

End-of-Life Photovoltaic Modules

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More than 78 million tons of photovoltaic modules (PVMs) will reach their end of life (EOL) by 2050. If they are not responsibly managed, they can (a) pollute our terrestrial ecosystem, (b) indirectly encourage continuous mining and extraction of Earth's finite resources, and (c) diminish the net environmental benefit of harvesting solar energy. Conversely, successfully recovering them could reduce resource extraction and waste and generate sufficient economic return and value to finance the production of another 2 billion PVMs by 2050. Therefore, EOL PVMs must participate in the circular economy, and business and political leaders are actively devising strategies to enable their participation.

Keywords: life cycle ; circular economy ; circularity ; end-of-life ; solar energy ; photovoltaics ; energy decarbonization ; energy systems ; photovoltaic modules ; sustainability

1. Recovery Methods for End-of-Life Photovoltaic Modules

1.1. Physical Methods

1.1.1. Mechanical Method

The mechanical recovery method starts with manually dismantling and removing the junction box and aluminum frame from the end of life (EOL) photovoltaic module (PVM) . After disassembly, the remainder is crushed and sorted by its physical properties ^[1]. After crushing, the ethylene-vinyl acetate (EVA) particles are sieved using a vibratory separator at low temperatures ^[2]. Physical separation typically concludes after the EVA particles are sieved. While no adverse environmental effects arise from physically taking the EOL PVM apart, the first mechanical recovery stage produces low-purity recyclates that need further sorting into their elements.

Additional separation steps may include flotation, electrostatic separation, eddy current separation, and mechanical screening ^[3]. The flotation method separates each substance by density, producing crushed glass sizes between 45 and 850 μm ^[4]. In comparison, electrostatic separation separates each substance by its electrical conductivity and properties ^[5]. The recyclates can be segregated into conductors (e.g., metals), semiconductors (e.g., silicon powder), and insulators through electrostatic action. A practical drawback of electrostatic separation is that glass is often separated into conductors alongside metals. Thus, electrostatic separation remains ineffective and needs further improvement before enjoying widespread adoption.

On the other hand, the eddy current separation method uses an electric vortex separator to separate the crushed recyclates. The elements are separated by their conductivity and shape ^[6]. The magnetic drum in the electric vortex spins at high speed to create an alternating magnetic field, creating an eddy current in the conductive metal as it passes through the field. The two opposing magnetic fields create a repulsive force (Lorentz) that separates the metal from the crushed recyclates. The recyclates' particle size must be optimized for this process to dissociate metals from the EVA remnants effectively.

Lastly, the mechanical screening method screens and sorts the recyclates by size, shape, thickness, and other differences. This method achieves more than an 85% recovery rate for silicon despite the inherent difficulty of separating the crushed recyclates mechanically. Correspondingly, the mechanical screening method is preferred as it is practical, requires the least energy, and requires no chemical reagents.

Recently, scholars have investigated the effectiveness of high-voltage pulse crushing, which has a higher selectivity than traditional mechanical crushing methods. High-voltage pulse crushing separates copper, aluminum, lead, silver, and tin ^[7] ^[8]. As components in the EOL PVM have different dielectric constants, conductive channels among components are formed and respond to an electrical breakdown in a high-voltage discharge. During an electrical breakdown, large amounts of high-temperature and high-pressure plasma expand the channels, leading to the selective separation of each

component ^[4]. A higher voltage (>100 kV) and pulse frequency are needed for high-voltage pulse crushing to be effective. However, the modern-day environment does not allow these conditions to be met, resulting in limited applications.

1.1.2. Pyrolysis

Pyrolysis, or heat treatment, is another physical EOL PVM recovery method. EOL PVMs are placed in furnaces with elevated temperatures above 400 °C to obtain clean PV cells ^[9]. At the same time, the EVA film, backplane—often Tedlar Polyester Tedlar (TPT) based—and other organic matters are decomposed, leaving the remnants of the aluminum frame, tempered glass, and PV cells. Unlike the mechanical recovery method, there is no crushing involved. Hence, the glass and PV cell remnants remain intact and can be recycled as it is ^[10]. Hydrometallurgy is then used to remove the metal electrodes and impurities from the remnant PV cells to acquire high-purity silicon (99.999%) ^[11].

As pyrolysis aims to decompose EVA films thermally, many studies have focused on its processes, products, and related mechanism. Scholars have observed no correlation between the heating rates and the weight of EVA films. Therefore, this suggests that EVA films can only be removed at an optimized temperature. The EVA removal process involves deacetylation (organic intermediate formation) and long-chain fracture with free radical reaction (hydrocarbon formation). The former mainly produces acetic acid and a small amount of acetaldehyde by deacetylation. Then, the latter carbonizes and degrades the organic intermediate to form the hydrocarbon. Recent investigations have also found that by combining nitrogen pyrolysis and vacuum decomposition, targeted hydrocarbon (such as linear alkenes, alkanes, and their isomers) could be recovered at a 100% organic conversion rate as the EVA decomposes ^[12].

Overall, pyrolysis efficiently separates glasses, PV cells, and backplanes. It is also a relatively simple treatment process. However, it is energy-intensive and requires high-specification equipment to operate. Furthermore, its by-products may adversely impact the environment and business viability. For example, heat-treating EVA in an oxidizing atmosphere may lead to the oxidization of silicon-based cells, which affects the quality of the silicon recovered.

Additionally, heat-treating EVA film and TPT backplane at a high temperature can produce a large amount of organic matter that pollutes the environment. Therefore, future studies using pyrolysis for EOL PVMs must focus on its industrial-scale viability and environmental aspects. It must also account for current trends, including the ongoing transition toward thinner polysilicon wafers—from 550 to 180 μm—which scholars speculate would increase the wafer's risk of breaking during pyrolysis ^[13].

1.2. Chemical Methods

Compared with the physical treatment methods, chemical treatment methods are relatively simple and consume less energy. Inorganic and organic solvent dissolution are both chemical treatment methods.

1.2.1. Inorganic Solvent Dissolution

The inorganic solvent dissolution method requires the EOL PVM to be immersed in an acid or alkali solution. Immersion dissolves the EVA film, leaving behind the PV cell sheet. Nitric acid and hydrofluoric acid are commonly used solvents. The process removes metal impurities and the silicon nitride anti-reflective layer (ARL) on the cell's surface to recover metal electrodes and high-purity silicon. Two methods have been developed to remove metal impurities from the cell's surface ^[13].

The first is a pure-acid method. First, phosphoric acid removes the aluminum electrode and silicon nitride ARL. Next, nitric acid and hydrofluoric acid mixture are applied to remove silver and other metal impurities from the cell's surface. Lee et al. ^[14] found that a mixed acid solution of nitric acid and hydrofluoric acid at a ratio of 83:17 can efficiently reclaim cleaned high-performing silicon wafers.

The second method uses both acid and alkali. Nitric acid and hydrofluoric acid mixture are applied to remove the silicon nitride ARL and the silver electrode. Following this, sodium hydroxide is used to remove the aluminum electrode. The drawback of this method is that it produces grooves on the surface of the silicon wafers, which unfavorably affects its performance for future applications. Additionally, as nitric acid is not adept at penetrating the EOL PVM, removal of the EVA film using this method is much slower. However, studies suggest that exposing nitric acid to higher temperatures could remove the EVA film faster. For instance, Klugmann-Radziemska and Ostrowski ^[11] reported that heating nitric acid to 40 °C during the dissolution process is critical to splitting each major EOL PVM component apart to extract the silicon wafer. In addition to increasing the temperature of nitric acid, scholars have explored various workarounds. Their proposed alternatives have primarily centered on nitric acid, potassium hydroxide, and phosphoric acid to remove all unwanted components to extract the silicon wafer. Regardless of the chosen route, the obtained silicon wafers via both

acid and alkali are reported to be of satisfactory quality to recreate lead-free PVMs with a comparable conversion efficiency to those made from new silicon wafers ^{[13][15]}.

Overall, while the inorganic solvent dissolution method is a fast and straightforward process with low energy consumption and high efficiency, the successful extraction of all valuable metals for recycling is not guaranteed. Additionally, as hydrofluoric acid is highly toxic and corrosive, the by-products from this process can pollute the environment. Similarly, nitric oxide and nitrogen dioxide are also aftereffects of this process; thus, equipment is needed to treat the tail gas.

1.2.2. Organic Solvent Dissolution

The EVA film expands and dissolves while being soaked in different organic solvents. While soaked, the various components are exposed and separated, allowing the PV cell sheet to be extracted. The expansion and dissolution reactions correspond to the crosslinked and non-crosslinked parts of the EVA films. The expansion process widens the gap between the tempered glass and the silicon panel while the dissolution process dissolves the EVA film into liquid. Studies have noted that EVA's fluidity in organic reagents increases as the reaction temperature increases. Other studies have noted that the dissolvent could easily overcome EVA's adhesive properties, thus allowing the EOL PVM to be separated and recycled ^[16].

Additionally, by comparing how the EVA film swells in different organic solvents, studies have suggested that the glass and silicon PV cells can be separated from the rest of the EOL PVM after two days of soaking ^[17]. As this method is relatively slower, Kim and Lee ^[16] developed an ultrasonic-assisted organic solvent method to accelerate the dissolution of EVA films. With their process, the EVA films could be wholly dissolved in o-dichlorobenzene within 30 min at 70 °C and 900 W irradiation power to recover PV cells in good condition.

Successful recovery of the EOL PVM depends on whether the EVA film can be thoroughly removed without implicating the rest of the valuable components. The organic solvent method is effective in this scenario as it only targets and treats the EVA film. However, its effectiveness depends on how tightly bonded and sticky each layer of PVM is. The tighter and stickier the layers are, the harder it is for the organic solvent to diffuse, penetrate, and react accordingly. Apart from its slow dissolution rate, it is worth noting that most organic solvents are toxic, volatile, expensive, and difficult to dispose of after use, affecting their feasibility for large-scale EOL PVM recycling efforts.

1.3. Physical-Chemical Method

As discussed earlier, both the physical and chemical methods have merits and drawbacks. Their primary limitations are their degree of separation and recovery. For example, the physical method is not adept at thoroughly separating the EOL PVM into its elemental components, while the chemical method is only effective at removing EVA films. Hence, the physical-chemical method was developed to take advantage of the merits while leaving the drawbacks behind.

Generally, the mechanical recovery methods are first used to pre-treat the EOL PVM. This process crushes and splits the EOL PVM into fragments between 0.08 and 0.4 mm. These tiny fragments cause the embedded EVA film to lose its adhesiveness. It also negates EVA's influence on metal leaching ^[18], thus paving the way for the fragments to be directly soaked in an acid leaching reagent consisting of nitric acid, sulfuric acid, and hydrogen peroxide at 60 °C. Glass will not be dissolved during this process and can be recovered directly ^[19]. In contrast, valuable metals are recovered with hydrometallurgical techniques ^[20]. As the EOL PVM has been pre-treated, the amount of chemical reagents needed for hydrometallurgy is significantly reduced.

As the mix of physical and chemical methods is increasingly accepted and adopted in contemporary environments, scholars have continuously worked with the industry to refine the method's efficiency. For example, Klugmann-Radziemska et al. ^[21] used the pyrolysis and chemical dissolution methods to recover clean silicon wafers from EOL PVM. Their process pre-treats and separates the cell sheet, backplane, cover glass, and aluminum frame using heat before using chemical reagents to remove the ARL and electrode coating on the surface of the cell sheet. On the other hand, Kang et al. ^[22] used organic solvents to isolate and recover the glasses before using pyrolysis to remove the EVA film and obtain the silicon. The recovered silicon was then chemically etched to remove all the metallic impurities on its surface to obtain 99.999% high-purity silicon. Tao and Yu ^[23] also noted that some industry actors used pyrolysis to remove plastics in the EOL PVM before manually separating the remaining remnants. They reported that this process, etching, and other further treatments could recover up to 84% of the EOL PVM. Additionally, they reported that the heat from the incinerator could be recycled for other processes, including preheating.

While most works have focused on refining the process, other scholarly works have focused on the interactions between the materials and the processes. For example, Pagnanelli et al. [24] attempted to understand and prescribe the optimal treatment methods based on the size of the crushed EOL PVM recyclates. They reported that recyclates with 0.4- and 1.0-mm particle sizes could be directly recycled. In contrast, recyclates with a particle size greater than 1.0 mm needed to be treated by pyrolysis to separate the EVA film and glass. For tiny recyclates (particle size smaller than 0.4 mm), chemical methods were prescribed to dissolve the metals and recycle the glass. They reported that 90% of the EOL PVM can be recovered by categorizing and treating the recyclates by size.

All in all, the physical-chemical method has a high waste utilization and recovery rate. When optimized, it can increase the specific surface area of metals, shorten their leaching time, and reduce chemical reagents' use. Hence, the researchers feel the industry will be steered towards using a mixed physical-chemical method to treat EOL PVM. However, as researchers actively investigate and accelerate industrial adoption of physical-chemical methods, one must be aware of its drawbacks. Presently, mechanically crushing the silicon cell into silicon powder increases the area of contact with air, leading to higher levels of silicon oxidation. Hence, a more potent reducing agent and more energy are needed to mitigate this issue in subsequent extractions. Additionally, D'Adamo et al. [25] noted the current state of the art as complex, energy-intensive, and generating waste liquids. Therefore, the researchers believe a viable and widely accepted solution must be simple and energy-efficient.

2. Recovery of Metals in End-of-Life Photovoltaic Modules

PVMs contain valuable metals. These metals must be recovered and reconditioned for future uses to participate in the circular economy upon reaching their EOL. These metals include silver and scattered metals.

2.1. Silver

Most studies suggest nitric acid is critical in recovering silver from EOL PVMs [26]. For example, Simon et al. [27] extracted silver powder using a multi-step process that involves dissolving nitric acid, precipitating silver chloride, co-heating sodium hydroxide, and hydrazine hydrate reduction. After extracting the silver powder, he used electrolytic refining techniques to obtain 99.99% pure silver. The EOL PVM must be pre-treated before the process to attain its desired outcomes. It needs to be crushed into recyclates of less than 0.5 mm. The fragments are then etched with nitric acid, allowing the silver particles to be precipitated and form silver chloride in the subsequent stages [18]. Other studies also demonstrated the possibility of attaining a 100% leaching recovery of silver by utilizing the nitric acid leaching reagents under optimal leaching conditions [28].

To reduce environmental pollution from the extraction of silver, Yousef et al. [29] proposed milling the EOL PVM to destroy the chemical bonds between the metal electrode and silicon layer before using nitric acid with a 60% or higher concentration to leach out silver. Following this, they proposed using a microfiltration process to extract the silicon particles and purify them with hydrofluoric acid etching to remove silicon nitride contamination. Lastly, centrifugation was proposed to separate the aluminum nanocrystals from the leached solution before adding hydrochloric acid to recover silver chloride from the remaining saturated solution.

2.2. Scattered Metals

Compound semiconductor solar cells such as gallium arsenide (GaAs) [30], cadmium telluride (CdTe) [31], indium phosphide (InP), and copper indium gallium selenide (CIGS) [32] are emerging alternatives to silicon-based PV cells. These emerging alternatives use scattered metals such as indium, tellurium, gallium, and selenium as their base material. Scattered metals are classified as critical metals and are limited in quantity. Given the current landscape, scattered metals will likely face material supply constraints when their global installed capacity exceeds a terawatt [33]. Hence, ensuring these emerging alternatives participate in the circular economy is crucial to developing and scaling next-generation solar energy systems. The means to recover them from their EOL PVMs include acid leaching, solvent extraction, and vacuum distillation.

3. End-of-Life Photovoltaic Module Recovery Process

3.1. Full Recovery End-of-Life Photovoltaics (FRELP)

The first process focused on maximizing material recovery from EOL PVMs. This concept was first mooted by the European Union (EU). In 2012, the EU changed its Waste Electrical and Electronic Equipment (WEEE) Directive to include PVMs [34]. With this enactment, all PVMs were effectively banned from being landfilled and mandated to be

recycled. At this time, there were limited means to recycle PVMs effectively and responsibly. Therefore, the EU funded research efforts to address this gap to ensure policy effectiveness and stakeholder compliance. Their efforts cumulated in the Full Recovery End of Life Photovoltaics (FREL P) project, which attempted to create a process to recover as many functional components from EOL PVMs as possible [35].

Ideally, the FREL P project partners aspired to achieve an economically viable method to recover 100% of EOL PVMs [36]. Based on the latest publicly available information, the FREL P project is in the pilot-scale stage, with a facility capable of treating roughly 1300 PVMs daily. However, the plant has stopped operating due to the insufficient and inconsistent volume of EOL PVMs arriving at its facility [37].

The FREL P process follows a four-phase physical-chemical treatment method [38]. In each phase, selected components are recovered through their appropriate means. For example, aluminum and connectors are recovered using mechanical detachment systems in the first phase. In the subsequent phases, acid, pyrolysis, and electrolysis are applied to recover silicon and other metals [39]. Recent reports have suggested that the FREL P process can recover more than 95% of glass, aluminum, silicon, and copper from EOL PVMs [35]. It can also recover 94% of silver but not tin or lead [37]. Compared to merely removing the aluminum frame and cables and shredding the rest, the FREL P process results in a 10% to 15% overall improvement in impact categories such as global warming potential, ozone depletion, and ecotoxicity [35].

However, the FREL P process is not perfect. While waste from EOL PVMs has been drastically reduced, it is not eliminated. For every ton of EOL PVMs that underwent the FREL P process, there will be close to 30 kg of residues that are landfilled or lost [35]. Additionally, the quality of silicon and metals recovered through the FREL P process is deemed inferior. As a result, this diminishes its economic value and makes it unsuitable for reuse in new PVMs. To this extent, experts estimated a return of only USD 3 on each PVM recovered through the FREL P process, which is less than its per-unit cost of recovery [39]. This means that loss is incurred in every unit of EOL PVMs recovered through the FREL P process. With poor unit economics, EOL PVM recycling through the FREL P process will not take off.

3.2. Arizona State University (ASU) Method

For EOL PVM recycling to take off, it must be a commercially viable and profitable pursuit. Recognizing this proposition, a team at the Arizona State University proposed a different strategy to recover EOL PVMs. Instead of maximizing the mass of materials recovered, they proposed strategies to maximize the value of materials recovered. Dubbed the ASU method, this process is estimated to produce a return of USD 13 on each EOL PVM recovered [39] without leaving much behind for landfilling. At USD 13 revenue per unit, this recovery model is suitable for private facilities to operate profitably without relying on state support [40].

The ASU method is also a multi-step process that uses physical-chemical treatment methods. It involves sequential electrowinning, using a heated nitric acid solution to extract and recover multiple metals from the highly compacted and interconnected PV cell. Following this, a secondary process called sheet resistance monitoring is applied to recover solar-grade silicon. While the ASU method does not recover the maximum mass, its process can recover metals of 99% purity and silicon that meet the standard for the solar grade. It can also recover silver, the most valuable solar material component, at 99% purity [39]. This explains the significant difference in the recovered material value between the FREL P and ASU methods.

As the ASU method is a recent discovery, its drawbacks and feasibility of operating sustainably at an industrial scale have yet to be thoroughly investigated. Nonetheless, even at a laboratory scale, where the conditions and feedstock are controlled, the ASU team has successfully demonstrated the technical and financial possibility of EOL PVMs being responsibly and effectively recovered.

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