μm. This mix was based on H₃PO₄ to etch the ARC and Al back contact and HF + HNO₃ to etch the Ag electrodes, emitter layer, and the p-n junction.

Dias et al. [110] conducted a characterisation and etching study in which they knife milled two separate modules and sieved the resultant powder based on particle size prior to etching in an aqua regia solution (200 ml aqua regia and 20 g of material) consisting of a 3:1 (HCl, HNO₃) ratio for 2 h at 60 °C. Also conducted in this work was the removal of the adhesive layer using two methods. The first consisted of using 100 ml of 40% Hydrofluoric acid (HF) and left immersed for five days. The second consisted of using 100 ml 98% H₂SO₄ and left immersed for five days. For HF, the adhesive layer was not removed or decomposed in any of the particle size groups. Instead, there was a partial dissolution of the glass layer observed. For H₂SO₄ the adhesive had appeared to be broken for all size groups. The larger particle size fraction showed better results were more silicon had detached from the glass layer. It is the author's view that the use of hazardous chemicals like HF poses more of a risk to human health and incurs a disposal cost for the recycler. The pre-treatment using the knife mill requires additional energy needed for the recycling of the modules and the destruction of the silicon wafer means that further refinement, purification and smelting is needed in order to re-use this silicon in new PV modules. This method disregards the energy requirements and the emissions produced in the production of silicon wafers and the additional steps required to produce the solar cells. This can be interpreted as a non-circular method to the recycling of PV modules.

The recycling of aluminium scrap waste such as aluminium foil waste into mesoporous γ-Al₂O₃ (catalyst/support) using a cost-effective green synthetic route is highly desirable. Recently, Osman et al. [111,112] reported an eco-friendly and less expensive synthesis of mesoporous γ-Al₂O₃ from aluminium foil waste where the produced γ-Al₂O₃ catalyst had surface and bulk characteristics better than commercial alumina in relation to the surface structure. The authors believe this approach could be applied to c-Si PV modules, due to the sheer quantity of aluminium it contains (both in the frame and also at a cell level), as well as reducing the need for landfill of aluminium waste materials. Additional chemical treatments of metals analogous to PV module constituents are outlined in Table S1 in the supplementary information.

Abu Bakar et al. [113] prepared porous silicon from silicon powder using chemical etching by mixing HF:HNO₃:H₂O with a ratio of (1:5:10 v/v) as the chemical etchant. They found out that the porosity and the

surface area increased with increasing the etching time (24 h) and the Si:etchant ratio (1:64 w/v). The produced value-added product such as porous silica was extensively used in the environmental catalysis of removing pollutants shown in Table S1. Recently, the silicon content (5.9%) within the lignocellulosic biomass ash was used as a heavy metal removal with a high removal capacity [114]. Additionally, scrap silicon could be used from broken wafers could be used in this way also.

Azeumo et al. [115] investigated the dissolution of EVA using eight solvents (water, toluene, xylene, 2,4-trimethylpentane, n-heptane, and N,N-dimethylformamide), where toluene showed the maximum detachment among the six solvents. Consequently, toluene was used as a solvent to optimise the process and found out that the optimal conditions were a temperature of 60 °C, residence time of <60 min, no thermal pre-treatment and with using ultrasound [115]. Although in some cases, the organic solvents could not remove the swollen EVA remaining on the surface, thus, additional thermal treatment such as pyrolysis is needed herein [108,116].

It is in the author's belief that as stand-alone processes, the methods outlined in this section utilise large amounts of hazardous chemicals and it could be argued that this is in no way follows the principles of a circular economy. However, if the methods outlined here and in previous sections could be integrated alongside the manufacture of PV modules (where hazardous chemicals are already used), where producers took responsibility for recycling, reusing and maintenance; then there are good arguments for efficiency and the economics of such an approach.

7. Discussion

7.1. Recent advances towards a circular approach

Recently, there have been a number of innovations in maximising the recovery of constituents within PV modules. The prime example would

be that of the silicon wafer where it can be recovered either intact or broken. If it is recovered intact, then certain offsets for the energy consumption and emissions of the silicon production would occur. Maximising recovery helps to steer the PV recycling industry towards a circular model.

Goris et al. [117] reported on a method of separating and recovering the glass, solar cell and backsheet layers from a c-Si module using a heated wire saw at approximately 200 °C. The separation was carried out in two steps: at the temperature when EVA starts to soften, the backsheet is peeled from the laminate and then at a higher temperature when EVA is viscous but not decomposing, the EVA between the glass and cell is separated. Goris has mentioned that an advantage to this method is with the glass separated, the heating required for the cell is reduced, as the heating of the glass layer consumes a lot of energy. However, there is still EVA left on the glass and solar cells with this method. Another step would have to be employed such as pyrolysis or chemical treatment to clean and remove the leftover EVA.

Following the heating wire saw method, the "hot knife" method was reported by Japanese company NPC [118,119]. Their process can separate the PV cells from the glass in approximately 40 s, leaving behind a sheet of cells. The module is placed between two rollers, which move it along and hold it steady until it runs past a heated knife. The knife is a 1 m-long, 1 cm-thick steel blade that is heated to 180–200 °C and slices the cell and the glass apart. Recycling solutions for this process exist in the form of machinery that was recently showcased [120].

One such innovation in the enhanced recovery of the silicon wafer and the minimisation of the damage came from Doi et al. [36]. They had noticed the cell was cracked due to the swelling force of the EVA encapsulant when immersed in a hazardous chemical (trichloroethylene) for several days to delaminate the module. To counteract this they employed the use of a binder clip to provide mechanical pressure and suppress the swelling as shown in Fig. 7a below. The wafer was

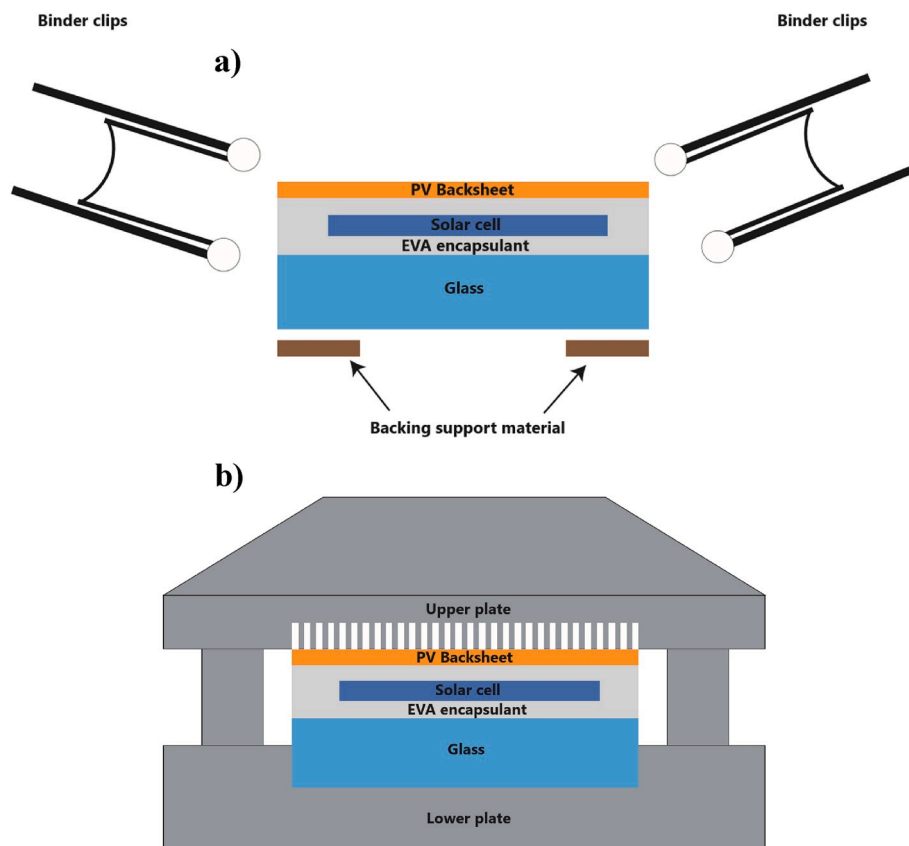


Fig. 7. Enhanced silicon wafer recovery using (a) Mechanical pressure and (b) Mechanical fixture. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

recovered without any noticeable form of damage when subjected to trichloroethylene at 80 °C for 7–10 days.

Recovery of intact solar cells from an EoL PV module to offset energy requirements and emissions associated with silicon and wafer production is extremely difficult. Therefore, an advanced thermal process is required in order to recover the unbroken solar cells [121,122].

Yu et al. [123] suggested that the current delamination methods that were used in already existing recycling processes were unreliable and have the disadvantage of degradation of breaking the photovoltaic cells. Yu opted for a hotwire cutting method that allowed more room for EVA swelling post chemical or thermal treatments. This subsequently meant that cracks and breakages in the cells were reduced. This was, however, before Park et al. [99] had established and published work using a mechanical fixture. The fixture was utilised to apply a light load to a PV cell to suppress some of the swelling and thermal stresses as shown in Fig. 7b. The gaps in the top plate were designed to facilitate the diffusion of gaseous product forming from EVA decomposition. Using a temperature of 480 °C, a ramp rate of 15 °C.min⁻¹ and the mechanical fixture ensured a 100% Si ratio.

One way in which to facilitate module delamination and the removal of the EVA encapsulation is pyrolysis. This involves heating the PV module in an inert environment (such as nitrogen or argon) under a strict heating regime so as to control the thermal stresses that occur in the module. Pyrolysis has the advantage of not promoting any chemical oxidation to any of the module constituents to help aid delamination and subsequently, recovery of the leftover constituents such as the cell, metal contacts, and glass. Therefore, it is proposed that pyrolysis could contribute positively to recycling rates and economics via means of tertiary recycling by processing the waste polymers into a potential fuel source [124–126] that could be utilised to delaminate further PV modules or used for an energy application on-site and thus facilitating the circular economy for the other PV module constituents.

To date, there are limited pyrolysis studies on PV modules in the literature; though all of the experimental findings have been positive. One pyrolysis study conducted by Zeng et al. [127] acted as the basis for a general reaction mechanism and basic kinetics of EVA decomposition which would mimic module delamination in a real-world scenario. Several atmospheric conditions and heating rates were tested to give meaningful insight into how EVA pyrolysis may occur in PV modules and help the industry by informing them of this promising route.

Some gaps are present however in existing research. For example, the lack of understanding on the full reaction mechanism of general degradation and pyrolysis of EVA is problematic when considering the construction of a process model to help aid the problem of PV module recycling at scale. Additionally, there are gaps in how the heating rate and light load affects the decomposition and delamination of the module. There are conclusions made from previous studies, but there is limited data for both cellular and module-level on what weight should be applied to allow the recovery of an intact silicon wafer.

Radziemska et al. [128] have reported previously that the thermal process allows fast, simple and economically efficient module separation. Additionally, they stated thermal degradation of EVA and subsequent separation of the module is, from an economical and ecological point of view, a more favourable alternative to chemical separation which requires the use and disposal of expensive and hazardous agents.

7.2. Circular material and energy flow analysis

Based on the literature in the previous sections, Fig. 8 shows four models outlined M1 to M4 when considering the three main phases of a PV module lifecycle (the manufacturing, the lifespan, and the recycling stages) to critical analyse the circularity of the process at hand. The flow of material is designated by black arrows, whereas the flow of energy in b,c is represented by red arrows. Internal arrows within the circle

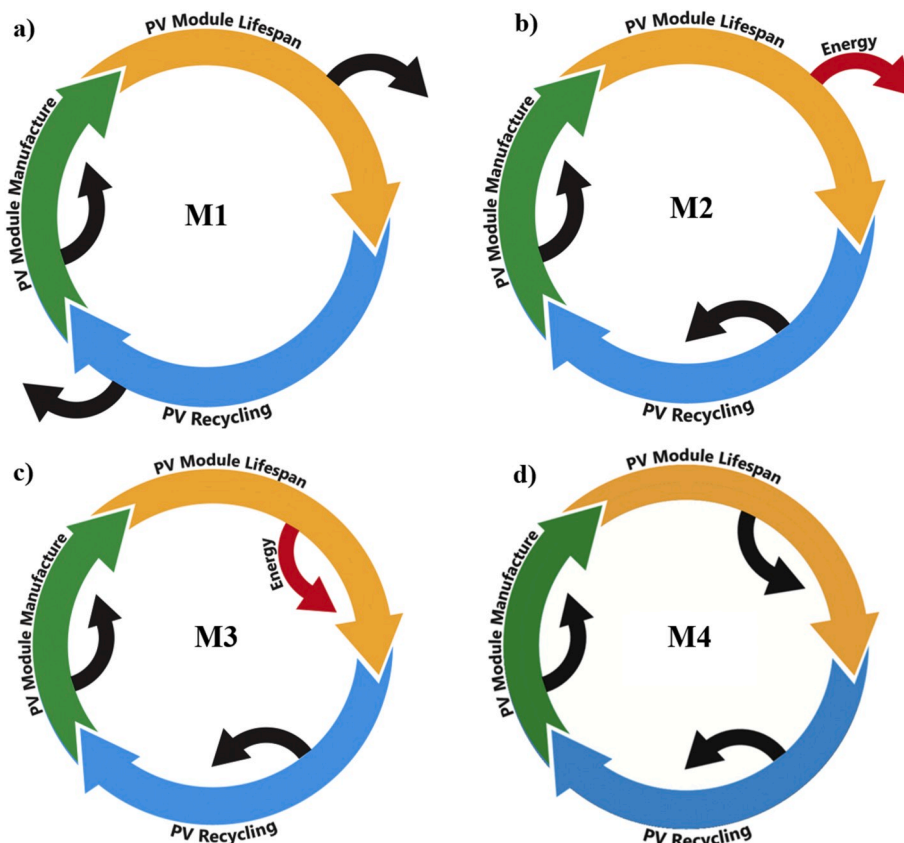


Fig. 8. Material and energy flow analysis of (a) Current PV industry; (b) Open-loop energy recycling; (c) Closed-loop energy recycling; (d) An ideal circular economy.

represent opportunities for closed-loop recycling where the materials can be re-used, recycled and recovered in the production of more PV modules. The external arrows to the circles represent downcycling where materials have a lower value and are not used within the PV production but are used elsewhere.

Fig. 8 shows four models outlined M1 to M4 when considering the three main phases of a PV module lifecycle (the manufacturing, the lifespan, and the recycling stages). The flow of material is designated by black arrows, whereas the flow of energy in b,c is represented by red arrows. Internal arrows within the circle represent opportunities for closed-loop recycling where the materials can be re-used, recycled and recovered in the production of more PV modules. The external arrows to the circles represent downcycling where materials have a lower value and are not used within the PV production but are used elsewhere.

Fig. 8a represents the current PV industry which is a physical and chemically lead process. This relates to the majority of the physical, thermal and chemical processes or treatment trains that have been described thus far. For example, in the module lifespan stage before the PV recycling stage is reached there is material lost in the failure to collect modules for an EoL treatment phase. Also, in the recycling stages, materials like the aluminium frame and glass layers are mechanically removed from the modules to be sold for commodity value or used in general aluminium or float glass recycling and are not incorporated or used in the manufacturing of new PV modules.

For example, in 2007 PV Cycle was initially founded as a voluntary collection and recycling system by the European solar industry as a joint project to research and develop high-quality recycling at an EU level with a drive to recycle 100% and reduce the energy consumption of a manufactured module [89,129].

It now caters heavily towards the WEEE requirement and has provided a benchmark of 96% recycling rate in world performance by weight [130]. The system mainly treats c-Si PV and has released figures of 79.4% feedstock based on c-Si and the remaining based on thin-film technology [44]. However, PV cycle remains a take-back and recycle system unless the consumer possesses more than 30–40 waste PV modules, in which PV cycle will collect the waste [131]. For anything less than 30 PV modules, they suggest that the de-installer transports the modules to the closest collection point. When considering the numerous journeys taken by de-installers and the emissions produced in transporting small numbers of modules each trip, this does not appear as an environmentally friendly system.

The choice to recycle these modules then falls to the consumer, which is alarming as there could be numerous PV modules with an abundance of rare and precious metals that could be neglected or unknown to the local recycling authorities.

Fig. 8b is analogous to M2, a thermally lead process where the energy from the thermochemical conversion process can be recovered and can be stored or used in other applications, whether it be on the recycling site or elsewhere. One such example of this was recently reported by Corcelli et al. [86], in which they refer to a high-rate recovery scenario. The heat produced by the plastics thermal treatment is recovered and then exploited for hot water generation or for heating purpose within the plant where the process takes place. The main constituents such as copper, glass and silicon are recovered through a manual separation after the thermal treatment. The aluminium frame is recovered mechanically prior to the thermal treatment.

Fig. 8c M3 outlines recent work, another thermally lead process and the concept of using energy from thermochemical conversion processes such as pyrolysis to be introduced into the same application of delamination and removal of problematic polymeric material, subsequently delaminating the PV module. The priority main gaseous pyrolysis product can be used in the form of heat recovery or the production of electricity to heat further EoL PV modules that can be delaminated in the same way and thus making the process somewhat self-sustaining [62]. The red energy arrow towards the end of the PV module lifespan indicates the energy recovery from the pyrolysis of the polymers and its

utilisation within the same system to help delaminate further modules.

Olson et al. [54,132] first put the idea into perspective that the silicon wafer and EVA have a minor contribution of approximately 14% to the weight, but account for a large amount of the embodied energy and climate change impact of 83% and 66%, respectively. The embodied energy of 1 kg scrap of a c-Si PV module was reported by Goe et al. [133] to be 78 MJ.m^{-2} . This is more than CdTe & CIGS and more than double the embodied energy compared to its closest counter-part a-Si at 37 MJ.m^{-2} . Of this energy reported, Silicon comprises 55%, next to Aluminium at 27%.

This signifies that it is ideal to recycle both of these constituents from an energy, environmental and subsequently, an economic perspective. The EVA should be recovered thermally due to the imminent degradation and yellowing in the polymer. This leaves behind the wafer and the metallization contacts which can be readily recycled again. To the best of authors' knowledge, there has been no testing on recovered wafers using pyrolysis without the utilisation of etchant chemicals and this can be seen as an avenue for future work.

Fig. 8d represents an ideal circular economy model, as yet this circular model does not exist. The reason for this is not due to constraints around recycling technologies, but rather a fundamental issue around the current design and construction of PV cells. The use of the EVA encapsulant that creates a complex laminate "sandwich-like" structure that can only be disintegrated using thermal methods or aggressive and hazardous chemicals is not good eco-design. The design of PV modules must include design for re-use, recovery and recycling.

With regards to the use of different materials to design out waste and to substitute current constituents for the make-up of a module, Hunt et al. [33] have stated this philosophy is not meant to prohibit the use of elements in current applications, but should be used to promote holistic strategies for extraction, manufacture, utilisation, and recovery of elements. Such activities are vital in order to develop a global sustainable circular economy.

The PV industry, however, prefers to minimise and prevent the amount of material included in the manufacture of a c-Si module [46]. This would instantaneously affect the recycling rates and efficiency as there would be a higher percentage of material recovered. This is limited on how efficient the cutting/sawing technology is that produces the silicon wafers.

When considering the argument for repair, re-use and remanufacture, a referral must be made to the current and previous market share of c-Si PV and the imminent waste flows that will arise in the future. This stresses the urgency of looking at recycling this particular PV technology and ensuring a sustainable recycling system where modules can be rebuilt using some of the constituents from older modules. This would allow a further lifetime guarantee of 20–25 years, making the constituents in use for a total of 40–50 years. Also, it would offset the energy requirements and emissions associated with the production of the silicon ingots, the sawing of the ingots and the manufacturing of the silicon solar cells.

One issue is that the recycled wafers have to meet the quality standards of existing PV modules being manufactured at this given time and perform close to that of a standard PV cell or module [134]. Both Frisson and Park et al. [99,135] previously have shown that recycled wafers have a close match to reference standard wafers in terms of electrical characteristics such as short-circuit density (J_{sc}), open-circuit voltage (V_{oc}), fill factor (FF) and electrical efficiency (Eff.).

Radziemska et al. [136] reported other issues surrounding the recycling of PV modules such as the geographical dispersion of PV module waste, their low content of valuable material (in terms of mass) and the decades-long interval in which modules are installed and discarded. It is vital that the proximity principle outlined by the EU (where waste should be disposed of as close as possible to where it is produced) is met when recycling PV modules [137].

Opportunities for re-use and recovery of PV cells and silicon wafers can be made more circular by incorporating re-use and recovery

methods alongside the production methods in the original PV production process. If PV cells can be designed to allow easy delamination allowing recovery of high-quality silicon wafers, then these could be incorporated directly back into production processes. For example, Frisson et al. [135] reported that the re-use of a recycled wafer improves the energy payback time of a PV module considerably, agreeing with Vellini et al. [138] and that the energy required for a PV module was 9.32 kWh/wafer over its entire lifespan with the majority of the energy coming from the silicon production phase (7.55 kWh/wafer). The recycled wafer, on the other hand, had a requirement of 2.17 kWh/wafer over its entire lifespan. As reported by Fthenakis et al. [139], during the life cycle of PV, emissions to the environment mainly occur from using fossil-fuel-based energy in the extraction or generation of the materials used for PV modules such as silicon, silver, copper, aluminium, and silica. The lower the EPBT, the lower the emissions will be.

This would require the PV producers to take more responsibility for the recycling, reusing and maintenance and to change from a business model of selling a single-use product to one that includes concepts such as collaborative consumption, incentivised returns and performance/service models [140,141]. This systematic and paradigm shift should be considered, as it is vital in reaching a true circular economy for the PV industry.

Having a proper recycling infrastructure and EoL systematic policy for PV modules in place will offer several benefits and sustainable solutions to the problems of resource availability, economic feasibility and EoL environmental impacts and risks [142]. The environmental benefits of recycling are related not only to the limited & decreasing space in landfills and the potential leak of toxic or heavy metals, but also energy savings and reduced emissions. Lowering energy consumption in the manufacturing stage is a strong argument for the recycling of PV modules [143]. This also helps alleviate pressure on the demand for these rare and precious metals for new module production. An increase in the recycling rate of PV modules or its constituents will reduce the life-cycle energy used to mine, refine, treat and manufacture PV material [133].

7.3. SDGs in PV recycling

At an upper level, the circular economy concept can be interpreted as a complementary part of sustainable development and touches on a number of the United Nations SDGs with goal 12 “responsible consumption and production” presenting itself as being the most meaningful [144].

In focusing on the efficient recycling of PV modules, there are ten of the seventeen total SDGs that can be considered relevant [145,146]. Ultimately, this means that if sustainability is achieved in a PV module recycling system, then it has numerous positive impacts on other issues and goals outside of this particular industry also. This could influence or incline other industries to do the same, making the PV industry lead by example.

The framework of how the SDGs work within circular approaches to PVs can be viewed in much the same way as the original waste hierarchy. This is illustrated in Fig. 9 and shows the top of the hierarchy to be ‘responsible consumption and production’ which infers to good design that allows PVs to have a circular lifecycle. This is facilitated by and in turn, facilitates the goals of ‘Industry Innovation and Infrastructure’, ‘Sustainable Cities and Communities’ as well as ‘Decent work and Economic Growth’. The top two levels of the hierarchy presented in Fig. 9 produce the outputs of ‘Good Health and Well Being’, ‘Affordable and Clean Energy’, ‘Climate Action’, ‘Life on Land’ and ‘Life below Water’.

To have such an SDG approach and an effective circular 3R framework again requires ecodesign to be implemented at first instance in the manufacturing of PV modules. The current model of a 20–25 year lifespan using a laminate structure with a strong encapsulant such as EVA is not ideal. However, due to EVA’s properties making it the current industry standard, the industry should consider a change in design to effectively re-use and maintain, even if this promotes a shorter lifespan and collection time. This would ensure that manufacturers have control of the supply chain and are not as affected to price volatility of the rare and precious materials used. This change would help promote the circular economy.

8. Prospective overview and conclusion

Considering the complex issue of PV module recycling, delamination and all of the associated constituents, it is crucial to assess if the current infrastructure and industry norm of recycling modules is in-fact sustainable long-term. To date, little PV waste has appeared, but there is an expectancy of large amounts in the very near future. The method of physical recycling may be suitable in terms of throughput now but fails to consider energy and emissions associated with the initial manufacture, which to date are having to be re-applied to physically treated modules. In addition, it fails to consider circular opportunities within the industry and lacks good ecodesign in which modules could be designed and built with delamination and recovery in mind for direct re-



Fig. 9. Hierarchy of SDGs applicable to the PV industry. Assets used from Ref. [147] with permission from M. Ike et al., “The process of selecting and prioritising corporate sustainability issues: Insights for achieving the Sustainable Development Goals”, Journal of Cleaner Production, 236, Elsevier, 2019.

use of constituents with the exception of EVA as it will likely remain the industry standard encapsulant.

In order to maximise recovery, value and contribute positively towards the circular economy and the environment; a forward-thinking approach to recycling this waste is needed. Ultimately, designing out waste the implementation of good eco-design would help achieve a truly circular economy for the materials used in PV module manufacture. However, the industry may not align with a change in design, and there is still a magnitude of modules that will reach their EoL stage using current design.

The various recycling methodologies of mechanical, chemical and thermal recycling and recovery of material from first-generation (c-Si) PV modules were reviewed and analysed in this work. It is in the authors' conclusion and the main finding of this review that pyrolysis with a focus on energy and silicon wafer recovery and utilisation (outlined in M3), is such a way to recycle current PV modules by delamination with clean results. This is very much dependent on controlling the heating rate and the thermal stresses that will arise in the module. Therefore, it is our recommendation that more experimental work is done in this field to help maximise recovery of full PV cells. Studies in the literature [63, 127] and our previous research [62] have shown that pyrolysis is efficient for removing polymeric material in the form of the EVA encapsulant and majority of the PV backsheets; with less than 1 wt% residual remaining for the EVA fraction and between 20 and 25 wt% residual left for the backsheet fraction.

The EVA encapsulant can be seen to be the most problematic material in PV modules due to its degradation over time. As it degrades, less light is transmitted through to the solar cells and this drops the overall power output of the module. This drop in power explains why the module is classed as EoL. The removal of EVA and the delamination of PV modules has been reported to be the most difficult step to recycle these modules [62].

Therefore, it is our view that pyrolysis not only has the benefit of no chemical oxidation of the constituents but also a relatively clean separation of the subsequent layers without the need for hazardous chemicals. Additionally, pyrolysis can provide energy which can help aid the delamination of further panels and positively affect the economic feasibility of the recycling process. This is particularly important as several studies have shown that the economics of current recycling methodologies are not profitable [148,149]. Although, Corcelli et al. [150] had suggested that attention should be given to more than just financial incentive when assessing environmental performance/benefits and choosing a waste treatment route from the perspective of sustainable development of the PV industry.

In order for the recycling of PV modules to occur at an efficient standard, the PV industry needs to constantly lower the price and incurred cost of recycling as a whole to be sustainable in a competitive recycling and energy market. This can only be done through rapid optimisation of both collection of PV modules and the material recovery stages in the near future [142]. The current method in Europe of delegating constituents to some of the 351 specialist recycling centres with PV cycle is not efficient as it effectively downcycles the PV module by creating a less valuable product in the form of individual constituents or low-quality recyclates such as mixed wastes. This is opposed to a complete and functional PV module which has value in producing renewable energy in the form of electricity, not to mention the emissions associated with the transport of these constituents to recycling centres.

As reported by Park et al. [151] and our previous research [62], there is a possibility of maximising the silicon solar cell recovery intact in the full wafer state, thus offsetting some of the energy requirements and emissions associated with the production of silicon, ingots, and wafers that were discussed. Couple with this the fact that the energy released from the EVA encapsulant fraction has a high calorific value that is relatable to biodiesel and heating oil (39.51 and 39.87 MJ.kg⁻¹ for the aged and virgin grade samples, respectively) [62,152]. This is surplus energy that exceeds the energy required for delamination.

For the current PV design, the possibility of reusing some of the constituents within the modules, whether in the physical or energy form will offset the potential deficit of rare and precious metals and aid sustainability for this renewable energy sector.

Barriers affecting PV recycling activities in the EU should be removed and existing prevention, re-use, recycling, recovery and landfill diversion targets for PV modules should be constantly reviewed so as to move towards a lifecycle-driven "circular" economy, with a cascading use of constituents found in the PV modules and subsequently, any residual waste that may be leftover is tending to and is close to zero by designing out waste with efficient ecodesign that focuses on design for deconstruction and remanufacture (Fig. 8, M4).

The presented information in this study on the recycling of first-generation c-Si PV module waste can be used by academics, industry and policymakers alike, as PV modules are under the legislative framework of the WEEE directive and these targets will constantly change.

Future research agenda in this area should start with looking at the emissions from the relevant polymers found in c-Si PV modules. The emissions study will ensure they are or could be at a suitable level where energy can still be harnessed at industrial scale in a closed or open-loop system. This will allow further delamination of modules or use for other energy applications on site. Additionally, process models can be created from these studies to help determine how the system will react at scale and over time with the development of slight deviations in PV module design as current PV modules reach their EoL stage. This would benefit the PV module supply chain and general availability of the scarce materials these modules contain. From a waste management hierarchy perspective, energy recovery is not the ideal scenario for constituents like EVA, but this will allow other materials like the rare and precious metals to reach higher up the waste management hierarchy with recovery and can remove the problem of EVA waste which is rendered useless due to the degradation over time. This has the potential to be more sustainable and efficient than what is currently done. This would only be the case until the industry is forced to change materials such as EVA and Tedlar backsheets in c-Si PV modules or the implementation of more circular ecodesign for PV modules. Finally, the contextual implication for PV production and manufacture is that there should be an uptake of technologies that promote the remanufacture of PV from current EoL PV stock. Additionally, there should be more of a focus on the design of future PV so that they can be easily reused, recovered and remanufactured.

Declarations of competing interest

The authors declare no conflict of interests.

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Appendix A. Supplementary data

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