Advancements in end-of-life crystalline silicon photovoltaic module recycling: Current state and future prospects

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ABSTRACT

With the rapid development of the photovoltaic industry, it has brought abundant renewable energy to society, but at the same time, it is also accompanied by a series of negative problems. In order to effectively address these issues and reduce the impact of end-of-life photovoltaic modules on ecological environment quality and industry sustainable development, many researchers are dedicated to studying rational recycling and utilization of photovoltaic modules. This study reviews and evaluates the recycling technologies for crystalline silicon photovoltaic modules (c-Si PV modules) proposed in recent years. Based on this, the following five suggestions and prospects are proposed: optimizing recycling strategies; promoting interdisciplinary integration; improving relevant policies and regulations; establishing a complete recycling industry chain; and providing appropriate economic subsidies. It is pointed out that efficient recycling schemes with universal applicability designed for broken c-Si PV modules are more worthy of support, and component sorting and the carbon emissions of recycling processes need to be taken into account. Properly recycle low-value materials and carry out high-value utilization of photovoltaic materials. Emphasize the producer responsibility system and establish admission thresholds and qualification requirements for photovoltaic recycling enterprises. The aim of this study is to provide valuable references for the sustainable development of the photovoltaic recycling field.

1. Introduction

With the depletion of global fossil energy reserves and the shift in policy orientation, the development of clean and renewable energy has become increasingly prominent [1,2]. Solar energy has been widely utilized due to its "inexhaustible" nature, and the most common way to utilize solar energy is to convert it into electricity through photovoltaic modules [3,4]. As of 2022, the contribution of photovoltaic power generation to global electricity generation is 4 %. With the continuous decrease in photovoltaic power generation costs and the continuous increase in installed capacity, China and even the global photovoltaic industry have entered a period of rapid development [5]. According to the reports from the International Energy Agency and the National Energy Administration of China, the global cumulative installed capacity of PV modules reached 1.65 TW in 2023 [6]. China is one of the major contributors to the installed capacity of PV modules, with a new installed capacity of 216.88 GW and a cumulative installed capacity of 610 GW in 2023 [7].

With the large-scale installation of photovoltaic modules, the amount of photovoltaic modules that end of their service life (EoL) is also showing a growing trend [8]. Given that the conventional service life of photovoltaic modules is approximately 25–30 years, those installed in the early 20th century are about to enter a peak period of wasting [9,10]. At the same time, with the steady growth of photovoltaic modules production and the evolution of solar cells manufacturing technology, the consumption rate of raw materials is accelerating, which has raised concerns about the potential depletion of these key materials [11,12]. Hallam et al. [13] and Verlinden et al. [14] pointed out that if the consumption of silver in the photovoltaic manufacturing process is not controlled, the silver reserves will face the risk of being depleted when the photovoltaic module production capacity reaches the TW level. Underwood suggested that under the scenario of widespread electrification in photovoltaic production, abundant materials such as copper, concrete, and aluminum may face shortages [15]. These phenomena not only pose challenges to the treatment and recycling of waste, but also poses new requirements for the sustainable development of the photovoltaic industry. Therefore, it is necessary to conduct in-depth research on the recycling and reuse technology of photovoltaic

modules in order to cope with the upcoming wave of wasting and resource crisis. According to the International Renewable Energy Agency's forecast, 1.7–8 million tons of EoL photovoltaic modules will be generated by 2030, and this number will significantly increase to 60–78 million tons by 2050 [16]. A large amount of discarded photovoltaic modules will bring enormous disposal and recycling pressure to the photovoltaic industry. The panel glass and aluminum frame contribute more than 80 % of the mass composition of photovoltaic modules. In addition, photovoltaic modules contain metals such as copper (Cu), silver (Ag), aluminum (Al), lead (Pb), and tin (Sn), as well as organic polymers such as ethylene vinyl acetate (EVA), ethylene terephthalate (PET), polyvinyl fluoride (PVF), and polyvinylidene fluoride (PVDF), as well as high-purity silicon (Si). Proper disposal and recycling of photovoltaic modules can not only prevent their impact on the ecological environment, but also allow for the secondary use of their constituent materials [17,18].

The disposal and recycling of EoL photovoltaic modules have gradually attracted the attention of governments worldwide. As early as 2012, the European Union defined waste photovoltaic modules as a new type of waste electrical and electronic equipment (WEEE), and introduced a series of mandatory regulations for the recycling of photovoltaic modules [19,20]. The United States, Australia, Japan, South Korea, India and other countries have also proposed waste management methods or recycling guidance for photovoltaic modules [21]. In early 2019, the European continent established the first systematic commercial photovoltaic module recycling facility [21]. Although the disposal and recycling of photovoltaic modules in China started relatively late compared to European countries, its development momentum is rapid. In 2022, China officially implemented the national recommended standard "General technology requirements for photovoltaic module recycling and recovery" (GB/T 39753 - 2021), providing clear technical guidance for the recycling and reuse of photovoltaic modules. In the same year, the China Renewable Resources Industry Technology Innovation Strategic Alliance (CIAR) also launched the development of the standard "Calculation Standard for Carbon Emissions in the Disposal and Utilization of Retired Photovoltaic Modules", marking a solid step for China in the management, disposal, and recycling of discarded photovoltaic modules.

The photovoltaic module market is mainly divided into two categories: crystalline silicon photovoltaic modules (c-Si PV modules) and thin film photovoltaic modules. At present, c-Si PV modules dominate the global installed capacity and market share [22]. Therefore, among wasted photovoltaic modules, c-Si PV modules also occupy the main share [23]. Given their dominant position and potential for environmental impact, research on the disposal and recycling technology of EoL c-Si PV modules is particularly important. This study reviews the recycling technologies for EoL c-Si PV modules proposed by researchers in recent years, mainly including module delamination, high value material recycling, and low value material recycling. On this basis, this study proposes five suggestions and prospects, including optimizing recycling strategies, promoting interdisciplinary integration, improving policies and regulations, establishing a complete recycling industry chain, and providing appropriate economic subsidies. It is particularly emphasized that current recycling technologies should focus on broken and incomplete EoL c-Si PV modules, and it is suggested that low-value materials such as PET in backsheet should also be included in the recycling process. The universal applicability and carbon emissions of the photovoltaic recycling scheme are crucial indicators for assessing its feasibility. Implementing the photovoltaic producer responsibility system and setting admission thresholds and qualification requirements for photovoltaic recycling enterprises is beneficial for the standardization and normalization of photovoltaic recycling development. This aims of this study is to provide clear ideas and valuable references for photovoltaic module recyclers and government departments, and jointly promote the green and sustainable development of the photovoltaic industry.

2. Structure of c-Si PV modules

As shown in Fig. 1, the c-Si PV module has a layered structure, consisting of key components such as tempered glass, EVA, solar cells, EVA, and backsheet. The aluminum frame serves as a support and protective structure, ensuring the mechanical strength and stability of the components. The junction box is installed on the back of the backsheet to connect with external circuits. In c-Si PV modules, the tempered glass on the frontside not only allows sunlight to penetrate, but also provides necessary physical protection. The backsheet, in addition to providing protection, also plays an important role in electrical insulation. Fluorinated polymers are widely used in the manufacture of backsheet due to their resistance to ultraviolet radiation and water vapor, such as: polyvinyl fluoride (PVF, DuPont trade name "Tedlar®") and polyvinylidene fluoride (PVDF, Arkema trade name "Kynar®"). The most classic backsheet material and structure is TPT (Tedlar® Film – PET Film – Tedlar® Film), which is made by pressing three layers of PVF – PET – PVF film. However, over the past decade, the market share of TPT backsheet has gradually declined, and KPK (Kynar® Film – PET Film – Kynar® Film) or KPE (Kynar® Film – PET Film – EVA Film) backsheet made from PVDF film instead of PVF film have become more popular in the market [24]. In recent years, industry has shifted towards using fluorocarbon coatings instead of fluoropolymer to coat PET on both sides or inner layer. Additionally, fluorine-free polymer backsheet such as polyethylene terephthalate core layer (PET backsheets), polyamide core layer (PA backsheets), and polyethylene core layer (PE backsheets) also have certain application markets [25]. EVA, which has good transparency, serves as an encapsulant to bond tempered glass, solar cells, and backsheet together.

As the core of the c-Si PV module, the solar cells have a multi-layer structure, which can be simplified from top to bottom as follows: silver electrode, antireflection layer (Si_3N_4), silicon wafer, and aluminum electrode. Solder strips containing copper, lead, and tin are evenly arranged on all solar cells to integrate and transmit the generated current. Table 1 provides the mass fractions of each component in the c-Si PV module per unit mass. It can be seen that 76 % of the whole c-Si PV module is made of low value glass, while solar cells containing high-value metals and high-purity silicon only account for 4 %. This is one of the reasons why the attractiveness of recycling c-Si PV modules to photovoltaic manufacturers is relatively low.

The unique layered structure of c-Si PV modules makes it extremely challenging to efficiently recover high-value materials from them. Especially for solar cells and solder strips with high economic value, they are tightly wrapped in the core position of the module, further increasing the complexity and difficulty of the recycling process. Therefore, the early disposal of waste c-Si PV modules was either landfilling together with municipal waste, or recycling the aluminum frame and tempered glass at the top through steps such as "disassembly - crushing - mechanical sorting" [8]. The remaining not delaminated EVA, solar cells, and backsheet were discarded or landfilled. In the current era of emphasizing product lifecycle management, from "cradle" to "cradle", simply disposing of EoL c-Si PV modules is clearly no longer a sustainable choice. In order to achieve effective utilization of resources and sustainable development of the environment, effective measures must be taken to promote the resource recycling of EoL c-Si PV modules. Components delamination and solar cells purification are the key points and challenges to be addressed in the resource recovery of EoL c-Si PV modules. The following content elaborates on the relevant technologies around these two points. The author observed that most research focus on the recovery of high-purity silicon wafers, high-value metals (Ag and Al on solar cells, and Cu in solder strips), while the recovery of low-value materials such as EVA and backsheet is often neglected. However, considering that the recycling of these low-value materials also has a positive impact on environmental quality and recycling costs, the author briefly introduces the recycling study of these low-value materials and suggests that these low-value materials can also be appropriately
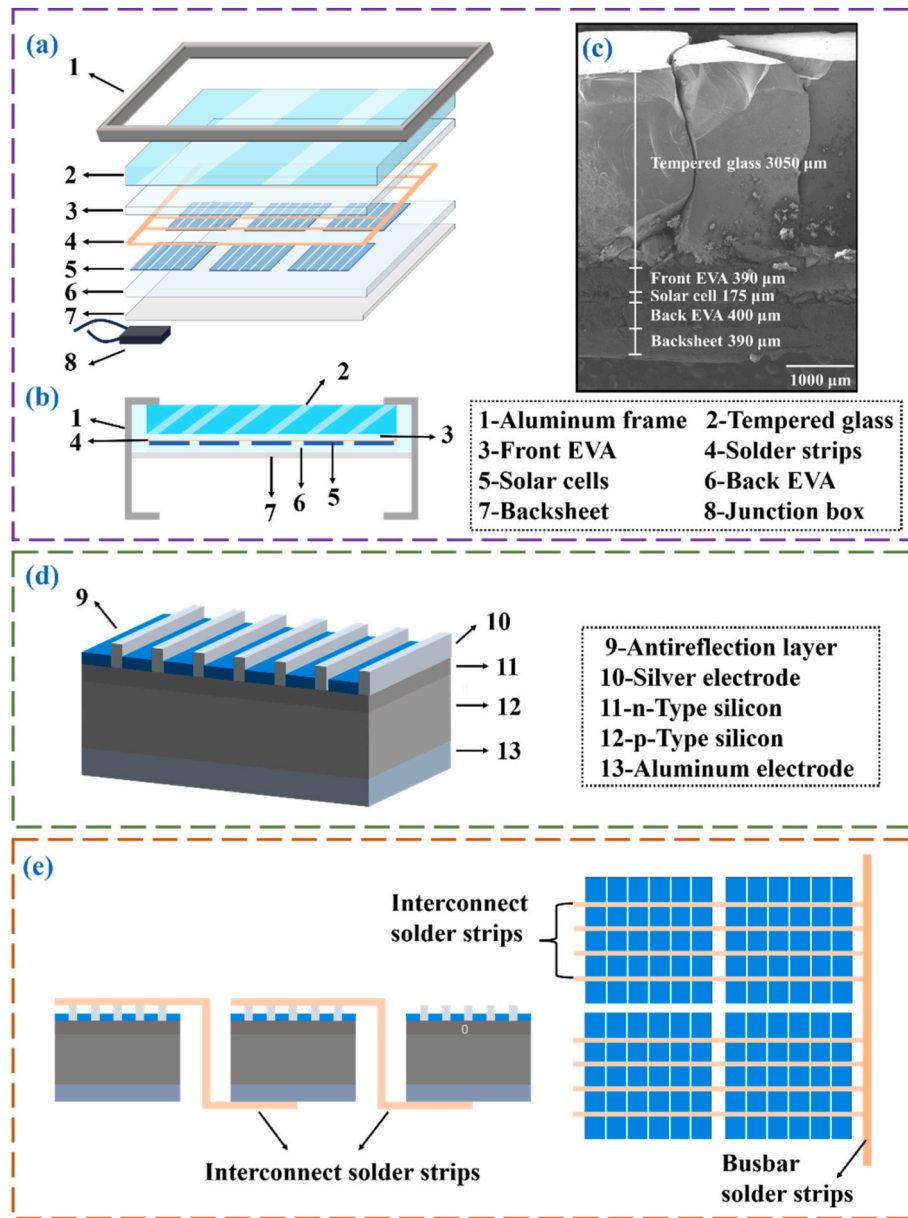
Fig. 1. Composition of c-Si PV module ((a) front view, (b) cross section view, and (c) SEM image), (d) solar cell and (e) schematic diagram of the connection between solder strips and solar cells.

Table 1
The main components and mass proportion of a typical c-Si PV modules [26].

Component	Percentage wt. (%)
Temper glass	76.00
Aluminum frame	8.00
Cable (containing copper and polymers)	1.20
Copper connector	1.00
EVA	5.80
Backsheet	3.60
Silicon wafer	3.60
Aluminum conductor	0.53
Sliver conductor	0.03
Other metals (tin, lead)	0.24

recycled in the recycling of EoL c-Si PV modules.

3. The delamination of c-Si PV modules

In the recycling process of EoL c-Si PV modules, mechanical dismantling can relatively easily disassemble the aluminum frame and junction box from the module. However, further delamination becomes crucial for the remaining tightly adhered components. Ensuring sufficient separation of layers in c-Si PV modules is a prerequisite for recovering valuable components from solar cells. Among them, the efficient removal of encapsulant EVA is a key step in achieving c-Si PV modules delaminating [27]. The delamination methods can be summarized as physical delamination, thermal delamination, and chemical delamination (Table 2).

3.1. Physical delamination

Ordinary mechanical crushing is an early proposed physical

Table 2
The delamination methods and technical parameters for c-Si PV modules.

Methods	Key parameters	Separation effect	Comment	Ref.
Physical delamination	Rotary crusher and hammer crusher	EVA and backsheet: d > 8 mm and 8 > d > 5 mm. Glass: 5 > d > 1 mm and d < 1 mm.	The purity of the product is low and further purification is needed.	[28]
	Multi-stage hammer crushing	Plastic, glass, and metals are enriched in the fractions with d > 1 mm, 1>d > 0.4 mm, and d < 0.4 mm, respectively.	The purity of the product is low and further purification is needed.	[29]
	Hot knife	Glass is completely separated from other components.	Obtain pure and intact glass, but the remaining components need further delamination.	[30]
	Devitrifying machine	Glass is completely separated from other components.	Obtain pure and broken glass, but the remaining components need further delamination.	[31]
	High voltage pulse crushing	High value metals: d < 0.5 mm, glass: 4 > d > 0.5 mm.	The purity of the product is higher than that of ordinary mechanical crushing.	[32]
Thermal delamination	500 °C, 30min, N_2	99 % of polymers are removed.		[33]
	Milling machine cuts off the backsheet before pyrolysis. 500 °C, 1 h, $N_2 + O_2$	EVA is completely removed.	Avoiding the production of fluorinated substances and Reducing the production of VOCs.	[34]
	First: 150 °C, 5 min, N_2 Second: 500 °C, 1 h, N_2	>99 wt% EVA is removed.	Two step pyrolysis method avoids the generation of fluorinated substances.	[35]
	First: 200 °C, 30 min, no airflow. Second: 500 °C, 1 h, $N_2 + O_2$.	EVA is completely removed.	Two step pyrolysis method avoids the generation of fluorinated substances.	[36]
	First: 40 °C, 3 min. Second: 650 °C, 30 min.	EVA is completely removed.	Low temperature treatment combined with thermal treatment avoids the generation of fluorinated substances and has lower energy consumption than two-step pyrolysis method.	[37]
	Ceramic filters, 500 °C.	Ceramic filters decompose EVA and PET to completely remove them.	The pyrolysis products are further decomposed, reducing the toxicity of exhaust gas.	[38]
Chemical delamination	TCE, 70 °C, 8 h.	9.4 wt% of EVA is dissolved.	The dissolved mass of EVA is very low.	[39]
	TCE, 80 °C, 10 d.	Successfully recycled solar cells.	Long time consumption.	[40]
	TCE (4 mol/L), microwave, 70 °C, Solid-liquid ratio is 50 g/L, 2 h.	Complete separation of components.	Reduced reaction time; Increased additional energy consumption.	[41]
	Toluene, ultrasonic (200 W), 60 °C, 60 min.	EVA completely dissolved.	Reduced reaction time; Increased additional energy consumption.	[42]
	Ortho-dichlorobenzene, ultrasonic (900 W), 70 °C, 30 min.	EVA completely dissolved.	Reduced reaction time; Increased additional energy consumption.	[43]
	D-limonene, ultrasonic (450 W), 60 °C, 120 min.	Complete separation of components.	Green agent, low toxicity, short reaction time, high cost of agents.	[44]
	dimethyl propylene urea, ultrasonic (600 W), 160 °C, 30 min.	Complete separation of components.	Green agent, low toxicity, short reaction time, high cost of agents.	[45]
	ethylene glycol diacetate, ultrasonic (900 W), 130 °C, within 30 min.	Complete separation of components.	Green agent, low toxicity, short reaction time, high cost of agents.	[46]
	Ethanol solution of 0.2 mol/L KOH, 200 °C, 3 h.	Complete separation of components.	No use of organic solvents, low cost of agents.	[47]

delamination method. For example, Granata et al. [28] used rotor crusher and hammer crusher to crush c-Si PV modules. Then, products with different particle sizes were obtained through screening, and the fractions with particle sizes d > 8 mm and 8>d > 5 mm enriched most of the EVA and backsheet fragments; And glass particles are mainly enriched in the 5>d > 1 mm fraction. Paganelli et al. [29] used a multi-stage hammer crushing operation to reduce the size of product fragments. After crushing, plastic, glass, and metals were enriched in the fractions with d > 1 mm, 1>d > 0.4 mm, and d < 0.4 mm, respectively. The viscosity and toughness of EVA and backsheet are one of the factors that reduce the crushing efficiency. Freezing c-Si PV modules with liquid nitrogen can effectively improve their brittleness, thereby promoting the crushing effect.

Some more effective physical delamination methods have been reported. Some companies in Japan have adopted a special device called a "hot knife" [30]. When the heated blade or wire passes through the EVA layer between tempered glass and solar cells, the EVA softens and its viscosity decreases, which can separate the tempered glass. However, this method requires extremely high precision control and is therefore not suitable for non-flat c-Si PV modules. Fiandra et al. [31] invented a novel recycling device (Fig. 2). The c-Si PV modules are placed flat with the glass surface facing downwards on a conveyor. An upwardly inclined guide plate is installed at the end of the conveyor. When the c-Si PV modules are transported to the end by the conveyor wheel, it moves upwards through the guide plate. At this time, the roller blade located at the end of the device shaves the glass. This device can almost completely remove the glass on c-Si PV modules, and the flatness requirements for
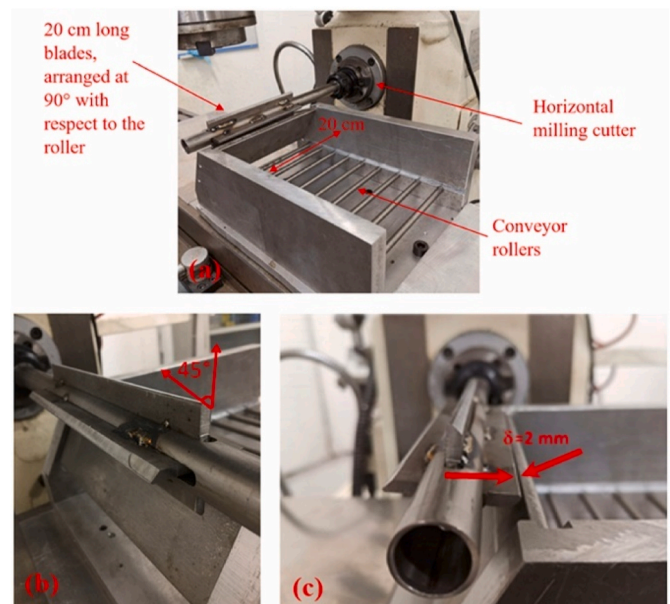

Fig. 2. Details of the devitrifying machine ((a) horizontal cutter and conveyor rollers, (b) blade profile: 45°, and (c) distance between the roller blades and the inclined stator: 2 mm [31].

c-Si PV modules are much lower than those of the hot knife method.

However, the above methods mainly achieve the separation and recovery of glass, while high-value metals are difficult to achieve enrichment and recovery. Compared with traditional crushing, high-voltage pulse crushing can more effectively release of metals in c-Si PV modules [60]. According to the differences in dielectric properties of the materials, the interfaces of materials with different dielectric constants will fracture under high voltage discharge, consequently achieving the liberation between materials. The experimental results of Nevala et al. [61] showed that 99 % of Cu, 60 % of Ag, and 80 % of Pb, Sn, and Al can be enriched in the portion with a particle size d > 4 mm through high-voltage pulse crushing of c-Si PV modules. The remaining Ag, Pb, Sn, and Al mostly exist in the portion with d < 0.5 mm, while high-purity (>99 %) Si is concentrated in the portion with 2>d > 0.5 mm. Zhao et al. [32] mentioned that the crushing selectivity of materials during high-voltage pulse crushing is: Ag > Si > glass, indicating the most significant enrichment effect on Ag. They mainly enriched high-value metals to d < 0.5 mm by adjusting the field strength and pulse number, while glass was enriched to 4>d > 0.5 mm. Song et al. [62] enriched Al and Ag in the particle size range of 2>d > 0.05 mm through high-voltage pulse crushing, while Cu, Pb, and Sn were enriched in the particle size range below 0.25 mm. They also fitted the quantitative relationship between crushing size and energy consumption.

Although the physical delamination method has lower energy consumption and less secondary pollution compared to chemical and thermal methods in the recycling process of c-Si PV modules, it cannot effectively remove EVA and liberate different components. Therefore, the purity of the obtained product is relatively low. Although the physical delamination methods can to some extent concentrate EVA in the coarser fraction, there are still many solar cells and metal particles adhering to EVA, resulting in a decrease in the recovery efficiency of the target product. To improve the recycling efficiency, further measures need to be taken to achieve effective separation of these components. This may require the combination of other technologies, such as chemical or thermal treatment, to more effectively separate EVA and other valuable components.

3.2. Thermal delamination

Thermal delamination often utilizes heating equipment such as tube furnaces, muffle furnaces, and box furnaces to soften and decompose EVA in c-Si PV modules at high temperatures, thereby separating components such as glass, solar cells, and solder strips from each other. The thermal decomposition process of EVA can be divided into two stages. The first stage is the acetylation reaction of ethyl acetate occurring at around 350–410 °C; The second stage occurs in the higher temperature range from 420 to 510 °C, during which the remaining C-C bonds on the main chain are completely broken [63,64]. Therefore, thermal treatment of c-Si PV modules at 480–550 °C is an effective delamination technique. Tempered glass, metals, silicon wafers and other components undergo minimal changes during this process, and the oxidation of metals can be avoided by injecting inert gases such as nitrogen during the thermal treatment process. The thermal treatment conditions given by Riech et al. [33] are to burn at 650 °C in the furnace for 30 min. Dias et al.'s [65] research indicates that pyrolysis at 500 °C for 30 min can remove over 99 % of the polymers in c-Si PV modules. However, due to the presence of fluorinated backsheet, c-Si PV modules will release harmful by-products during the pyrolysis process, such as HF, fluoroalkanes, and aromatic compounds, causing the atmospheric environment pollution [66]. Aryan et al. [67] investigated the environmental impact of fluorinated and non-fluorinated backsheet during pyrolysis and incineration processes through life cycle assessment (LCA). They emphasized that pyrolysis cannot be an effective way to treat fluorinated backsheet from multiple dimensions of economy, environment, and technology. Specifically, the HF gas released during pyrolysis is corrosive and toxic, requiring further alkaline treatment to prevent

environmental pollution. At the same time, the pyrolysis oil and pyrolysis carbon generated during the process may contain fluorine elements, which have adverse effects on the subsequent energy recovery process.

To avoid the negative impact of fluorinated backsheet during the thermal treatment of the module, it is advisable to separate the backsheet before the thermal treatment. Fiandra et al. [34] used a milling machine to scrape off the backsheet before pyrolysis, and it took about 8 min to remove a 100 × 45 cm backsheet. Wang et al. [35] and Fiandra et al. [36] proposed a two-stage pyrolysis method. In the first stage, the c-Si PV module was preheated at low temperature (150–200 °C) for 5–30 min. At this time, EVA became soft and its adhesion decreased. The backsheet was then mechanically or manually peeled off, and finally, the EVA was completely removed by a second stage pyrolysis treatment at high temperature of 500 °C. Wu et al. [37] proposed a novel approach that combines low-temperature treatment and thermal treatment to achieve delamination of the c-Si PV modules. Placing the c-Si PV module in a low temperature environment of −40 °C for 3 min made it easy to separate the rear EVA and the backsheet. The separated rear EVA and backsheet were kept at 100 °C for 30 s to soften the EVA and achieve the separation of the EVA and backsheet. The remaining glass, front EVA, and silicon wafers were maintained at 650 °C for 30 min, and EVA was decomposed to obtain glass and silicon wafers. This method not only avoids the generation of fluorine-containing by-products during pyrolysis, but also allows for the recovery of some EVA and all of the backsheet. Fiandra et al. [36] also changed the pyrolysis atmosphere by heating c-Si PV modules to 500 °C in a mild oxygen atmosphere for 1 h. This not only achieved complete removal of EVA, but also reduced the production of harmful gases. The gases produced under this atmosphere were mainly CO_2, CO, and a small amount of VOCs. In addition, Sasai et al. [38] used ceramic filters loaded with catalysts to catalytically decompose molten PET and EVA in a low-temperature thermal decomposition furnace (aerobic atmosphere), successfully suppressing the generation of black smoke. Huang et al. [68] proposed connecting a washing tower behind the thermal treatment device to recover the fluorinated gas generated by direct combustion of c-Si PV modules in the form of hydrofluoric acid solution.

Thermal delamination technology, with its unique advantages such as short time consumption, high delamination efficiency, and easy operation, has received widespread attention in the field of c-Si PV modules recycling and is regarded by many scholars as a technology with industrial production potential. However, in practical applications, the by-products such as waste gas generated by this technology cannot be ignored. In order to prevent and reduce the production of by-products such as waste gas from the source, a series of measures need to be taken. Firstly, by optimizing the process parameters of thermal delamination, such as temperature, pressure, time, and atmosphere, the generation of by-products such as waste gas and pyrolysis oil can be minimized to the greatest extent. Secondly, advanced waste gas treatment technologies and equipment should be adopted, such as adsorption, catalytic oxidation, etc., to efficiently treat the generated waste gas and ensure that its emissions comply with environmental standards. In addition, the possibility of reusing by-products such as pyrolysis oil generated during the thermal delamination process can be explored. Through refining and purification methods, some pyrolysis oil can be converted into valuable chemicals or fuels, thus achieving resource recycling. However, caution should be exercised as high-temperature processes may be limited in the future carbon neutral society, because they are not conducive to reducing carbon emissions. Therefore, it is necessary to develop a delamination process that does not require high temperatures and has minimal impact on environmental quality.

3.3. Chemical delamination

Chemical delamination technology is a method that uses chemical solvents to dissolve or swell EVA, causing EVA to lose its adhesion while having a relatively small impact on the properties of other components.

EVA is composed of non-polar ethylene segments and polar vinyl acetate segments. This special structure allows some non-polar solvents (such as benzene, toluene, trichloroethylene) and polar solvents (such as acetone) to dissolve EVA. The dissolution process of EVA conforms to the normal dissolution model, where the solvent plasticizes the polymer, forming a swollen gel-like polymer layer [69]. Prasad et al. [39] investigated the dissolution of EVA using solvents such as toluene, tetrahydrofuran, trichloroethylene, dichloromethane, and methanol. They pointed out that using trichloroethylene (TCE) as a solvent, the ratio of c-Si PV modules to TCE was 1:7.44, and the reaction temperature was 70 °C for 8 h. The best dissolution effect of EVA was achieved, but only 9.4 wt% of EVA was dissolved under this condition. The experimental results of Doi et al. [40] also demonstrated that cross-linked EVA can be dissolved in a TCE solution at 80 °C, but it requires up to 10 d.

In order to improve the dissolution efficiency of EVA and shorten reaction time, the techniques including microwave and ultrasound assisted dissolution methods, have been proposed. Pang et al. [41] introduced microwave enhanced swelling of EVA in TCE. Under the conditions of 4 mol/L TCE, a solid-liquid ratio of 50 g/L, and a temperature of 70 °C, microwave reaction was carried out for 2 h resulted in the layers of the c-Si PV module were completely separated. This is because the excellent microwave absorption performance of the anti-reflection layer and the difference in thermal expansion coefficients of each layer in the c-Si PV module accelerate the swelling separation. Azeumo et al. [42] and Kim et al. [43] investigated the dissolution effect of ultrasonic enhanced EVA in toluene and o-dichlorobenzene, respectively. The results showed that when toluene was used as a solvent, EVA could be completely dissolved under the conditions of ultrasonic power of 200 W and a temperature of 60 °C for 60 min. When using o-dichlorobenzene as a solvent, higher ultrasonic power (900 W) and slightly higher temperature (70 °C) were required, and the reaction time could be shortened to 30 min to achieve complete dissolution of EVA. The enhanced effect of ultrasonic can be understood as the formation, rapid growth, and implosion collapse of cavitation bubbles, resulting in a high-pressure and high-temperature environment around the bubbles, thereby accelerating the reaction process [70]. However, Kim et al. [43] pointed out that hydroxyl radicals generated by ultrasonic cavitation have a certain degree of degradation on organic solvents.

However, it is important to maintain caution as the organic solvents mentioned above, used for EVA dissolution, are highly toxic and pose significant risks to environmental quality and operators. To address this issue, some low toxicity solvents or novel technologies have been introduced. D-Limonene, a chemical substance extracted from natural fruits that can effectively remove adhesives, was used by Abdo et al. [44] to dissolve EVA. Their research shows that using D-limonene as a solvent, combined with ultrasonic with a power of 450 W, can effectively delaminate the layers of c-Si PV module by reacting at 60 °C for 120 min. Li et al. used green reagents dimethyl propylene urea (DMPU) [45] and ethylene glycol diacetate (EGDA) [46] as solvents for the delamination of c-Si PV modules and analyzed the delamination mechanism. However, it should be noted that the expensive cost of these chemical agents cannot be ignored.

An international invention patent (WO2017009062A1) [71] proposes an innovative method aimed at achieving the separation of different layers in c-Si PV modules and the depolymerization of backsheet and encapsulant by adding oxidants in hydrothermal subcritical systems. This method utilizes the special environment under hydrothermal subcritical conditions to enhance the decomposition of polymers, providing a new technological approach to delaminate c-Si PV modules. Lovato et al. [72] used supercritical carbon dioxide and toluene technology combined with planetary grinding to achieve c-Si PV modules delaminating, achieving 100 % recovery of glass, solder strips, and backsheet, as well as 85 % recovery of solar cells and EVA. Yan et al. [47] placed the c-Si PV modules in a high-pressure reactor and then added an ethanol solution containing a certain amount of KOH. The separation of different layers was achieved by reacting at 200 °C for 3 h.

The high toxicity, high consumption, and high economic cost of solvents and long reaction time are key factors limiting the industrial application of chemical delamination technology for c-Si PV modules. Researchers need to continue to make efforts to explore low toxicity, low cost, and efficient chemical solvents. Moreover, there may still be some incomplete separation between EVA and solar cells after chemical treatment, and further thermal treatment is needed, which undoubtedly increases economic costs.

During the thermal delamination process, internal stress is generated due to the different thermal expansion coefficients between the layers of the c-Si PV module, as well as the inability to release the pyrolysis gas smoothly. When these stresses exceed the limit that solar cells can withstand, the cells may break. In the process of chemical delamination, the swelling rate of EVA in organic solvents is greater than its dissolution rate, resulting in volume expansion of the EVA layer during the dissolution process, thus exerting compressive pressure on the solar cells. If such compression force exceeds the capacity of the solar cells, it will also lead to breakage. Xu et al. [73] and Doi et al. [40] effectively reduced the breakage of solar cells during thermal or chemical delamination processes through strategy of s solvothermal swelling with thermal decomposition (SSTD) or applying external mechanical pressure, respectively. In the whole process of recycling EoL c-Si PV modules, the phenomenon of module breakage is an inevitable fact. From modules disassembly, loading, road transportation, to unloading at the recycling companies and entering the recycling production line, modules are inevitably subjected to external physical impacts and collisions, resulting in structural damage. In addition, the decreasing thickness of solar cells in newly produced c-Si PV modules increases the risk of their breakage [74]. Furthermore, in large-scale industrial recycling, production efficiency is an important indicator of enterprise benefits, and the production efficiency of recycling intact silicon wafers is lower than that of recycling broken silicon wafers. Therefore, the author proposes that the focus of recycling c-Si PV modules should not be on preventing the breakage of solar cells and recycling intact silicon wafers, but on solving the challenges of recycling broken silicon wafers. This contradicts the views of Tao et al. [75], who argue that complete c-Si PV modules have higher economic value (complete modules cost $22/module vs broken modules cost $10.61/module) and advocate for special measures to be taken during packaging and transportation to prevent damage to c-Si PV modules.

3.4. Sorting

After removing EVA and backsheet of the broken c-Si PV modules through thermal or chemical delamination, the remaining low-value glass particles, high-value solar cells, and solder strips will mix together. Necessary measures need to be taken to separate these three types of components for subsequent step-by-step recovery. Or the mixed materials obtained through physical delamination still need further sorting and enrichment. Utilizing the differences in physical properties such as density, shape, conductivity, and magnetism of different components, using physical sorting techniques is a good choice. Dias et al. [76] used a roller electrostatic separator to sort the ground c-Si PV modules based on differences in conductivity. The results indicate that organic polymers fell into non-conductive grooves, silicon particles fell into intermediate grooves, and glass particles fell into conductive grooves together with metals due to their heavy weight. Bogust et al. [77] developed a special slit sieve that utilizes the differences in size and shape between glass particles and solar cells to allow the solar cells to pass through the slit sieve while the glass particles remain upon the sieve. Optical sorting machines can distinguish components of different colors, and some European photovoltaic recycling companies use optical analysis equipment to separate glass fragments from mixed materials [78]. Gas/solid fluidized beds are commonly used in the separation and processing of coal and other minerals, achieving the separation of different components based on differences in density, particle size and

shape [79,80]. The author's team achieved efficient separation of glass particles and solar cells in the mixed material after pyrolysis using a gas/solid fluidized bed [81]. The results showed that the recovery and purity of the solar cells in the mixed materials with a particle size of d > 4 mm were 91.09 % and 84.4 %, respectively; For the mixed materials with 2<d < 4 mm, the recovery and purity of solar cells were 82.29 % and 80.52 %, respectively. The author's team also sorted the solder strips from the mixed materials of solar cells and solder strips using pulsing air flow device, which could enrich 96.45 wt% of the solder strips into heavy fraction.

Physical mechanical sorting is an indispensable part of the c-Si PV modules recycling process, and its low operating cost and simple operation make it highly suitable for commercial promotion. But there are still some shortcomings, such as: the product purity after sorting is not high enough and the sorting of small-sized materials is difficult to achieve. Therefore, it is recommended that future research focus on further improving product recovery efficiency, purity, as well as the sorting of small-sized mixed materials.
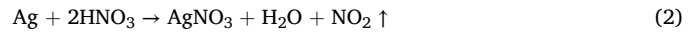
4. High-value material recycling

4.1. Purification and metal recovery of solar cells

Pure components such as glass, solar cells, and solder strips are obtained through a series of physical, thermal, chemical, and sorting treatments. Glass, as a low-value material, can be directly recycled. However, further purification is required for solar cells and solder strips to recover valuable metals and Si Table 3 lists some processes and related parameters reported in recent years for purifying solar cells. The purification of solar cells normally uses acidic or alkaline solutions to remove metals (Ag, Al), antireflection layer, and n-p junction from their surfaces. Consequentially, the resulting metal leaching solution is processed through precipitation, electrodeposition, extraction, and other

methods to obtain metal salts, metal oxides, and elemental metals.

Kang et al. [48] used a mixed acid solution of $HF + HNO_3 + H_2SO_4 + CH_3COOH$ to simultaneously remove Al, Ag, antireflection layer, and n-p junction, obtaining Si with a purity of 99.999 %. This one-step etching method is not convenient for the separation and recovery of metals, so the step-by-step etching method is more favored by researchers. A study [49] demonstrated a typical step-by-step etching method, including: First, dissolving the Al electrode in 18 wt% - 24 wt% HCl solution at room temperature (10–15 min); Second, placing the solar cells obtained in the first step into a 30 wt% HNO_3 solution and leaving them at 50 °C for 10 min to completely remove Ag; Finally, the solar cells obtained from the second step were immersed in a 20–30 wt% HF solution at room temperature for 5 min, successfully removing the antireflection layer. The relevant reaction equations are as follows (Eqs. (1)–(3)). The flowchart of the sequential etching process for solar cells is illustrated in Fig. 3.

$$2Al + 6HCl \rightarrow 2AlCl_3 + 3H_2 \uparrow \tag{1}$$

$$Ag + 2HNO_3 \rightarrow AgNO_3 + H_2O + NO_2 \uparrow \tag{2}$$

$$Si_3N_4 + 12HF \rightarrow 3SiF_4 \uparrow + 4NH_3 \uparrow \tag{3}$$

Similar studies have been conducted, for example, Wang et al. [50] using a mixed solution of $HCl + H_2O_2 + H_2O = 1:1:5$, 5 % HF, and 25 % NaOH to sequentially remove Al, the antireflection layer, and Ag with Si-Al alloys. Ultimately, 62 % Si (purity ~8N) was recovered. Klugmann Radziemska et al. [51] immersed the solar cells in a 30 % KOH solution at 60–80 °C, and the removal of Al could be achieved within 2–3 min; Then place the solar cells in a mixed solution of HNO_3 (65 %) + HF (40 %) + CH_3COOH (99.5 %) + Br_2 and leave them at 40 °C for 9 s to remove Ag, antireflection layer, and n-p junction. In order to avoid the use of highly toxic and corrosive HF, some scholars have suggested using H_3PO_4 instead of HF to remove the antireflection layer or n-p junction

Table 3
The purification methods and technical parameters for solar cells.

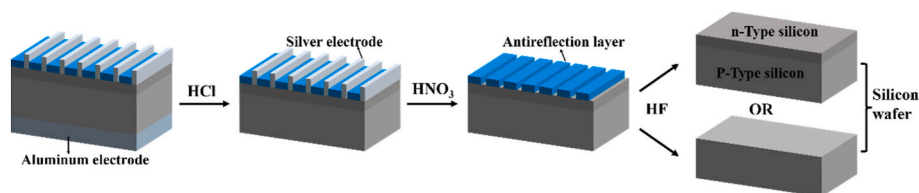| Etching methods	Recovery effect			Comment	Ref.
	Recovery efficiency	Purity efficiency	Etching efficiency		
HF (48 %), HNO$_3$ (70 %) H$_2$SO$_4$ (97 %), CH$_3$COOH (99 %)	Si (86 %)	Si (99.999 %)		One step etching method, metal is difficult to classify and collect.	[48]
HCl (18 wt% - 24 wt%), 10–15 min, etching Al HNO$_3$ (30 wt%), 10 min, etching Ag HF (20–30 wt%), 5 min, etching antireflection layer			100 %	High etching efficiency, different metals are separated during the etching process.	[49]
HCl + H$_2$O$_2$ + H$_2$O = 1:1:5, etching Al HF (5 %), etching antireflection layer and Ag NaOH (25 %), etching Si-Al alloy	Si (62 %)	Si (99.999999 %)		Low recovery efficiency and high Si loss.	[50]
KOH (30 %), 2–3 min, etching Al HNO$_3$ (65 %) + HF (40 %) + CH$_3$COOH (99.5 %) + Br$_2$ mixed solution, 9 s, etching Ag, antireflection layer and n-p junction.			100 %		[51]
HNO$_3$ (5 mol/L), 1 h, etching Ag, Cu, Sn, and Pb H$_3$PO$_4$ (90 %), 160 °C, 60 min, etching antireflection layer KOH (40 %), 10 min, etching Ag	Si (80 %) Cu (79 %) Ag (90 %)			Not using HF , more environmentally friendly, but the recovery efficiency is not high.	[52]
NaOH (10 mol/L), 5 min, etching Al HNO$_3$ (6 mol/L), 5 min, etching Ag H$_3$PO$_4$ (90 %), 45 min, etching antireflection layer		Si (99.9984 %)		Not using HF, more environmentally friendly.	[53]
HNO$_3$ (69 %) + H$_2$SO$_4$ (50 %) + H$_2$O$_2$ (30 %) + H$_2$O mixed solution, etching Ag, Al, and Pb	Ag (82 %) Pb (91 %) Al (86 %)			Not using HF, more environmentally friendly.	[54]
H$_2$SO$_4$ (1 mol/L) + 1 % H$_2$O$_2$, 24 h, etching Ag	Ag (96.75 %)			Not using HF and HNO$_3$, more environmentally friendly.	[55]
MSA: H$_2$O$_2$ = 9:1, within 4 h, etching Ag		Ag (99.995 %)		Not using HF and strong acids, more environmentally friendly and safe.	[56]
CuCl$_2$ (0.75 mol/L), 80 °C, 60 min, Solid-liquid ratio is 50:1, etching Ag			Ag (100 %)	Not using strong acids or alkalis, but with high reagents cost and consumption.	[57]
FeCl$_3$ (0.5 mol/L) + Choline chloride + H$_2$O, 65 °C, 10 min, etching Ag	Ag (95 %)	Ag (98 %)		Not using strong acids or alkalis, but with high reagents cost and consumption.	[58]
FeCl$_3$ (0.5 mol/L), 30 °C, 1 min, etching Al. HNO$_3$ (64 %), room temperature, 2 h, etching Ag.	Ag (94 %)			Replacing other Ag precipitants with NaCl, green and safe.	[59]

Fig. 3. Flowchart of the sequential etching process for solar cells.

[82]. This is because hot H_3PO_4 can react with Si_3N_4 (Eq. (4)). For example, Jung et al. [52] placed the solar cells that have removed Ag and Al in a 90 % H_3PO_4 solution at 160 °C for 60 min to remove the antireflection layer. The mechanical polishing method can also avoid the use of HF. As shown in Park et al.'s [83] study, polishing the solar cells at a speed of 20 rpm for 20 min on a grinding disc can effectively remove the antireflection layer and n-p junction.

$$Si_3N_4 + 4H_3PO_4 + 6H_2O \rightarrow 3SiO_2 + 4NH_4H_2PO_4 \tag{4}$$
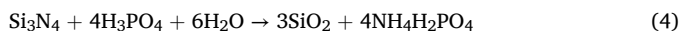
Solar cells can be cleaned with different etchants to obtain high-purity silicon wafers, but further recovery measures require to be taken for metals that exist in the etching solution as ions. Punathil et al. [53] proposed an effective recovery strategy, which involves holding the solar cells in a 10 mol/L NaOH solution (63 °C) for 5 min to remove Al and form $Al(OH)_3$ precipitate; Subsequently, maintaining the solar cells in a 6 mol/L HNO_3 solution (70 °C) for 5 min to remove Ag and generate $Ag(NO_3)$ solution; Finally, keeping the solar cells in a 90 % H_3PO_4 solution (70 °C) for 45 min to remove the antireflection layer. During the treatment process, the $Al(OH)_3$ precipitate in NaOH solution can be recovered through filtration. For the $Ag(NO_3)$ solution, AgCl precipitate can be generated by adding HCl to recover Ag in the form of AgCl. Jung et al. [52] first leached Al and Ag using KOH and HNO_3 respectively, and then recovered Al and Ag through 2 and 4 steps, respectively. Recovery of Al: (1) $Al(OH)_3$ was Filtered out from KOH solution; (2) It was then heated at 1200 °C for 3 h, 94 % of Al was ultimately recovered in the form of Al_2O_3. The recovery route they proposed for Ag is more elaborate and includes: (1) Adding 5 M HCl solution to HNO_3 leachate to obtain AgCl; (2) Transferring AgCl to a 5 M NaOH solution to obtain Ag_2O; (3) Using hydrazine hydrate ($N_2H_4 \cdot H_2O$) to reduce Ag_2O in a solution of distilled water and ethanol (volume ratio 2:1) and obtain elemental Ag; (4) Ag metal with a purity of 99.99 % (recovery of 90 %) was obtained through melting, solidification, and electrolysis steps. In Lim et al.'s [54] study, a mixed solution of $HNO_3 + H_2SO_4 + H_2O_2 + H_2O$ was used as an etching solution to leach Ag, Al, and Pb in one step. Then, NaOH was added to adjust the pH to 6 to precipitate Al, which was filtered and recovered. Carbon fiber was used as the working electrode and counter electrode, and AgCl was used as the reference electrode to electrodeposit Ag and Pb from the filtrate at +0.41 V and −0.13 V battery potentials, respectively. The recovery efficiency of Ag, Pb, and Al were 86 %, 95 %, and 97 %, respectively. Huang et al. [68] conducted in-depth research on the application of electrodeposition technology in Ag recovery. They first used HNO_3 to leach Ag, and then recovered Ag from the leaching solution through electrodeposition. During the experiment, they used a ternary electrode system, with Pt foil as the counter electrode, Ti foil as the working electrode, and Ag/AgCl foil as the reference electrode. Through this electrodeposition method, they successfully recovered 74 % of Ag with a purity of up to 99 %. Although electrodeposition technology has shown good recovery results on a laboratory scale, it still faces challenges in large-scale applications. Especially, the construction and operation of large-scale electrodeposition reactors will be crucial to industrializing this technology. Future research should focus on how to design and optimize these reactors to improve electrodeposition efficiency, reduce costs, and ensure high recovery efficiency and product quality can still be maintained in large-scale operations.

Some scholars have used the combination of other acids + H_2O_2

instead of HNO_3 to etch Ag on solar cells, in order to avoid the generation of NO_x. Yashas et al. [55] pointed out that H_2O_2 can be used as an oxidant to etch Ag, and its concentration does not need to be too high. A combination of H_2SO_4 and H_2O_2 was used by them to etch Ag from solar cells, then a catalyst column coated with platinum activated carbon (Pt/AC) was used to selectively concentrate Ag, and finally recovered Ag through electrodeposition. The experimental results from Yang et al. [56] demonstrated that methanesulfonic acid (MSA) + H_2O_2 can effectively etch Ag from solar cells, and then added HCl to the leaching solution to promote the precipitation of AgCl. Subsequently, AgCl reacted with NaOH and H_2O_2 successively to recover Ag metal powder, and finally the metal powder was refined to increase the purity of Ag metal to 99.995 %.

The strong corrosiveness of strong acids and alkalis imposes extremely high requirements for the corrosion resistance of reaction vessels, and poses a threat to the health of operators and environmental quality. Therefore, the demand for green and environmentally friendly etchants has attracted the attention of researchers. Deep eutectic solvents (DESs) are a novel solvent system that is considered a green solvent due to their low toxicity and high biocompatibility. DESs have applications in organic compounds separation/catalysis, element extraction from minerals, and recycling of waste lithium-ion battery cathode materials [84–86]. Zhang et al. [57] and Zante et al. [58] demonstrated through experiments that DESs have high applicability and strong recyclability in etching metals from solar cells. For example, Zante et al.'s experiment first used aqueous solutions of $FeCl_3$ or $AlCl_3$ to remove Al from solar cells, then etched in a DES composed of $FeCl_3$, choline chloride, and water for 10 min to completely remove Ag, and finally, a large amount of water was added to the DES to precipitate AgCl for recovery. The relevant reaction equations are as follows (Eqs. (5) and (6)). However, using DESs to purify solar cells is a high-cost method. From an economic perspective, conventional acids and alkalis systems are more attractive than DESs.

$$Al + 3Fe^{3+} \rightarrow Al^{3+} + 3Fe^{2+} \tag{5}$$

$$Ag + Fe^{3+} \rightarrow Ag^+ + Fe^{2+} \tag{6}$$

The author believes that chemical etching is a crucial step in the purification process of solar cells to remove impurities. Although many etchants are volatile, corrosive, and toxic, this method is very fast, efficient, and thorough. In particular, when it is necessary to obtain solar grade silicon (Si with a purity of over 99.9999 % [87]), chemical etching is essential. But in addition to the issues discussed above, one point that needs to be particularly emphasized is that precise control of the etching process is crucial. This is because during the etching process, especially when using reagents that can react with Si (such as using HF to etch the antireflection layer), if not precisely controlled, it may lead to unnecessary sacrifice of Si.

4.2. Reuse of silicon materials

Although significant progress has been made in material recycling research, exploring the secondary market and ultimate use of recycled materials is equally crucial. In the process of recycling solar cells, preliminary purification can obtain silicon material with a certain purity, which provides a foundation for subsequent reuse. However, further

upgrading these preliminarily purified silicon materials to solar grade purity not only requires more economic costs, but also raises the question of whether photovoltaic recyclers or their downstream enterprises are willing to engage in this purification production. A study on the synthesis of spherical nano SiO_2 particles from bulk silicon materials through hydrothermal reaction has been reported [88]. SiO_2 particles with controllable particle size in the range of 8–50 nm were synthesized by reacting at a temperature of 300–453 K for 2–96 h. Nano SiO_2 particles have wide applications in the production of electronic devices, nanocomposites, building materials, adsorbents, and catalysts [89,90]. Eshraghi et al. [91] and Zhang et al. [92] prepared preliminarily purified silicon materials into nano silicon and porous silicon for using as anode materials in lithium-ion batteries through ball milling treatment and alloying/dealloying treatment in molten salt, respectively. They obtained high electrical capacities of 1400 mAh/g and 2427 mAh/g, respectively. These interesting and meaningful attempts have broadened the perspectives for the recovery and reuse of silicon in c-Si PV modules.

4.3. Purification and recovery of solder strips

The metal copper (Cu) in the solder strips is the most abundant among all metals in c-Si PV modules, with a value second only to Ag. However, there is limited research on the recycling of solder strips, which to some extent limits the integrity and efficiency of c-Si PV modules recycling. Huang et al. [68] dissolved the metals by placing the solder strips and solar cells together in HNO_3, and then used sequential electrodeposition to recover the metals, achieving a recovery of 83 % for Cu (with a purity of 99.99 %). Chen et al. [93] proposed three methods for recovering solder strips, including physical polishing, chemical etching, and leaching - extraction, obtaining copper wires with a purity of 99.63 %, copper wires with a purity of 99.57 %, and CuO with a purity of 99.70 %, respectively. Gao et al. [94] adopted a strategy of air roasting followed by alkaline leaching to remove the lead and tin coating on the solder strips, and then used electro-deposition technology to recover lead and tin from the leachate, effectively recycling the full composition of metals from the solder strips. Recycling solder strips can yield metals such as Cu, Pb, Sn, etc., which can increase economic value and be very beneficial for recyclers. Therefore, more attention should be paid to the recycling of solder strips. Although these existing studies have made good performance in the purification of the solder strips, there is a lack of work on how to separate and concentrate the solder strips from the mixed materials of c-Si PV modules, which should receive more attention, as discussed by the author in section 3.4.

5. Low-value material recycling

Materials such as glass, EVA, and backsheet have not attracted much attention from researchers due to their low recycling value. However, exploring the potential recycling pathways and ultimate applications of these low-value materials is also meaningful for development. Due to the presence of fluoropolymers, the recovery of backsheet is often overlooked. However, the majority of material that make up backsheet is PET, which is widely used in the synthesis of plastics and resins [95,96]. Morita et al. [97] treated the backsheet with a hot alkaline solution, degraded PET into soluble disodium terephthalate, and then filtered and recovered the separated PVDF, achieving complete recovery of the backsheet. Pham et al. [98] used PET from the backsheet and Al on the solar cells to synthesize MOFs materials, which was used for the adsorption of methylene blue and methyl orange, with adsorption capacities of 190.1–262.5 mg/g and 244.5–296.7 mg/g, respectively. This indicates a new approach for the high-value recycling and utilization of c-Si PV modules. Chitra et al. [99] placed c-Si PV modules on a heating plate, first heating them to 130 °C to separate the backsheet, and then heating them to 170 °C to mechanically remove the EVA, achieving the recovery of EVA. The recycled EVA can be used as a potential fuel because it has a high calorific value of 39.51 MJ/kg, which is equivalent to the calorific value of biodiesel and natural gas [100]. It is worth noting that due to the variability in backsheet composition, structure, and EVA content of c-Si PV modules produced by different manufacturers or at different times, as well as the varying degrees of deterioration of these polymers in waste c-Si PV modules, the complexity of polymer recycling processes increases [24,25]. This requires a universally applicable recycling scheme.

6. Life cycle assessment

Life cycle assessment (LCA) is a method used to evaluate the potential environmental impacts of a product or service throughout its entire lifecycle [101]. LCA is one of the key factors in the management of EoL c-Si PV modules and has attracted the attention of researchers [102]. Disa et al. [4] demonstrated through LCA that high-throughput, large-scale processing of EoL c-Si PV modules have a positive environmental impact compared to direct landfilling, but it is not economically profitable. The LCA analysis by Lunardi et al. [103] and Mao et al. [104] showed that the upcycling of EoL c-Si PV modules, which involves dismantling, delamination, pyrolysis, and hydrometallurgy to recover valuable materials from PV modules, is the most environmentally beneficial. However, its economic feasibility is not as good as the downcycling (only recycling aluminum frames, junction boxes, glass, and copper cables). And they also pointed out thermal treatment and transportation have a significant impact on the recycling process. The sales revenue of silver, aluminum, and silicon is the most sensitive parameter affecting the economic viability of the recycling process [105]. Therefore, from the perspective of environmental impact and economic benefits, more detailed upcycling of valuable materials from EoL c-Si PV modules is more favored. However, the environmental benefits and economic feasibility of different recycling methods vary.

Li et al. [106] compared the costs and benefits of mechanical recycling, chemical recycling, and thermal recycling technologies based on LCA. The results showed that mechanical recycling has the smallest lifecycle energy use (−28,949 MJ/ton), chemical recycling has the smallest industrial water use (−65,069 kg/ton), and thermal recycling has the smallest global warming potential (−5525 kg CO_2 eq/ton). Del Pero et al. [107] conducted LCA on mobile mechanical processing plants for handing EoL c-Si PV modules, and indicated that mobile recycling systems have greater environmental benefits because they reduce the impact of transportation of EoL PV modules. Pagnanelli et al. [108] pointed out that although chemical solvent treatment of polymers have negative impacts on human health and the environment, it has advantages over thermal treatment methods because it can obtain high-value glass. The thermal delamination – acid etching – sequential electrodeposition process was used by Wei Lim et al. [54] to recycle EoL c-Si PV modules. LCA analysis showed that using this recycling method to process EoL c-Si PV modules significantly reduced the global warming potential by about 393 %. Furthermore, by implementing more effective processing techniques to reduce and prevent the release of toxic substances into the environment, its impact can be mitigated.

In summary, the comprehensive recycling of valuable materials from EoL c-Si PV modules is crucial for reducing ecological and environmental impacts and achieving sustainable development in the photovoltaic industry. However, it may not be economically profitable. Additionally, existing studies with LCA have not taken into account the contribution of the different component sorting process, and the recycling of low-value materials has also been omitted, which is a direction that subsequent research needs to strive towards.

7. Policies on the management of EoL photovoltaic modules

In 2012, the European Union classified EoL photovoltaic modules as WEEE (Directive 2012/19/EU), which is based on the extended producer responsibility (EPR) principle [109]. This directive requires photovoltaic suppliers to take responsibility for collecting and recycling

waste photovoltaic modules and stipulates a recycling efficiency of 85 % for secondary raw materials. The German government implements business to consumer (B2C) and business to business (B2B) regulations to manage electronic waste from photovoltaic modules [110]. In the United States, EoL photovoltaic modules are recognized as solid waste, and if they meet the toxicity characteristics, they will be treated as hazardous waste [111]. Some states have enacted regulations and guidelines for the recycling of waste photovoltaic modules. For example, New Jersey has established a solar panel recycling committee and recommended the construction of new solar panel recycling centers or the management of EoL photovoltaic modules as general waste in existing facilities, including charging an advanced recycling fee for each solar panel when consumers purchase or expanding producer responsibility [112]. In California, the Department of Toxic Substances Control has established regulations to designate EoL photovoltaic modules identified as hazardous waste as general waste and implement general waste management for them [113]. There are differences in guidelines and specific management requirements among states in the United States, and greater coordination is needed between federal and local governments, as well as among the states [114].

In September 2022, the National Energy Administration of China proposed the development of standards for the retirement, recycling, and reuse of photovoltaic modules [115]. In July 2023, the Chinese government enacted the "Guiding Opinions on Promoting the Circular utilization of Retired Wind Power and Photovoltaic Equipment", which supports photovoltaic equipment manufacturing enterprises to establish photovoltaic recycling systems through independent recycling, joint recycling, or commissioned recycling [116]. In January 2024, the Ministry of Ecology and Environment of China assigned the waste code 900-015-S17 to waste photovoltaic modules [117]. In May of the same year, the China Resources Recycling Association released a draft of the "Technical Specification for Environmental Pollution Prevention and Control for Recycling of Waste Photovoltaic Modules" for comments [118]. This standard specifies the technical requirements for the entry, storage, disposal and utilization process of waste photovoltaic modules, as well as the process of product pollution control. It can be seen that China's development in the field of EoL photovoltaic modules recycling is gradually shifting from a broad horizontal layout to a deep vertical targeting. However, more detailed regulations, requirements and region-specific strategies require continuous efforts.

8. Discussion and outlook

Green and harmonious sustainable development strategies are essential for many enterprises and industries, and the recycling of EoL c-Si PV modules should also be implemented in this way. However, the current recycling technology of EoL c-Si PV modules is still in its early stages and requires in-depth and continuous research to optimize production techniques [119]. Although many methods have been reported, most of them only remain at the laboratory scale, and their effectiveness in semi-industrial or industrial scale applications require systematic verification. And all recycling methods have some problems that need to be solved urgently, such as the low efficiency of physical delamination; The management of by-products such as waste gas and pyrolysis oil under thermal delamination; The extensive use of toxic reagents in chemical delamination; The optimization of reagent types and combinations; As well as physical sorting of the mixed materials with fine particle size. In addition, due to the extremely low proportion of valuable materials in c-Si PV modules, a low-cost recycling system is necessary, otherwise the economic income obtained from recycling c-Si PV modules may difficult to balance the production process expenses.

It should be emphasized that this study only discusses a series of recycling technologies for EoL c-Si PV modules (Al-BSF type solar cells). However, due to the updating and development of manufacturing technologies, the types of solar cells and the structures of photovoltaic modules have become diversified. For example, PERC solar cells are

gradually replacing Al-BSF solar cells and becoming the mainstream technology [120]. The types of solar cells include: full area aluminum back surface field (Al-BSF), passivated emitter and rear cell (PERC), tunnel oxide passivated contact (TOPCon), and silicon heterojunctions (SHJ), among others. In order to reduce the consumption of scarce materials and maintain sustainable TW level manufacturing capabilities in the future, the manufacturing technology of solar cells needs to be adjusted and innovated [121], which will undoubtedly increase the diversity and complexity of solar cells in the future market. The types and consumption of materials used in the production process of different types of solar cells have changed, which poses strict requirements for the universality of recycling technology. As reported by Gao et al. [94], a method of etching solar cells using molten hydroxide was applied to four different types of solar cells: Al-BSF, PERC-I, PERC-II, and TOPCon, all of which achieved excellent etching effects. The etching process only took 180 s, and more than 99 % and 98 % of silver and silicon could be recovered, respectively. In addition to c-Si PV modules, different types of photovoltaic modules such as amorphous silicon (a-Si) photovoltaic modules, cadmium telluride (CdTe) photovoltaic modules, and copper indium gallium selenide (CIGS) photovoltaic modules are also applied to a certain extent in reality. Although their market share is far lower than that of c-Si PV modules, the recycling of valuable materials from them is a good measure to address the sustainability crisis of rare materials [122]. Therefore, research on the recycling technology of photovoltaic modules must continue, and recycling strategies need to be constantly enriched to cope with the recycling challenges caused by the diversity and variability of photovoltaic module structures and materials.

Developing solar cells and photovoltaic modules that are easier to recycle is a good way to promote photovoltaic recycling, which helps simplify the recycling process. Meanwhile, using recycled materials to manufacture new photovoltaic modules can effectively promote material recycling, which not only solves the problem of selling secondary materials produced by recyclers, but also reduces the carbon footprint of photovoltaic module products [123,124]. This requires emphasis on producer responsibility, and manufacturers of photovoltaic modules should take on more responsibilities related to module recycling, just as some scholars have suggested that manufacturers of power batteries and new energy vehicles need to take on recycling responsibilities [125, 126].

In summary , To the best of our knowledge , We propose the following suggestions and prospects.

(1) Optimizing recycling strategies: This is includes focusing on effective recycling of broken c-Si PV modules. The contribution of the sorting process for different materials and components to the photovoltaic recycling process cannot be ignored. It is essential to explore green, low toxicity, and efficient chemicals, and combining multiple techniques to improve the purity and recovery efficiency of recycled products. To address the diversity of composition and structure of future photovoltaic modules, recycling solutions with universal applicability are more worthy of support. At the same time, it is necessary to strictly control secondary pollutants during the production process and implement measures for pollutant reduction. In addition, appropriately recycling low-value materials to increase economic income and achieve the economic sustainability of recycling technology. And in the recycling process, high-value utilization of photovoltaic materials can be considered, such as preparing electrode materials. Finally, in the design of recycling schemes, carbon footprint needs to be taken into account to meet the requirements of society and government for carbon emissions during the recycling process.

(2) Multidisciplinary integration: The recycling of EoL c-Si PV modules is an interdisciplinary task that involves multiple fields such as pyrometallurgy, hydrometallurgy, mineral processing, chemistry, computer science, automation systems, and environmental

science. Among them, the delamination of c-Si PV modules and the etching/purification of solar cells require professional knowledge in pyrometallurgy and hydrometallurgy. The processes of crushing, sorting, and enrichment are closely related to mineral processing technology. The study of the decomposition characteristics of polymers relies on the support of the field of chemistry. And computer science and automation systems are responsible for monitoring equipment and coordinating processes in the recycling production line; Finally, environmental scientists are responsible for evaluating the environmental impact of the whole recycling process and proposing appropriate solutions for waste gas, wastewater, solid waste, and noise management. This interdisciplinary and integrated approach is crucial for promoting the development of c-Si PV modules recycling technology.

(3) Improving relevant policies and regulations: A reasonable and comprehensive institutional system is a favorable guarantee for the recycling of EoL c-Si PV modules. Positive policy guidance and mandatory recycling requirements are necessary to strengthen the recycling of EoL c-Si PV modules. Developing industry or national standards for the quality requirements of recycled products, optimizing pollution prevention and control technical specifications, and emphasizing environmental management during the production process to ensure the standardization and sustainability of EoL c-Si PV modules recycling. At the same time, setting admission thresholds and qualification requirements for recycling companies to curb the disorderly production of informal small workshop firms and ensure the standardization of EoL c-Si PV recycling. Meanwhile, the producer responsibility system should be emphasized, and photovoltaic manufacturers need to take on more responsibilities related to photovoltaic module recycling, such as: developing photovoltaic modules that are easier to recycle and using recycled materials for the production of new photovoltaic modules. Furthermore, establishing specialized supervision departments, implementing responsibility mechanisms, strengthening supervision of the recycling process, enhancing the sense of responsibility of recycling enterprises, governments, and society, and jointly promoting the healthy development of the photovoltaic modules recycling industry.

(4) Establishing a complete recycling industry chain: This includes the establishment of a full process service units for the disassembly, collection, transportation, recycling production, and the final flow of recycled products. For large-scale photovoltaic power stations, the disassembly, collection, and transportation of EoL c-Si PV modules are relatively easy to manage and implement. However, for decentralized photovoltaic installation points (such as rural residential rooftops, lakes, and reservoirs [127], secondary or tertiary collection points and transportation units are necessary. For both centralized and decentralized photovoltaic power stations, a complete and professional collection network can improve the efficiency of collecting and transporting EoL c-Si PV modules. Furthermore, downstream enterprises of recycling companies, namely the purchasers and users of recycled products, are vital for the healthy and sustainable development of the entire recycling industry chain. Recycled products need to be accepted by downstream companies either for high-value utilization or as raw materials for secondary production. Therefore, it is necessary to strengthen the exploration and cultivation of these downstream enterprises to promote the effective utilization of recycled products.

(5) Providing appropriate economic subsidies: Currently, it is difficult to achieve economic benefits solely relying on the profits generated from the recycling of EoL c-Si PV modules, which hinders potential recyclers and investors [4]. Therefore, the government should actively intervene and establish a subsidy mechanism to provide appropriate subsidies and welfare support for recyclers. These subsidies can include forms such as funding subsidies, tax incentives, loan support, etc., aiming to reduce the operational costs of recyclers and improve their profitability, thus attracting more capital and enterprises to enter this field.

If the recommendations mentioned above could be implemented ideally, the author believe it will have a very positive impact on the healthy development of the photovoltaic recycling industry.

9. Conclusion

In order to effectively respond to the trend of retirement of c-Si PV modules, this study conducts a systematic analysis and evaluation of recycling technologies for EoL c-Si PV modules. Through a detailed analysis of the advantages and disadvantages of existing technologies, combined with the actual demand of the c-Si PV modules recycling market, this study proposes a series of suggestions and prospects, including optimizing recycling strategies, promoting interdisciplinary integration, improving relevant policies and regulations, establishing a complete recycling industry chain, and providing appropriate economic subsidies. Specifically, it is.

1. Recommend designing an efficient recycling scheme with universal applicability for broken c-Si PV modules, where the component sorting process requires more attention and the carbon emissions of the entire recycling process need to meet relevant requirements.
2. Consider appropriate recycling of low-value materials and high-value utilization of photovoltaic materials.
3. Emphasize the producer responsibility system for photovoltaic manufacturers and establish admission thresholds and qualification requirements for photovoltaic recycling enterprises.

In addition, the construction of a rational and comprehensive institutional system and policy guidance, as well as ensuring the integrity of the recycling industry chain, are of vital significance for the healthy and sustainable development of the EoL c-Si PV modules recycling industry.

CRediT authorship contribution statement

Pengxin Su: Writing – review & editing, Writing – original draft, Methodology, Data curation. **Yaqun He:** Writing – review & editing, Writing – original draft, Supervision, Methodology, Data curation. **Yi Feng:** Writing – review & editing, Writing – original draft, Methodology, Data curation. **Qiuyue Wan:** Writing – review & editing, Writing – original draft, Methodology, Data curation. **Tao Li:** Writing – review & editing, Writing – original draft, Methodology, Data curation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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