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Master Thesis

Secondary Resource Potential and Design-for-Recycling of Silicon- and Chalcogenide-based Photovoltaic Panels

submitted by

Dina Mūsiņa, BSc

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Supervisor:
Ao.Univ.Prof. Dipl.-Ing. Dr.nat.techn. Stefan Petrus Salhofer
Institute of Waste Management
Department of Water, Atmosphere and Environment

Affidavit

I hereby declare that I have authored this master thesis independently, and that I have not used any assistance other than that which is permitted. The work contained herein is my own except where explicitly stated otherwise. All ideas taken in wording or in basic content from unpublished sources or from published literature are duly identified and cited, and the precise references included.

I further declare that this master thesis has not been submitted, in whole or in part, in the same or a similar form, to any other educational institution as part of the requirements for an academic degree.

I hereby confirm that I am familiar with the standards of Scientific Integrity and with the guidelines of Good Scientific Practice, and that this work fully complies with these standards and guidelines.

Vienna, 25.02.2022

Dina Mūsiņa

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I would like to thank everyone who accompanied me during my master thesis.

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Without the considerable support and motivation from my family, I would probably not have studied in the first place. Also, I would like to thank my friends who selflessly proofread parts of this work. Indeed, thank you Leona for reading this right now! And I thank all my fellow students and BOKU employees who made my time at BOKU so cherishable. BOKU is indeed a university of life and has taught me way more than the pure contents of the study program.

Kurzfassung

Im Zusammenhang mit der Erreichung der Pariser Klimaziele steigt die Anzahl der installierten Photovoltaikanlagen weltweit an. Gleichzeitig ist ein Ausbau von selbiger geplant. Die damit einhergehenden wachsenden Abfallmengen stellen ernste Umweltrisiken für die Zukunft dar. Sowohl Produktion, als auch Recycling von Leiter- und Halbleitermaterialien sind sehr energie- und rohstoffaufwendig.

Photovoltaikmodule (PV-Module) enthalten viele Wertstoffe, unter anderem auch kritische Rohstoffe mit hohem Versorgungsrisiko für die EU. Dadurch wird eine geeignete Verwertungsmethode noch wichtiger, um die Materialien möglichst im Kreislauf behalten zu können. Zurzeit ist Recycling die umweltfreundlichste Lösung von allen zur Verfügung stehenden Maßnahmen für die ausgedienten PV-Module. Derzeit ist die Recyclingquote weltweit jedoch noch sehr gering.

In dem letzten Jahrzehnt erreichten die ersten größeren kommerziellen Photovoltaikanlagen das Ende ihrer Lebensdauer. Große PV-Abfallmengen sind laut International Renewable Energy Agency und International Energy Agency (2016) ab dem Jahr 2030 zu erwarten. Geschätzt liegen diese Abfallmengen weltweit zwischen 0,4 und 1,3 Mio t pro Jahr. Die aktuellsten Daten aus dem Jahr 2016 schätzten die kumulative weltweite Abfallmenge auf 43.500-250.000 t. Aufgrund der derzeit geringen Abfallmengen ist die stoffliche Verwertung noch nicht vollends ausgereift und wird noch nicht ausreichend umgesetzt. Infolgedessen werden PV-Module in Entwicklungsländern deponiert.

Für diese Masterarbeit wurde eine umfassende Literaturrecherche durchgeführt, um den aktuellen Stand der Technik im Recycling und Design-for-Recycling-Konzepten von kristallinen Silizium- und Dünnschicht-PV-Modulen zusammenfassend darzustellen. Zusätzlich wurden Expert*innen-Interviews durchgeführt. Der Aufbau und die Materialzusammensetzung von den untersuchten PV-Modultypen spielen hierbei eine entscheidende Rolle. Der Fokus dieser Arbeit liegt auf Verfahrenstechniken, mit denen diese PV-Modultypen für das Recycling aufbereitet werden können. Die Technologien, die sich derzeit noch in der Entwicklungsphase befinden, werden ebenfalls bedacht. Design-for-Recycling-Konzepte werden zusätzlich von bestehenden Modultechnologien abgeleitet, wie zum Beispiel der Einsatz von Glaspaste anstatt von Polymeren als Randabdichtung oder von Fluorpolymer-freien Rückseitenfolien. Die Hypothese besagt, dass die Recyclingfähigkeit der Module mit Design-for-Recycling-Konzepten verbessert werden könnte.

Die Entsorgung von ausgedienten PV-Modulen besteht aus mehreren Schritten: Demontage, Sammlung, Vorbehandlung und Nachbehandlung. Die kommerziell eingesetzten Vorbehandlungstechnologien führen nicht alle technisch möglichen Materialien zurück oder führen zu einer niedrigeren Qualität der Rezyklate als es möglich wäre. Es wird an Alternativen weitergeforscht, beispielsweise an der Anwendung von Blitzlampen, um die Laminate aufbrechen bzw. Kompositmaterialien trennen zu können. Die Nachbehandlungsmethoden in Form von hydrometallurgischen Verfahren sind in der Industrie ausgereifte Prozesse. Im Zuge der unterschiedlichen Recyclingtechnologien können Aluminium, Glas, Silizium, Kupfer, Polymere, Silber, Cadmium, Tellur, Indium und Selen zurückgewonnen werden.

Das Recycling der untersuchten PV-Module wird aufgrund der eingesetzten Verbundwerkstoffe und der nötigen Delaminierung der Materialverbunde erschwert.

Eine mögliche Verbesserung zur Wiederverwertbarkeit bei den untersuchten PV-Modulen ist die Substitution der Verbundwerkstoffe durch Materialien wie Glas, Polyolefine, Polyamide, Polyester oder thermoplastische Silikonelastomere. Eine weitere Verbesserungsmöglichkeit ist die Substitution der Lamination durch Vakuum oder Klebstoff-Pins. Tests belegen die Haltbarkeit solcher Modulaufbauten. Im Zuge dieser Studie wurden nur wenige zusätzliche Design-for-Recycling-Konzepte gefunden, nämlich alternative Klebstoffe zum Abdichten, Substitution von Blei und alternative Einkapselungsmethoden. Im Sinne der Circular Economy und in Hinblick auf die weltweit zunehmenden PV-Abfallmengen sollte daher die Weiterentwicklung von Design-for-Reuse- und Design-for-Recycling-Konzepten („Ecodesign“) in der PV-Industrie vorangetrieben werden.

Abstract

The number of installed photovoltaic (PV) systems is increasing worldwide to adhere to the Paris Climate Agreement. A further future increase of PV system instalments is planned. Due to this increase, the growing amounts of end-of-life PV panels pose serious environmental risks for the future. Both production and recycling of conductor and semiconductor materials are very energy- and raw material-intensive.

PV modules contain many valuable materials, including critical raw materials with a high supply risk for the EU. This makes appropriate resource utilization at the end of life even more important in order to keep materials in circulation as long as possible. Currently, recycling is the most environmentally friendly solution of all available end-of-life options for the discarded PV modules. However, the recycling rate worldwide is currently still very low.

The first commercial-sized PV installations have reached their end of life in the last decade. Large amounts of PV panel waste are anticipated by the early 2030s and are estimated to lie between 0.4 and 1.3 Mio t annually. The latest assessment of the global PV waste streams estimates the cumulative PV waste volume to have reached 43,500-250,000 metric tons by the end of 2016. Recycling of PV panels is not sufficiently developed and implemented mainly due to the currently low PV panel waste streams. Consequently, PV panels are landfilled in developing countries posing an environmental burden and waste of resources.

The aim of this master thesis is to obtain an overview of the current status of crystalline silicon and chalcogenide-based PV panel recycling technologies, and design-for-recycling concepts for these panel types. This is done through a combination of literature review with semi-structured interviews by experts from the field. The structure and material composition of the investigated PV module types play a decisive role in achieving the aforementioned aim. The focus lies on technologies intended to process these PV module types for recycling. Both the commercially implemented technologies and the ones under research and development are subjects of this study. Design-for-recycling concepts are additionally derived from existing module technologies, e.g. the use of glass paste instead of polymers as edge sealant and the use of backsheets without fluoropolymers. The hypothesis is the possible improvement of module recyclability through design-for-recycling.

The disposal of discarded PV modules consists of several steps: disassembly, collection, pre-treatment and post-treatment. The commercially used pre-treatment technologies either do not result in all technically recoverable materials or result in lower quality recyclates than technically feasible. Research activities are ongoing in the search for alternatives, such as the use of flash lamps for delamination. Post-treatment methods in the form of hydrometallurgical processes are mature processes in the industry. Aluminum, glass, silicon, copper, polymers, silver, cadmium, tellurium, indium and selenium can be recovered through the use of various recycling technologies.

The recycling of the investigated PV modules is impeded due to the composite materials and the necessary delamination. One possible improvement for recyclability of the PV modules in question is the substitution of the composites with materials such as glass, polyolefins, polyamides, polyesters or thermoplastic silicone elastomers. Another possibility is the substitution of lamination by other technologies, e.g. vacuum or adhesive pins. Tests have proven the durability of such modules. Few additional design-for-recycling concepts have been found during the course of this study, namely

alternative adhesives for sealing, the substitution of lead, and alternative encapsulation methods. In the sense of the circular economy and in view of the increasing PV waste volumes worldwide, the further development of design-for-reuse and design-for-recycling concepts ("ecodesign") should be promoted in the PV industry.

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Abbreviations

a-Si	amorphous silicon
c-Si	crystalline silicon
CdTe	cadmium telluride
CdS	cadmium sulfide
CIGS	copper indium gallium selenide
CIS	copper indium selenide
CRMs	critical raw materials

DEMOC	double encapsulation module with optical coupler
ECAs	electrically conductive adhesives
EVA	ethylene vinyl acetate
ITO	indium-tin oxide
R&D	research and development
PET	polyethylene terephthalate
PV	photovoltaic
POE	polyolefin
PVF	polyvinyl fluoride
WEEE	waste electrical and electronic equipment

1. Introduction

Photovoltaic (PV) technology is expected to play an important role in the clean energy transition. Its impact on the environment is one of the lowest of all energy and electricity generation technologies (International Energy Agency, 2018). Since PV technology has become economically competitive in the energy market, PV deployment is expanding exponentially with national plans to increase the PV capacities even further to reach the ambitious targets of the clean energy transition.

At the end of 2020, the worldwide installed PV capacity has reached 707.5 GW_p with China being the biggest market with more than 250 GW_p installed (Our World in Data, 2021). Asia and Pacific together have around 420 GW_p installed, Europe has installed more than 160 GW_p while United States has more than 70 GW_p of solar PV.

Together with this expansion comes a respective increase in waste PV panel quantities. According to the waste management hierarchy, if a product cannot be repaired or reused, recycling is preferred over disposal. Reuse and repair of PV panels are currently extremely limited (Chowdhury et al., 2020; Allesch et al., 2019). Different panel types and the rapid development of PV technology impede reuse since the PV panels are electrically connected into strings that need to be calculated and planned. For an easier planning of PV arrays, one PV panel type is used making the reuse of panels generally unpractical. Design of panels impedes repair because the panels are typically laminated which makes an exchange or repair of a component practically impossible. Consequently, recycling is currently the most environmentally friendly end-of-life management option for all so far installed and manufactured panels. If recycled, impacts associated with toxicity are greatly reduced, secondary raw materials are used for generating new products, thus virgin materials and energy is saved, and generally less waste is landfilled (Vellini et al., 2017). Studies confirm this by concluding that recycling is the most environmentally friendly option for PV panel waste management (ISWA and IPV, 2012).

Design of panels also impedes their recyclability. Panels are designed in a way that ensures their durability while making their disassembly harder at the same time. Therefore, design-for-recycling is important to facilitate the recyclability of PV panels.

This study aims to get an overview of the current status in crystalline silicon (c-Si) and thin-film PV panel recycling technologies and design-for-recycling concepts for these panel types. Both the commercially implemented technologies and the ones under research and development (R&D) are subjects of this study. This paper uses a literature review combined with semi-structured interviews by experts from the field to reach this aim. Design-for-recycling concepts have been either derived from the existing c-Si PV technologies or found in the literature as existing design-for-recycling concepts.

The superior aim is to contribute to the minimisation of the environmental impact of PV panels through environmentally friendly design and prevention of landfilling. The possibility of keeping the secondary raw materials in the material circle as effectively as possible contributes to this aim too.

The research object is the PV panel itself. Balance of system components, junction boxes and cables are not part of this study. Organic photovoltaic cells have been omitted due to their early development state and their negligible market share. Manufacturing waste is not explicitly reviewed, however is included if the literature reviewed for the waste PV panels includes manufacturing waste.

The research topics of this study are the various steps of the pretreatment, namely the delamination and material separation technologies. The process of waste panel dismantling and the recycling process itself are beyond the scope of this master thesis. Metal extraction and refining after the pretreatment steps is outlined for the sake of a complete description of some recycling pathways.

The basic assumption of this study is the urgency of further development of the PV panel recycling technologies together with the improvements of the design-for-recycling of the panels because the current conventional designs impede their recyclability.

1.1 Research questions

This master thesis aims to answer following research questions:

1. What are the estimates for the global PV panel waste volumes currently and the projections thereof?
2. Which recycling technologies are currently implemented and being researched for the silicon-based (c-Si) and thin-film panels? Which secondary raw materials are obtained? An overview with a list of the companies/research institutions, including their geographical location, will be created.
3. Which design-for-recycling concepts have been developed for the selected PV technologies and which materials are used in those? These concepts will be either derived from the existing unconventional c-Si PV technologies or existing design-for-recycling concepts (best case practice) will be compiled.

2. Description of PV technologies

Jäger-Waldau (2016) estimates that around 92% of the commercial modules on the market are Si-based. Thin-film modules take up 8% of the market share of which 5% for cadmium telluride (CdTe) modules, 2% for Copper Indium Gallium Selenide (CIGS) modules and less than 1% for amorphous silicon (a-Si) modules. Since a-Si technology has such a small market share and is predicted to be phased out before 2030 (International Renewable Energy Agency and International Energy Agency, 2016), this technology is not considered in this master thesis.

The module type that is typically chosen to be installed commercially is a compromise between its market price and efficiency. As it is visible in Figure 1, the silicon- and chalcogenide-based panel types are one of the most efficient ones explaining their leading market position.

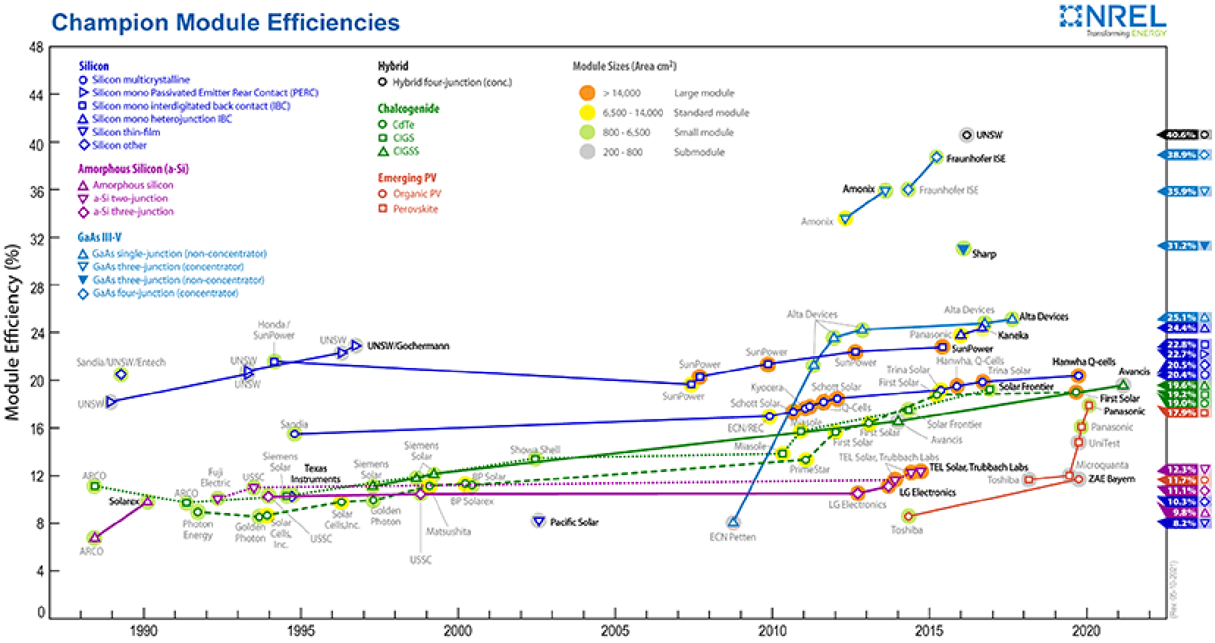
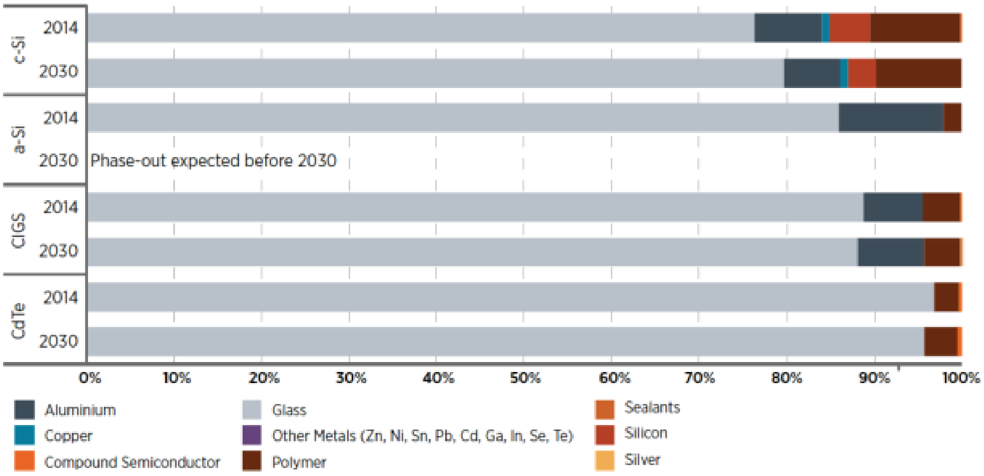


Figure 1: Efficiency of different PV module types (Green et al., 2020)

2.1 Material composition of PV modules

Before explaining the recycling processes and design-for-recycling concepts, it is important to understand the structure and material composition of PV panels first.

The design of the modules is different even within the same PV module family, which is the reason why no general listing of the material composition can be made. An approximation is used in the following subchapters and an overview of the material composition by PV panel technology is given in Figure 2.



Based on Marini et al., (2014); Pearce (2014); Raithel (2014); Bekkelund (2013); NREL (2011) and Sander et al., (2007)

Figure 2: Evolution to 2030 of materials used for different PV panel technologies as a percentage of total panel mass (International Renewable Energy Agency and International Energy Agency, 2016)

2.1.1 Crystalline Silicon (c-Si)

Crystalline silicon (c-Si) cells are obtained from slices of silicon that are called wafers (Casini, 2016). They are 160–240 µm thick, cut from a single crystal or a block. Depending on the Si wafer manufacturing process, three main types of c-Si cells can be produced:

- Monocrystalline
- Polycrystalline/multicrystalline
- Ribbon /sheet c-Si

Maani et al. (2018) made an overview of the percentage of the total mass of the solar panel each component takes up. Most of the c-Si solar panels consist of more than 68% of glass. It protects solar cells from damaging external influences, such as ultra-violet radiation, humidity and vapour penetration, dryness, wind, dust, sand, and chemicals (Roekens-Guibert, 2007). At the same time, glass transmits as much sunlight as possible into the panel. Some c-Si panels have glass on both sides, however, most of the time c-Si panels are manufactured as glass-foil panels, meaning that the bottom side of the panel is protected by a backsheet. A backsheet is the last layer and is typically made of a polymer or a combination of polymers such as polyvinyl fluoride (PVF) and polyethylene terephthalate (PET) (Bradley, 2015). It protects the solar panel from external factors.

The ethylene vinyl acetate (EVA) layer is a transparent polymeric resin that protects the solar cell from moisture, dirt, ice, and other conditions expected during operation. It is used as an adhesive between the glass and the Si wafer. EVA accounts for about 7% of the total mass of c-Si panels. Other materials such as liquid silicone rubber or polyolefin (POE) can be used as cell encapsulants as well, however, they are used rarer than EVA (personal information, 2014-2018).

Si is the light absorber layer and takes 4% of the total mass of the panel. Si wafers are connected to the backsheet by soldering copper wires onto them. Sn, Pb and Ni are used for the coating (Scherhaufer et al., 2020). In the case of glass-glass modules, Si wafers are encapsulated by EVA or other encapsulation material which is attached to the rear glass plate.

The Si cells have an anti-reflective coating on the front side consisting of silicon nitride. Metal pastes containing Ag, Al, Pb and Cd create the electrical connection between the front and the back side of the solar cells.

It is common to use Pb/Sn solder to attach tabbing/ribbons to the cells and busbars as is seen in Figure 3.

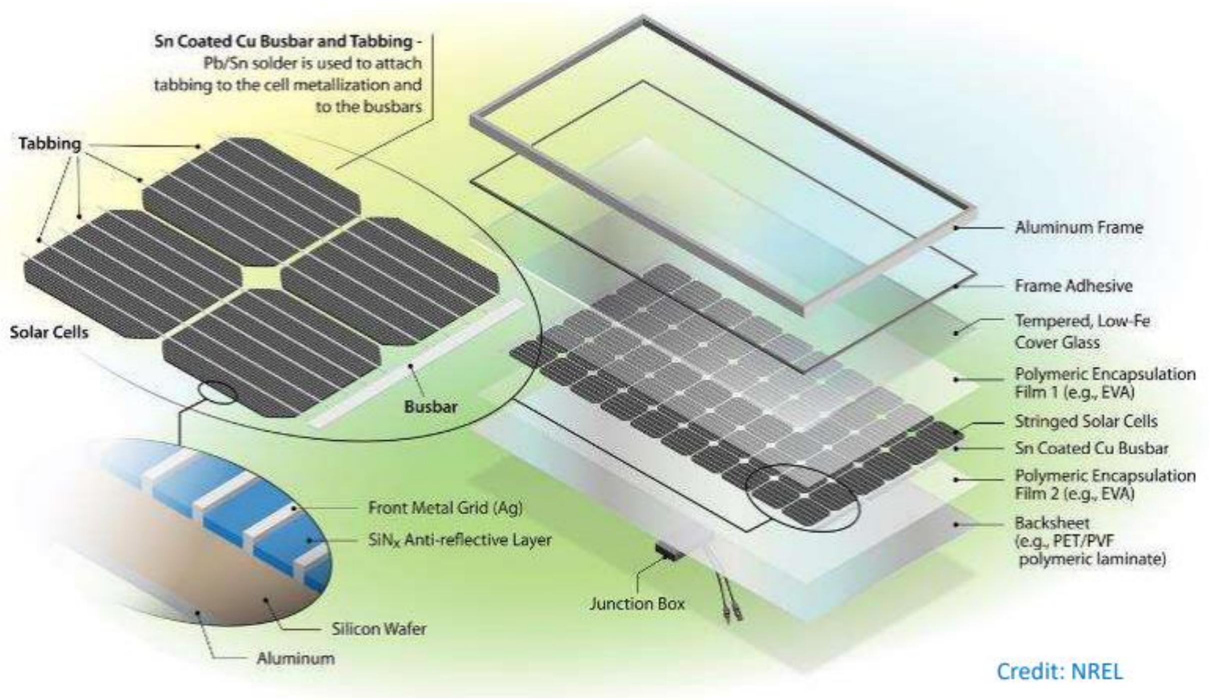


Figure 3: Typical structure of c-Si PV panels (NREL, 2019)

A junction box is attached to the backside of the panel for electrical connection. The electrical connections are made up of Ag in combination with Cu, Ni and Fe that together make up 1.5 % of the total mass (Grandell and Thorenz, 2014).

The aluminium frame gives mechanical stability for the laminates (frameless panels). Aluminium makes up 17% of the solar panel mass and the frame also includes Mg in the form of aluminium alloy ($AlMg_3$) (Domínguez and Geyer, 2017).

2.1.2 Cadmium Telluride (CdTe)

International Energy Agency (2018) states that an important difference between the c-Si and compound technologies is the deposition of the compound semiconductor layer directly on the substrate glass as opposed to the c-Si cells being separated from cover glass and backsheet or back glass.

Maani et al. (2018) summarized the portions of each material mass used in the CdTe solar panels. Glass makes up 92% of the CdTe panel mass. Glass protects the laminate from damaging external factors, e.g., water and dirt. The glass is also mostly used as the substrate meaning that the CdTe layers are deposited on it (Eiffert et al., 2009).

The CdTe itself takes up 0.12% of the total panel mass. It is the light-absorbing layer and generates charge carriers. This layer is encapsulated between two glass plates or sturdier backsheets are being used that cater for enough mechanical stability. Therefore, CdTe panels do not need an aluminium frame. This not only reduces manufacturing costs but also improves the electrical isolation of the panel (Ridge et al., 2017). An unintended conduction path for leakage currents can emerge if the metal is in contact with the glass.

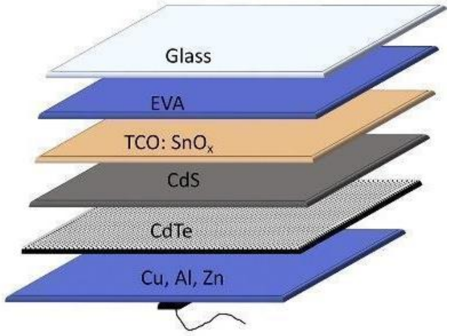


Figure 4: Typical structure of CdTe PV panels (Maani et al., 2018)

The EVA accounts for 4% of CdTe panel mass and is located right below the glass substrate.

3% of the total panel mass takes up front and back contacts that are responsible for reducing series resistance for current flowing from the solar cells.

2.1.3 CIS/CIGS

Copper Indium Selenide ($CuInSe_2$ or CIS) is a ternary compound p-type absorber material (Ong et al., 2018). Kazmerski et al. (1976) described the beginnings of CIS technology in 1976. The CIS thin-film solar cell had a buffer layer of cadmium sulfide. The efficiency of 4-5% was achieved by evaporating $CuInSe_2$ powder in the presence of excess Se vapour.

Ong et al. (2018) documented that higher efficiency rates were achieved by emerging technologies "such as alloying CIS with gallium (Ga) to become Copper Indium Gallium Selenide (CIGS), incorporating sodium (Na) into the CIGS absorber layer, and replacing thick cadmium sulfide (CdS) buffer layer with thin CdS layer".

In the CIGS panels, light enters the cell through the antireflection coating consisting of MgF_2 (Ong et al., 2018). Then it moves further through the transparent conducting oxide consisting of ZnO that is doped by Al and passes through the buffer layer of CdS. The light is being absorbed by the CIGS and reaches the back contact, usually made of molybdenum (Mo), which is deposited on the substrate. The substrate is made of glass, metal foil or polymers.

The back contact acts as an optical reflector to reflect the light to the absorber layer in CIGS solar cell. Ong et al. (2018) explain the process:

"The layer after Mo back contact is Copper Indium Gallium (CIG) before going through the process of selenization. During the selenization process, selenium (Se) vapour will react with CIG to become CIGS and react with Mo to form the $MoSe_2$ layer. This interfacial layer between Mo and CIGS is beneficial in terms of having a wider bandgap (1.35–1.41 eV) than CIGS, hence it can absorb more near-infrared light to improve the cell performance [24]."

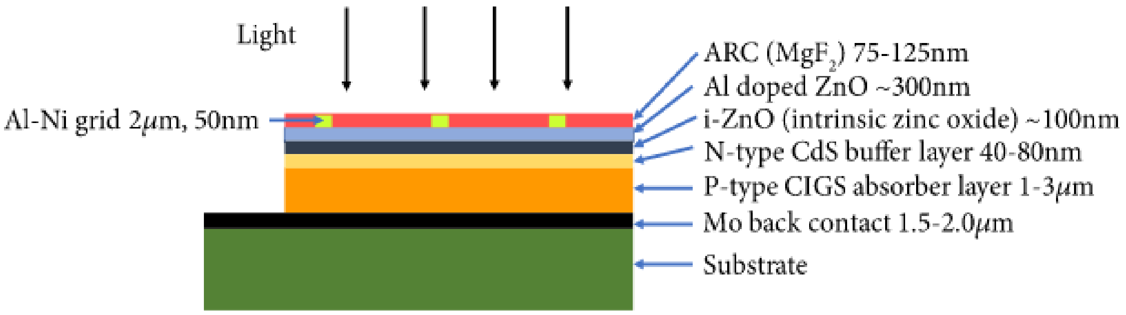


Figure 5: Structure of CIGS thin-film solar cell (Ong et al., 2018)

2.2 Critical raw materials in c-Si and thin-film photovoltaic modules

Security of supply of raw materials used for PV fabrication is very important in the European Union (EU) because many semiconducting materials or rare earth metals need to be importet into the EU. Especially, so-called critical raw materials (CRMs) are irreplaceable in solar panels (European Commission, 2018). Silicon is crucial in crystalline or amorphous solar cells as semiconductor (European Commission, 2020). Germanium is also used as a semiconductor for multi-junction solar cells. Boron serves as a p-type dopant in the crystal lattice of the silicon-based wafers. Gallium and Indium are used in CIGS technology or as the dopant in semiconductors and as indium-tin oxide (ITO) conductive layer, respectively. All of these materials have a high supply risk and are defined as CRMs for the EU. Projections of Ga, In and Si demand for PV industry at the level of EU are depicted in Figure 6 on a logarithmic scale. In the high scenario, the demand for all these CRMs is expected to rise by a factor of 6 until 2030 while in the low scenario the demand is expected to fall compared to values in 2015.

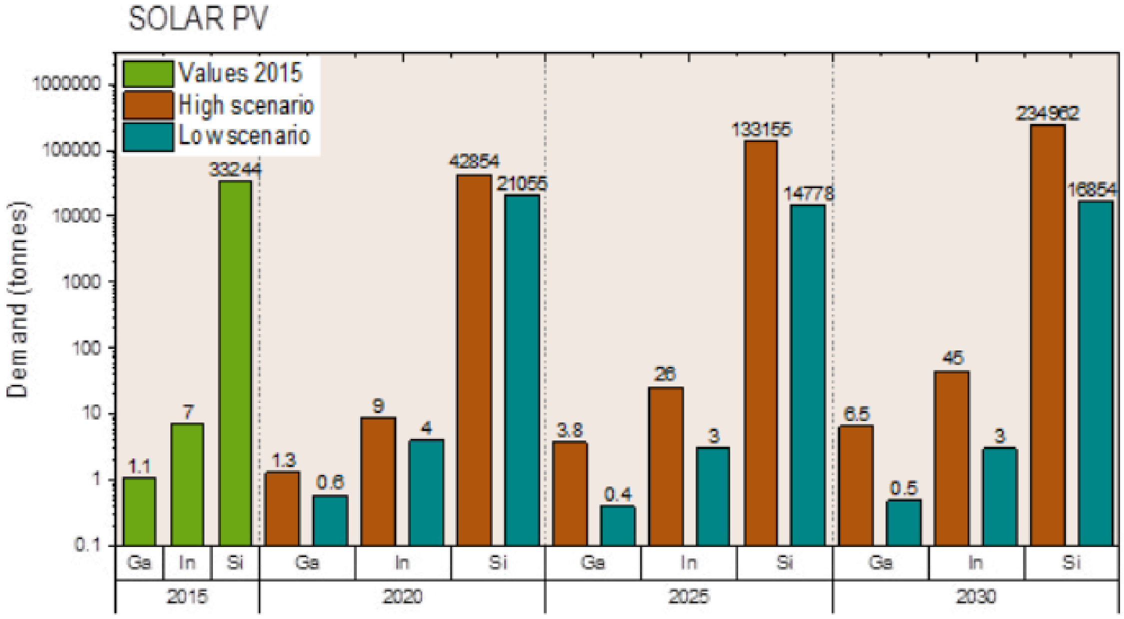


Figure 6: Projected evolution in EU demand on a logarithmic scale for the three CRMs required in PV sector (European Commission, 2018)

Project CABRISS aimed at implementing recycling technologies to recover In and Si (Draoua, 2017). The first solar cell results from the CABRISS project confirm the feasibility of using recycled Si in industrial production lines. The silicon kerf is recycled in diamond wire cutting processes (Halvorsen, 2017). The recycled material is a powder with “2-4 N” (which means 99,00% to 99,99%) in purity. Oxidation levels are acceptably low (1-4%). However, several safety issues related to Si kerf recycling are still to be addressed.
Indium is mostly recycled only in the production processes because its end-of-life recycling is very costly (Hoffmann et al., 2017). Project CABRISS successfully demonstrated the feasibility of indium recycling from thin-film panel waste and manufacturing of indium sputtering targets and ITO targets from secondary indium. These targets have been successfully tested in thin-film PV manufacturing.

2.3 End-of-life-PV modules and waste volumes

PV technology has experienced rapid successive technological improvements, which resulted in a significant price drop of PV panels (Peeters et al., 2017). As a result, PV technology is one of the most economically and environmentally promising electricity generation technologies worldwide. The decarbonization of energy systems is a high priority nowadays, therefore it is anticipated that PV deployment will continue to expand globally. The expansion of the PV market means an increase in waste PV modules in the future (International Energy Agency, 2018).

Peeters et al. (2017) state that forecasting the volumes and material composition of PV waste is a relatively new area of research with a high degree of uncertainty. Part of the uncertainty comes from the variability among manufacturers and the rapid technology change (Mahmoudi et al., 2019).

PV modules have an average estimated lifespan of 30 years. A small share of panels experiences damage during the transport, installation or operation phase due to technical or physical failures caused by external environmental influences. This share of panels appears in the waste stream earlier than after the estimated 30 years.

The latest assessment of the global PV waste streams estimates the cumulative PV waste volume to have reached 43,500-250,000 metric tons by the end of 2016. This corresponds to 0.1%-0.6% of the cumulative mass of all installed panels globally. These projections were based on PV system deployment trajectories of the International Renewable Energy Agency (2030) and International Energy Agency (2030-2050), converted through a Weibull distribution incorporating data on early failure modes of PV modules (International Renewable Energy Agency and International Energy Agency 2016).

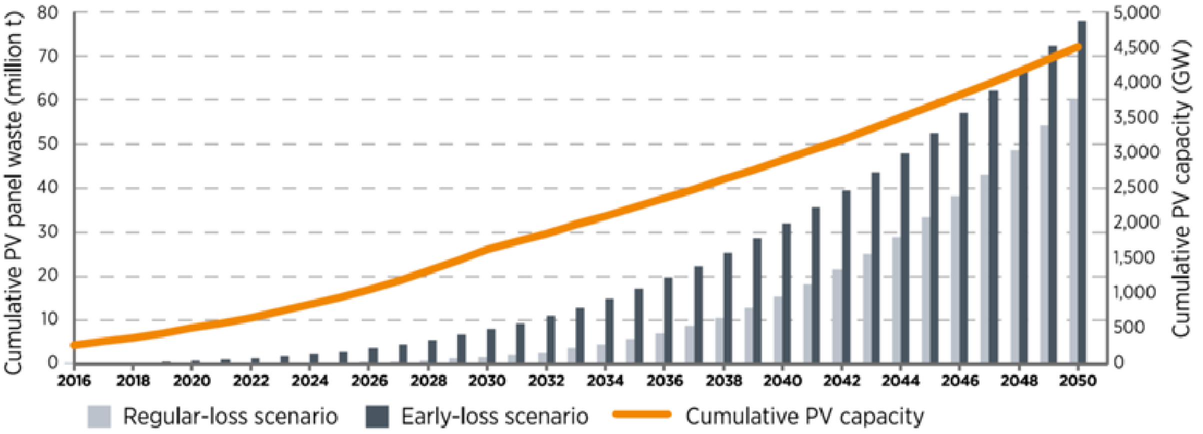


Figure 7: Estimated cumulative global waste volumes of end-of-life PV modules (International Renewable Energy Agency and International Energy Agency, 2016)

The difference in estimates results from two scenarios. The early-loss scenario projection estimates much higher total PV waste streams as it assumes a higher percentage of early PV module failures than the regular-loss scenario. The report of the International Renewable Energy Agency and International Energy Agency (2016) expects the actual future PV panel waste volumes to fall between the regular-loss and early-loss scenarios. Large amounts of annual waste are anticipated by the early 2030s, starting from 0.4 - 1.3 million tons.

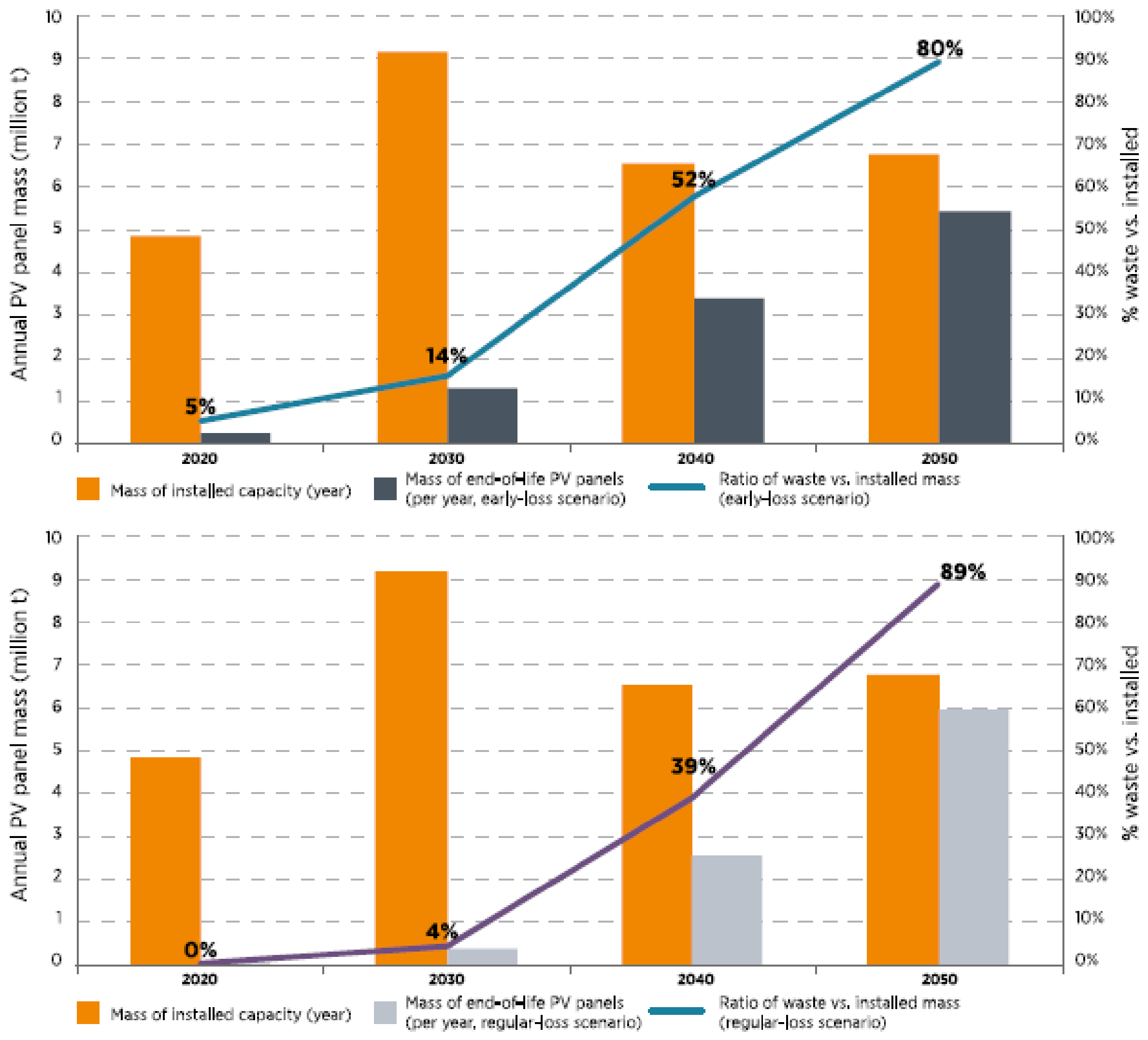


Figure 8: Annually installed and end-of-life PV panels 2020-2050 (in % waste vs. t installed) by early-loss scenario (top) and regular-loss scenario (bottom) (International Renewable Energy Agency and International Energy Agency, 2016)

3. Applied methods

The separation technologies of compound materials and the design-for-recycling concepts of PV panels have been studied by the means of a literature review and semi-structured interviews by experts from the field. Design-for-recycling concepts have been either derived from the existing unconventional c-Si PV technologies or found in the literature as existing design-for-recycling concepts.

For the literature review, a combination of the following keywords was used: “end of life”, “recycl*”, “waste”, “solar”, “photovoltaic”, “PV”, “c-Si”, “thin-film” and the snowball method was applied to find further relevant literature. The terms “design for recycling” AND “solar” OR “photovoltaic” were searched for separately. The scholarly databases used were Scopus, ScienceDirect, BOKU:LITsearch as well as the physical library of the University of Natural Resources and Life Sciences, Vienna was visited. The most important search criteria were set to be the year of the research combined with the reliability of the source. The literature corresponding to these criteria in English and German languages was reviewed.

Merely the PV panel has been studied. The junction box and cables, as well as the balance of system and energy storage components are beyond the scope of this master thesis. The focus of this work lies on the pre-treatment methods of PV panels, namely delamination and material separation technologies.

Information on the costs for the technologies reviewed has not been collected since the majority of technologies is under R&D or the information on costs is not publically available.

4. End-of-life management

4.1 End-of-life regulatory framework

Sharma et al. (2019) have reviewed the global policies for the waste PV panels and found that very few countries outside the European Union have taken any measures to regulate waste PV panels. These countries have started to develop some regulations or have them in the planning stage. Apart from European Union, this waste category falls under the general waste legislation. Consequently, the PV panels are improperly landfilled, especially in developing countries. This contributes to environmental pollution.

In the EU member states, PV panels are adressed and considered by the Directive 2012/19/EU on waste electrical and electronic equipment (WEEE directive, 2012) since 2014. This directive regulates the appropriate collection, treatment and financing of end-of-life products and requires that manufacturers, rebranders and importers of electronic products (in the following regarded as manufacturers) comply with all national waste management obligations and reach the minimum collection and recovery targets set (WEEE directive, 2012). Under this law, manufacturers putting the PV panels on the market of the European Union are responsible for take-back, treatment, financing and declaration of their products and may fulfil their obligations individually or by joining a collective scheme. This means that PV panels are now under the scope of the extended producer responsibility. Not complying with these legal obligations can result in fines or prohibition of sales.

The RoHS Directive currently restricts the use of Pb and Cd among other hazardous substances in electrical and electronic equipment. However, PV panels intended to be used for energy production are exempt from this directive.

The WEEE directive currently demands an 85% collection rate and an 80% recycling rate of the overall PV panel weight. The limit values of the glass output fraction for all PV panel types are regulated for Cd, Se and Pb, and they differ depending on the panel type. The limit values for Cd and Se represent the removal and separation of 95% of the semiconductor layer for thin-film modules.

The WEEE directive allows for the reuse of the panels (Bellini, 2020). This presents an opportunity for legal limbo because the guidelines for defining a PV panel as waste or as capable of reuse are lacking. This enables the shifting of electronic waste to developing countries where the products are eventually discarded in an environmentally improper manner.

4.2 Recycling technologies

In this study current state-of-the-art technologies for the recycling of PV modules were investigated. The focus was hereby on separation techniques for delamination of the composite materials. In the literature (cf. e.g. International Energy Agency, 2018 and Xu et al., 2018), these technologies are mostly referred to as recycling technologies, therefore, in the following, the separation technologies will often be referred to as recycling technologies as well. Both established separation and recycling technologies and promising ones in the research or pilot phase will be considered. The list of the technologies explored in this master thesis is not intended to be exhaustive. The end-of-life management of disposed PV panels generallay consists of several process steps depicted in Figure 9.

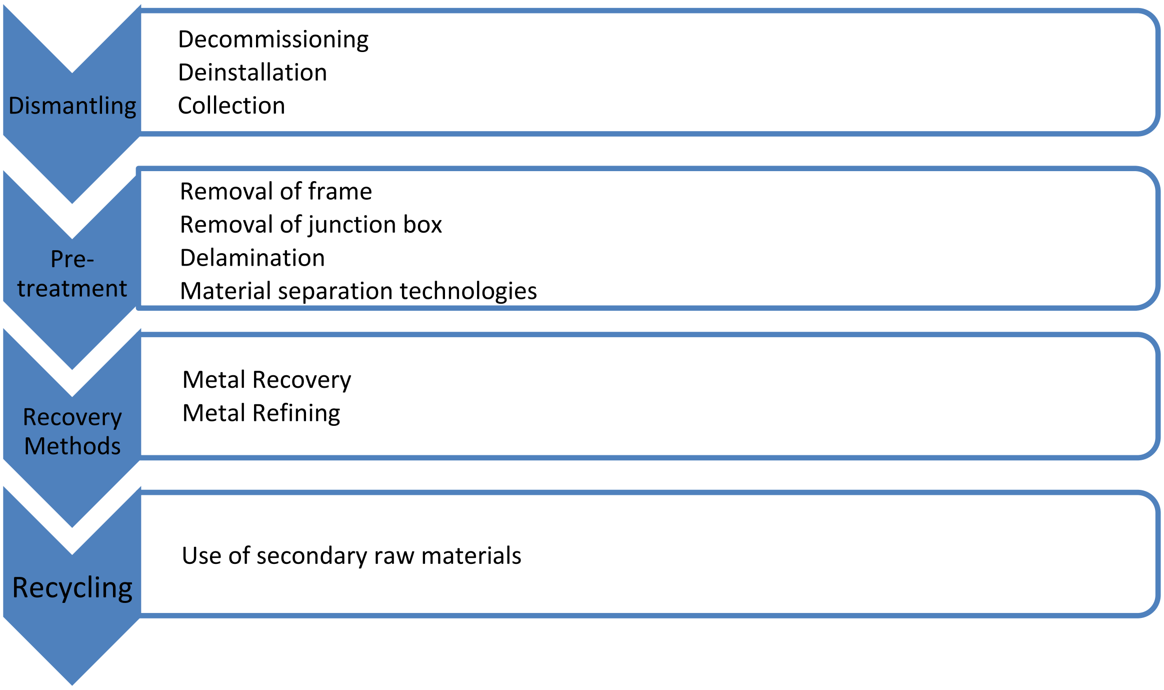


Figure 9: End-of-life process steps for end-of-life PV panels based on Tao and Yu (2015) and Marwede et al. (2013)

International Energy Agency (2018) summarizes the emerging PV recycling industry. Typically, end-of-life PV panels are treated in separate batch runs in existing recycling plants for the main components of a PV module, namely glass or metal recycling plants. In the long term, dedicated PV module recycling plants have the potential to increase treatment capacities and maximise revenues due to the better quality of recyclables and the ability to recover a greater fraction of materials.

In some cases, the end-of-life PV panels end up with WEEE recyclers as explained by Ledersteger (2020). However, these quantities are so small that the panels are shredded together with other electronic devices. In some cases the frame and junction box are not even removed before the shredding. The resulting fraction is the aluminium and sometimes also glass which can only be downcycled because of the impurities. In cases where the glass cullet has an attached polymer layer with it, it is treated as commercial waste or is incinerated, depending on the composition.

A comprehensive patent filing study done by International Energy Agency (2018) gives a good overview of what is currently the status and trend in the PV recycling industry.

Total 178 PV recycling patents were filed, out of which 128 pertained to c-Si module technology and 44 to compound module type. Most probably the dominance of the c-Si recycling technologies reflects the installation market trend because at present c-Si modules occupy most of the installation market.

As the c-Si-based and thin-film panels have differences in the panel structure and the contained metals, their recycling technologies have different aspects as well. The aim of eliminating the encapsulant from the laminated compound PV structure is to recover both the cover glass and the substrate glass which has the semiconductor layer deposited on it. The aim of eliminating the encapsulant from the c-Si-based panels is to separate and recover glass, Si cells and other metals.

Regarding the targeted materials for recovery of the c-Si panels, most patents are for technologies recovering aluminium, solar cells and glass. An extreme increase in patent filings occurred in China since 2011 (almost half of the c-Si recycling patents) and in some other Asian countries. Various components of the module can be targeted as a way to separate the module. 45% of patents focus on separating modules by removing encapsulants. Mechanical methods account for 40% of the total patents. Many patents have been filed for the recovery of aluminium frames, glass and solar cells but only a few concentrated on the recovery of Si, Ag and Cu.

In contrast to that, most patents for thin-film panels address the total recycling process without concentrating on a specific component or material. The reason is the thin-film panel structure. They are manufactured by a continuous deposition process rather than an assembly of various components as is the case with the c-Si panels. Therefore, a single method with a focus on disassembly is not effective for thin-film technology. A good example is glass. It cannot be recovered by a single method because semiconductor material remains on the substrate after the module separation. An additional step is needed for the separation of the semiconductor material from the substrate glass. This is the reason why a combination of separation methods is used for thin-film modules. The approach of combined methods of more than two single methods accounts for 64% of the total patents and is followed by single methods: thermal, optical, mechanical (sometimes referred to as physical methods in the literature) and chemical.

Interestingly, 95% of the patent assignees for compound PV recycling are corporations while for c-Si-based panels both corporations and public entities have filed the patents. This difference leads to the assumption that the patent technologies for compound modules are more likely to be commercialized. The greatest interest for compound PV module recycling is in the United States with 27% of the total patent number, followed by Japan, China and European countries.

Tao and Yu (2015) give a structure to the end-of-life treatment of PV modules, which is also illustrated in Figure 10. Since the PV panels mainly consist of laminated layers, the major processes can be divided into three stages:

1. Delamination of the substrate-semiconductor composites
2. Material separation of non-/ferrometals, polymers, glass and other components
3. Metal extraction or refining of the valuable metals and metaloids

Before the panel is delaminated, frame and junction box are removed. This could be done manually but would not be economic on a large scale. Noda et al. (2014) name an automated frame removal method by an air cylinder actuator.

First, processes for discomposing different layers and exposing semiconductor layers are necessary. Then, processes for separating different materials are utilized. And finally, processes for extracting useful or harmful materials are required. Different technologies can be applied for each of the stages as visible in Figure 10.

At the very first, the solar panels need to be dismantled from their installation site. Then, they need to be collected and brought to the treatment site. The pre-treatment is typically the same for both c-Si and compound technologies, namely the mechanical removal of the aluminium frame and junction box. The aluminium can be recovered through secondary metallurgy. Other elements present in small quantities (iron, silicon, and nickel) are typical components of aluminium alloys (Dias and Veit, 2018).

The further treatment depends on the panel type and the technologies chosen. The separation technologies are divided into mechanical, chemical, thermal, optical methods and combinations of thereof (International Energy Agency, 2018). After the selected separation technologies, the post-treatment in the form of recovering methods takes place that are mainly divided into hydrometallurgical processes to recover metals and pyrolytic processes.

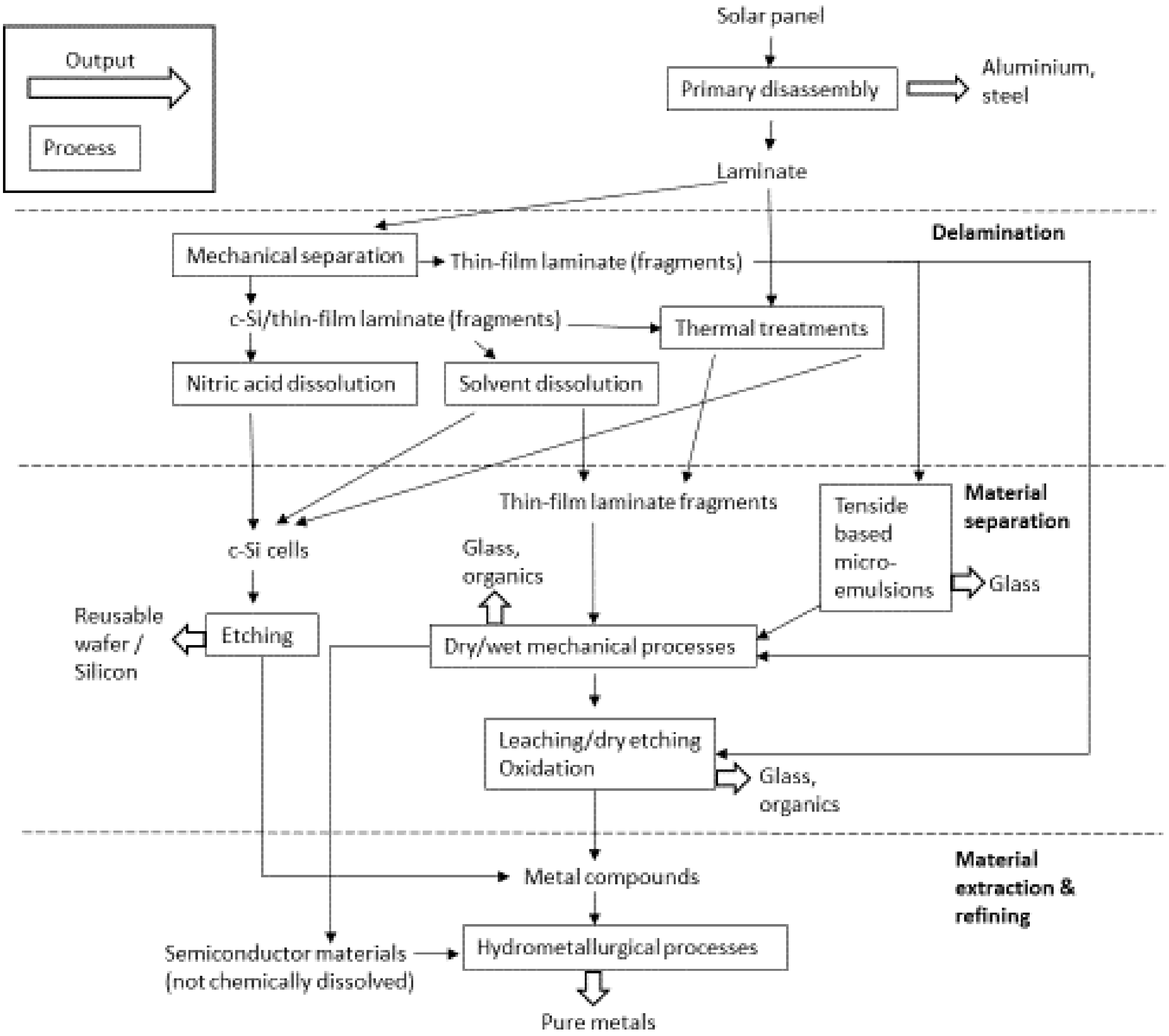


Figure 10: Feasible pathways of end-of-life PV module recycling modified according to Tao and Yu (2015) and Marwede et al. (2013)

4.2.1 Delamination

Eliminating the encapsulant from the laminated structure (also referred to as delamination) is the most important aim for recycling technology R&D (International Energy Agency, 2018). It is also the most difficult and at the same time a crucial process for separating the different materials from each other. The reason is that the Si cells are coalesced with the encapsulation material through the thermal treatment. Panels are laminated to withstand external influences and mechanical loads for 30 years on average. For prolonging the operational lifetime of the panels, adhesives are being developed to be used instead of encapsulation foils impeding the delamination process even further (Marwede et al., 2013). In the following, different delamination technologies for c-Si and chalcogenide-based solar panels are described.

4.2.1.1 Two-Step thermal delamination

International Energy Agency (2018) claims that the thermal approach is expected to have an advantage of recovering glass and Si cells without damage.

Wang et al. (2012) from the Green Energy and Environment Research Laboratories, Industrial Technology and Research Institute proposed a thermal approach that results in obtaining a whole glass plate without breakage. It can be reused for new solar modules if the temperature is well controlled in the process. Almost 62% of silicon is recovered with a purity of “8 N” (i.e. % of purity) that can be reused for new solar panels. 85% of copper is recovered in further acid treatment. The recovery of glass results in an almost 100% recovery rate as reported in experiments.

The thermal delamination occurs in two-step heating. The encapsulation material (in the respective experiment EVA was used) could be fully oxidised to CO_2 and H_2O. Nevertheless, Tao and Yu (2015) argue that this process requires high energy consumption and that the thin c-Si cells degrade due to high temperature.

4.2.1.2 Electro-thermal heating

The electro-thermal heating process was developed at Padua University as described by Doni and Dughiero (2012). By this approach, the glass is easily removed from the c-Si solar panels. This method uses the thermal effect of an alternating magnetic field on a dielectric material work. A radio frequency electro-thermal heating device heats the core of a c-Si panel. Afterwards, the glass fragments can be easily removed from the laminate. This method works at temperatures lower than the decomposition temperature of EVA and the substrate, which reduces the environmental impact of the recycling of PV panels to a large extent.

4.2.1.3 Combinations of thermal and mechanical methods

The bankrupt Solarworld GmbH from Germany had commercialized a thermal treatment method for primarily its production waste, but it was also applicable for end-of-life c-Si panels (Tao and Yu, 2015). In the beginning, the solar panels were combusted at 600 °C in a complex semiconductor-protecting process where the polymer layers were oxidized and thus eliminated. Then, manual separation of the remaining materials (solar cells, glass, metals) was carried out. Glass and metals were recycled. Afterwards, etching was applied to solar cells to obtain wafers again. After the solar cell processing, the recycled wafers are of equal value to new wafers and can be reused in a standard solar cell production line.

The material recovery rate of this method is 84% of the input module weight with up to 6 N purity of the fraction. More than 90% of the glass can be reused in new products and 95% of the semiconductor material can be reused for new solar modules. Silver, copper and other metals can be added to their primary production. The polymer layers can be used as a substitute fuel. The solar cells are recovered up to 98% depending on the initial status of them, the solar panel type and the solar cells used. If the cells have damages, such as micro-cracks or edge chipping, they can typically not be recycled into an intact wafer, however, they could be used for obtaining silicon raw material. The thicker the wafers, the higher the recovery yield. One disadvantage of this method is the inefficiency of the semiconductor-protecting thermal delamination process for modules with thin wafers.

Another combination method suitable for c-Si, compound and CIGS PV modules was developed by Kitakyushu Foundation for the Advancement of Industry, Science and Technology from Japan together with others (Noda et al., 2014). After frame removal, the backsheet is removed by a milling machine for preventing the glass from thermally cracking in the next steps. The rest of the panel is heated, and the EVA is decomposed in a muffle furnace. The decomposition gas is sucked out and burned. The generated heat can be recycled to the furnace. Glass substrates that have CIGS layers attached and the cover glass are recovered from the CIGS modules. Solar Frontier was part of the project and developed wire brushes that scrape the CIGS layer on the substrate glass. CIGS metals are recovered through a cyclone collector and afterwards can be recycled by metal refinery companies.

Granata et al. (2014) investigated a combination method suitable for c-Si, amorphous Si and CdTe PV panels. First, the frame is removed. Subsequently, two approaches were applied as a shredding step. One is a simple crushing method using a two-bladed rotor followed by thermal treatment; the other one uses the two-bladed rotor followed by hammer milling. An analysis for the distribution of sizes of the crushed module pieces was performed, X-ray diffraction and X-ray fluorescence analyses of obtained fractions were carried out to evaluate their properties. For all types of modules, the mechanical approach followed by the other mechanical approach performed better.

Kushiya et al. (2003) have developed a simple and low-cost recycling method for CIGS panels. First, the modules are heated to below 250°C for softening the EVA. Then, the cover glass is mechanically separated. The residual EVA is removed by immersion in an acetic acid solution.

4.2.1.4 Thermal and mechanical treatment

During the European project *Full Recovery End of Life Photovoltaic* one recycling technology by Sasil, S.p.A. and other organizations was developed (Ercole, 2016). After the removal of the frame and junction box, the laminated structure is heated to 90°-120°C by an infrared heater. Then the laminate is inserted into a roller mill and vibrating knife equipment, that separates and recovers the glass. The remains are heated to 500°C, combusted at 850°C, and metals in the Si cells and electrodes are separated. During the burning of polymers, waste gas is generated. It can be recovered for combustion. The separated metals are further treated by chemical methods.

4.2.1.5 Mechanical scraping with solvent-based treatment

Mitsubishi Materials Corporation (2016) from Japan has developed a scraping method for recovering glass from the PV modules. The cover glass is mechanically scraped to avoid contamination of the glass with the encapsulation layer. The treatment time is one minute per module. Then, the recovered glass is sieved and can be reused. The remaining solar panel layer contains a small amount of glass that can be treated by metal refinery technology. The main target of the metal refining process is Ag because of its high current value.

Another scraping process is proposed by the Japanese Toho Kasei Co. Ltd. (2016). Their approach is to treat the non-glass layers mechanically. First, the backsheet and the encapsulation layer, including the Si cells and electrodes, are scraped. Second, the scraped encapsulation layer is treated by a solvent developed by Toho Kasei Co. Ltd. The Si, other metals and polymers are then recovered. The recovery of high-purity Si is one of the goals of the project. The encapsulation polymer is recycled as fuel or material. The scraped backsheet is disposed of as industrial waste. The cover glass is not damaged in the scraping process, however, a small amount of the encapsulation layer remains attached. These remains can be removed by another solvent that is developed by Toho Kasei Co. Ltd. These solvents could also be used before scraping and the layers would be resolved, however, this would be a very time-consuming process.

4.2.1.6 Refrigeration and grinding

An interesting mechanical method has been developed by Yingli Solar, Electrical Engineering Institute and the Chinese Academy of Sciences in China (International

Energy Agency, 2018). PV module has its frame and junction box removed and is consequently crushed. The crushed pieces are then refrigerated at -197°C by using liquid nitrogen. The refrigerated pieces are ground, and the particles of the encapsulation layer, glass and mixed powder containing Si, Ag, Cu and other materials undergo a physical separation. A mixed powder of Si, Ag, Al etc. is separated from the pieces of backsheet and encapsulant. The expected recycling rate is approximately 90% but the Si cannot be recycled for new PV modules because of its low purity.

4.2.1.7 Commercially established combination of mechanical processes

According to PV Cycle (2018), the organisation is globally present with its PV panel end-of-life service and has collected more than 27.000 tons cumulative of end-of-life PV panels. 95.2% of treated panels in 2018 are silicon-based.

International Energy Agency (2018) states that most of these modules are treated by glass recyclers. Flat glass recycling companies can implement recycling of the laminated glass component of c-Si modules with little additional investment to their daily business. An example of the possible process steps is seen in Figure 11. The glass recovery is frequently run in batches to be able to adjust processing parameters. For removing impurities such as polymer residues or screws from the glass cullet typically magnets, crushers, sieves, eddy-current devices, optical sorters, inductive sorters and exhaust systems are used. The resulting crushed glass fraction may still be heavily contaminated with silicon, polymers and metals. It can be blended with other recycled glass and used as a thermal insulating material in the glass-foam or glass fibre industries. With an increase in end-of-life PV streams, this market may become saturated though and technologies leading to higher quality recyclables will be in demand.

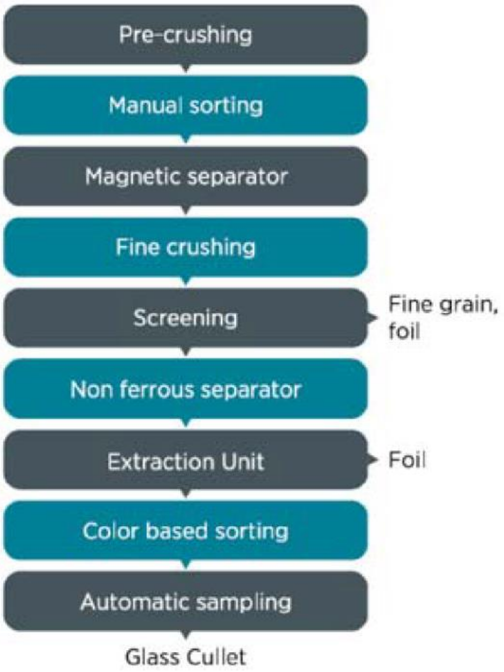


Figure 11: Process for laminated glass recycling (International Renewable Energy Agency and International Energy Agency, 2016)

Currently, the mechanical approach dominates for separating the laminated structures of c-Si PV modules. It results in a high material recovery rate measured in mass. Some high-value materials in small quantities may not be fully recovered though. Also lead and lead compounds are still found in the output glass fraction. In some countries, in addition to glass, certain valuable metals such as silver are recovered by intermediate processors for technical and/or economic reasons.

In 2018 the first dedicated solar panel recycling factory in Europe, possibly even in the world, was opened in France by the French water and waste group Veolia (De Clercq, 2018). Veolia has a contract with PV Cycle France. In the factory, robots are used to dismantle simultaneously the frame, junction box and cables (Veolia Group, 2018). Then they are shredded and treated according to the procedure described above. The treatment time is between one and one and a half minutes per panel.

Even in this dedicated factory exclusively for solar panels only the aluminium frame and glass are being recycled, the rest is incinerated and accounts for 5.3% of the total mass. Rollet and Beetz (2020) investigated this non-recycled fraction and found that it is “mainly dust trapped in the filters after shredding (..) The dust can also be incinerated or used as a substitute for sand in construction, since glass, silicon and silicone are all derived from sand”. The backsheet is incinerated as well. The EVA or Tedlar used in backsheets could be used as a binder for paint, however, it would require further cleaning, making it more environmentally friendly to incinerate them in a filtered incinerator.

4.2.1.8 Solvent-based methods

According to calculations of LuxChemtech, the currently commercially used recycling method described in chapter 4.2.1.7 results in a loss of two-thirds of the silicon-based PV module value (Deutsche Rohstoffagentur in der Bundesanstalt für Geowissenschaften und Rohstoffe, 2019). LuxChemtech (earlier Loser Chemie GmbH) has developed a method that separates components in pure fractions of glass, encapsulation foil, fluor plastic foil, silicon and silver. Glass, silicon and Ag can be recycled in the PV panel manufacturing process. According to Palitzsch (2021), the LuxChemtech technology uses aluminium chloride and water to separate the PV module components. Currently, a recovery rate of about 95% is reached. Tests with full-sized panels were accomplished. A further process is needed for the separation of the Si cell components.

Yokohoma Oils & Fats Industry developed processes for eliminating the encapsulant from the laminated structures (International Energy Agency, 2018). After the removal of the frame and junction box, the backsheet is removed mechanically. The remaining laminate is immersed in a neutral solvent, and glass, encapsulation layer, Si cells and the electrodes are separated. Usually, the separated glass does not get damaged. The remaining layer is crushed and immersed in an alkali solvent. Afterwards the encapsulant, silicon and electrode ribbon are recovered. An additional process is required to recover metals from Si cells. The processing time is expected to be almost a day for a commercial module. However, compared to acids and organics, the developed solvents are environmentally friendly.

Since the chemical separation requires a long treatment time, the Korea Research Institute of Chemical Technology together with Kangwon National University introduced additional ultrasonic irradiation to overcome this shortcoming (International Energy Agency, 2018). The panels are submerged in an organic solvent, and a temperature of 70°C needs to be attained at an irradiation power of 900 W for 30 minutes to completely dissolve EVA. Different organic solvents can be used - trichloroethylene, O-dichlorobenzene, benzene, and toluene (Kim and Lee, 2012).

Different solvent combinations, temperatures, ultrasonic power and radiation times were tested. According to their experiment results, the PV cells were recovered without any cracks (Tao and Yu, 2015). The disadvantages of this approach are the relatively expensive equipment and waste solution treatment required. Lead resulted as a hazardous by-product. After the elimination of the encapsulant from the laminated structure, a further process is required to recover metals from Si cells.

4.2.1.9 Irradiation by laser

Another researched method for the destruction of EVA is irradiation by a laser through the glass (Tao and Yu, 2015). The test was conducted by 5 N Plus, a Canadian metal refiner, however, it is far from industrial application. The process is slow, and the required equipment is very expensive.

4.2.1.10 Pyrolysis

Disposer company Suez uses pyrolysis for the treatment of end-of-life PV panels together with Institute Fraunhofer (EU-Recycling, 2018). More than 90% of the materials can be recovered by this method. The panels are heated in a furnace under high pressure and exclusion of oxygen. This induces pyrolysis by which the polymers are decomposed in methane, propane and butane. The delamination frees metals, glass and silicon for further electrochemical treatment. A pilot facility in Knittlingen (Germany) in 2019 was successful. Nevertheless, the energy demand for this method is very high due to the heating of the whole panel (Heuschkel, 2021).

4.2.1.11 High-intensity light

Heuschkel (2021) explains that Flaxres GmbH from Germany has developed a chemical-free, low-energy, mobile and fast method to separate all components of composite materials like PV modules. Other electrical appliances can be broken down by this technology as well. This innovative thermal approach uses a well-established process of a high-intensity and low-energy light source on an unprecedented scale to illuminate the whole PV module surface at once with visible light. Plasma jet created by flashbulbs heats selected layers of the solar panel depending on the parameters set. In this way, less energy is required than if the whole panel is heated up as in the case of pyrolysis. The heated layer expands, and this leads to the compartmentalization of the respective layer from the rest of the solar panel. It results in homogeneous recyclables with no impurities that can be used for manufacturing new solar panels, except for encapsulation foils and backsheets. The flashbulbs are suitable for all types of solar modules, including organic PV and thin-film silicon modules. If the panels are damaged in a way that they are bent, they can still be treated, however, if the panels are folded (as may be the case after hurricanes), then treatment by flashbulbs is not possible.

Flaxres GmbH was founded in 2017. Currently a funded EU research project of over 12 Mio. € is being implemented together with Veolia and Evonik Industries. Under this program, EIT RawMaterials has linked Flaxres with strategic industry partners to validate the feasibility of high-quality recycling after the dismantling process. The technology has been approved in tests with real-sized PV modules.

The recycling process begins with the removal of the junction box by mechanical means. Silicon-based solar panels are recycled by using fewer illumination steps than thin-film panels. Polymers lose their adhesion to the heated silicon during the first light exposure. By changing the illumination period and light intensity, the bus bars can be unsoldered during the first illumination step as well. The contact fingers remain attached. Consequently, this first step has separated the module into five parts: front side glass with the attached polymer layer, silicon wafers with the attached contact fingers, bus bars, backsheet or back side glass together with the adjacent polymer layer and frame.

During the second light exposure, the polymer layer attached to the glass can be removed using different parameter settings. In this way, panels with shattered front glasses can be also treated because during the first illumination step the glass pieces are held together by the adjacent polymer layer. Various wafer thicknesses and polymer types can be treated by changing the exposure parameters.

In the first light exposure step for the thin-film panels, the light-absorbing layer is heated up. A polymer layer is commonly laminated on one side of the light-absorbing film, and it loses its adhesion irreversibly during this process. Edge sealing material, such as silicone or butyl rubber, can be separated along with the polymer layer. During the second illumination step using a different parameter set the light-absorbing layer is stripped from the bottom electrode layer. For removing the bottom electrode layer including the bus bars a third light exposure is performed. The light-absorbing layer and the bottom electrode layer generate particle flakes that can be collected separately. This is induced by excessive thermo-mechanical stress. Evaporation or combustion of the films does not take place. Whether the separation of the two different kinds of flakes from each other is cost-effective remains open. It could be that reducing the number of illuminations by one and stripping both thin films at once can be more cost-effective.

In the final exposure step, the polymer layer is delaminated from the other glass sheet. The several light exposures result in the following fractions: two clear glass sheets, bus bars, flakes from the light-absorbing layer and the electrode layer, one polymer layer and edge sealing material. In the case of CdTe modules, it is possible to set the parameters in such a manner that the complete removal of toxic materials like Cd is assured.

During the heating process, a negligible amount of gas is released required for the separation of the back side glass or back side polymer like polyvinyl fluoride as well as the front side glass. A deaerator is installed to protect the workers at the facility and the environment, however, the exact amounts of the released gases are not yet measured.

The target for the speed of this recycling method, including the separation of the resulting recyclables, is 10 seconds per one PV module. The flashbulb equipment fits into a standard sea container, therefore it can be directly transported to PV installation sites, PV panel production factories or end-of-life panel collection sites to minimize transportation routes, especially for the heavy fraction glass.

Even though the conversion of electricity into light by flashbulbs and subsequently into heat in the light-absorbing film creates a significant loss of energy, the electricity required is very low. This dry process is estimated to be less expensive than other currently available recycling technologies. The profit using this recycling technology is estimated to be around 600-900 €/t.

4.2.1.12 Heated cutter

A mechanical cutting of the laminate was introduced by Yu and Yang (2013) to reduce the cracks of the silicon wafers during the recycling process. A hotwire is used by this approach. After the laminate has been cut, the cover glass and the substrate can be removed. This gives more space for the swelling of EVA during the following thermal or chemical treatments, which should reduce wafer cracks.

Hamada Corporation and NPC Incorporated (2016) have also worked on the method with a heated cutter. First, the frame is removed and the backsheet is scraped. Then, a heated cutter incises the laminate and cuts it alongside the cover glass. The cutter is inserted into the bonding plane between the glass and the encapsulation layer. When the encapsulation material is removed from the glass plate, the glass is recycled as a cullet. The remaining layers can be treated by chemical processes or a metal refinery to recover any metals.

The thin-film panel manufacturer Solar Frontier K. K. (2016) has researched the heated cutter approach also for the compound panels. First, the frame and junction box are removed. Then, the cutter is inserted between the cover glass and the substrate glass. The speed of the treatment is 400 seconds per module. The cover glass has some EVA attached. The substrate glass is typically broken and, in the case of CIGS modules, has CIGS, Mo and some polymer layers attached. Both types of glass are treated chemically afterwards. Glass and metals are recovered.

4.2.1.13 Commercially established shredding

According to the company itself (2016), First Solar is the largest thin-film solar module manufacturer in the world. The company describes its recycling method for CdTe solar panels. It is implemented at all their production factories on a commercial scale and is used for both, end-of-life solar panels and manufacturing scrap. The process requires no pretreatment.

First Solar provides packing materials and collects end-of-life modules upon customer request. Once they reach the recycling plant, the modules are collected in hoppers and loaded by forklift into a shredder. The facility is equipped with an aspiration system. It is used for dust control in all dry recycling process parts and is equipped with pre-filters and high-efficiency particulate air filters which are 99.95% efficient. Collected dust and filters are disposed of in an environmentally safe manner.

Inside of the shredder, the modules are reduced in size in a two-step process. First, the modules are broken into large pieces. Second, the hammermill crushes the larger pieces into approximately 4-5 mm small pieces to ensure the lamination bond is broken.

4.2.1.14 Optical and chemical approach

LuxChemtech (earlier Loser Chemie GmbH) from Germany is piloting a technology involving an optical approach for the separation of compound solar panels (Palitzsch, 2021). First, the frame and junction box are removed. Then, panels are automatically loaded into the optical treatment equipment. The laser treatment was tested, however, due to the shortcomings of this technology, new methods with different forms of light are currently under research, including flash lamp annealing. The aim of the optical treatment is the separation of the cover glass from the substrate glass. Vacuum suction cups were formerly used to pull the components away from each other without damaging the glass plates. Currently, the suction cups are not used anymore.

4.2.1.15 Laser

Strachala et al. (2017) describe another possibility of recycling PV cells after the separation of them from the PV module. Laser surface cleaning was performed with a neodymium laser pulse with λ = 1064 nm wavelength and frequency up to 120 Hz. Aluminium metallisation, anti-reflective coating, and passivation layer were removed from both mono- and polycrystalline cells. Pulse duration was 10 ns. Beam energy reached 300 mJ per pulse. The created irradiation heats localised areas and removes unwanted layers.

The laser method is not economically advantageous. It is expensive and very time-consuming. The needed time is about one minute per cm^2. During this time it is possible to clean the whole surface of the cell by the chemical etching.

4.2.2 Separation of metallic from non-metallic compounds

After separating the module laminate, the thin-film materials can be removed from the substrate and the Si cells can be separated from glass and other metals. Several separation technologies of non-metallic fraction (encapsulation foil, glass) from metallic fraction (semiconductors, metals) for c-Si and chalcogenide-based solar panels are described in this chapter.

4.2.2.1 Manual material separation

One obvious way to separate the materials after delamination is a manual separation that was used by Solarworld GmbH for primarily its production waste and end-of-life c-Si panels as described in chapter 4.2.1.3. Tao and Yu (2015) concluded that the manual material separation should be automated though.

4.2.2.2 Vacuum blasting

As a material separation step, vacuum blasting can be applied to remove the non-metal materials. It is suggested for intact modules to reduce the material input for the subsequent leaching or etching process (Tao and Yu, 2015). If the material input is reduced, the required chemicals for the processes are reduced as well. The vacuum blasting removes the semiconductor layers without a chemical dissolution, however, the process is relatively slow and emissions of metallic fraction with abrasives are generated. A further chemical or mechanical treatment is needed before the refining step.

4.2.2.3 Material separation after mechanical delamination

After the mechanical delamination process described in chapter 4.2.1.3, the crushed pieces were treated in three different ways depending on their size. Those with a diameter >1 mm are combusted at 650°C to separate and oxidize the polymers, those with a diameter > 0,08 mm are recovered as glass and those with a diameter <0,08 mm are further treated by a hydrometallurgical process to recover metals.

4.2.2.4 Commercially established etching, precipitation and glass recovery

The crushed pieces of thin-film solar panels are etched with a mixture of sulphuric acid and hydrogen peroxide (International Energy Agency, 2018). This removes the semiconductor layers in a slowly rotating, stainless steel drum (First Solar, 2016). The drum is emptied into a classifier where glass is separated from liquids. A rotating screw conveys the glass up an incline, leaving the metal-rich liquids behind, which are pumped to the precipitation unit. The metal compounds are precipitated in three stages at an increasing pH.

The precipitated materials are concentrated in a thickening tank where a metal-rich filter cake is an outcome. It is packaged for processing by a third party. Up to 90% of semiconductor material (Te and Cd) can be recycled for use in new solar modules (First Solar, 2021).

The glass pieces are treated by a vibrating screen that separates the glass from the larger pieces of the attached laminate material. The glass is further rinsed to remove any residual semiconductor films. About 90% of the glass is recovered as cleaned glass that can be used for new glass products.

Altogether the separation process currently results in about 90% of the module weight being recovered, most of which is glass (First Solar, 2021).

4.2.2.5 Mechanical scribing and dissolvent

After the removal of residual EVA described in chapter 4.2.1.3, the CIGS-based absorber is collected as a metal powder by mechanical scribing. The back glass is recovered by dissolving the MO layer with a diluted nitric acid solution.

4.2.2.6 RESOLVED method

EU-LIFE project RESOLVED resulted in two recycling methods for compound modules (Berger et al., 2010). The compound panels are delaminated thermally into the substrate glass and the cover glass. The cover glass can be recycled without any further treatment. The substrate glass is then treated by blasting to remove the semiconductor layers.

The second recycling method was developed for broken panels. At first, they are treated mechanically by crushing and milling to reduce the grain size and to uncover the semiconductor layer that is removed by attrition. As explained in more detail by Maani et al. (2018):

“Attrition is a wet mechanical process using shear and frictional forces on the surface of the particles to be separated. During attrition, the thin film fragments are divided into glass, EVA and semiconductors. This product is then taken through screening/sieving whereby glass greater than 150 µm and EVA are sieved out. Glass smaller than 150 µm and semiconductor materials are passed through a floatation process.”

As Tao and Yu (2015) describe, flotation is performed for the pre-concentration of the CIS- and CdTe-fines. Flotation is a relatively simple process and uses relatively few chemicals. However, flotation results in considerable losses of valuables during rinsing and sieving and results in inadequate purities of the separated materials. Experiments have shown that a multi-step flotation improves the valuable yield and related enrichment of the semiconductors. Te and In were enriched by factor three due to flotation. The CdTe flotation products need to be purified to semiconductor-grade material for recycling within the PV industry.

4.2.2.7 Dry etching and precipitation

Antec Solar GmbH from Germany holds a patent for the recycling of compound PV modules (Antec Solar GmbH, 2021). Tao and Yu (2015) describe the patented process. It includes mechanical treatment for the physical disintegration followed by pyrolysis of EVA. Pyrolysis is performed at a temperature of at least 300°C in an oxygen-containing atmosphere. Then, the module fragments are exposed to a chlorine-containing gas atmosphere at a temperature of more than 400°C. This causes an etching process wherein $CdCl_2$ and $TeCl_4$ are generated, which are further condensed and precipitated by cooling. A schematic overview of the process is provided in Figure 12.

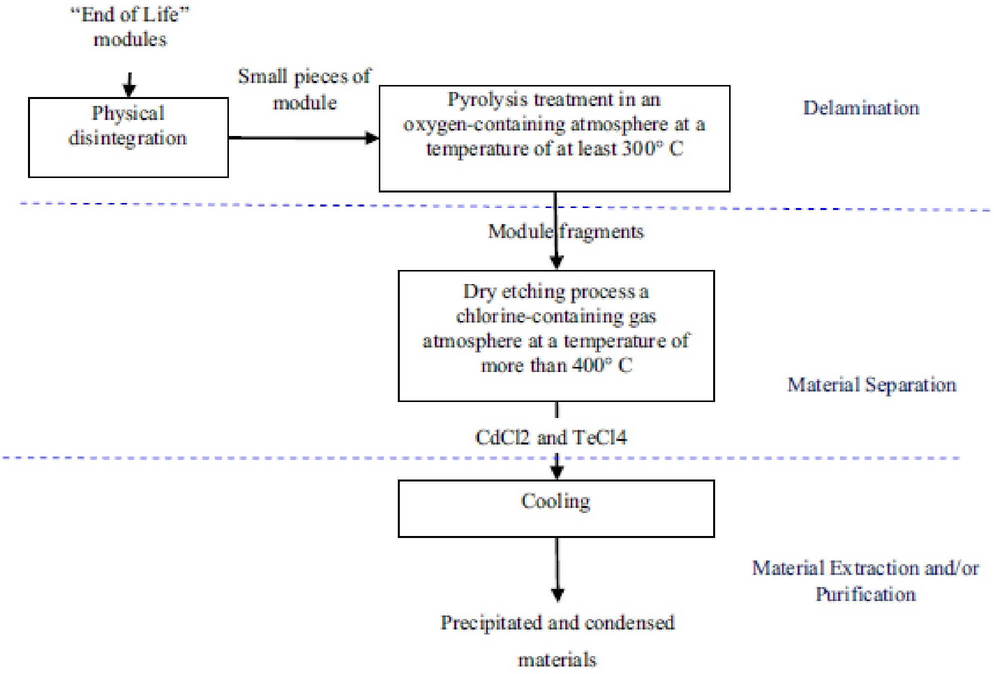


Figure 12: Recycling process of ANTEC Solar GmbH (Tao and Yu, 2015)

This approach is suitable for mixed waste treatment, requires relatively low usage of chemicals and results in a high recovery rate of glass and semiconductor material, however, it requires a lot of energy. In addition, the solar cells get broken, so it is impossible to reuse them. Wafer reproduction is required.

4.2.3 Metal extraction and refining

After delamination of the solar panels and material separation, metals are extracted and purified. Marwede et al. (2013) describe the technologies used for this purpose. Dry and wet mechanical processes, hydrometallurgical and pyrometallurgical treatments have already been commercially implemented.

Although the pyrometallurgical process is an established industrial process and the feedstock can contain different materials, the smelter needs a high throughput, and the feed requirements are very strict. A pre-treatment is necessary to reach high target material concentrations, and heavy metals or process contraries are unwanted. A high amount of glass would have to be processed as well. Therefore, pyrometallurgical processes are not used for recovering metals from PV modules anymore.

Hydrometallurgical processes are more suitable for PV panel waste fractions. Moreover, they induce low and controllable emissions. These processes are robust and technically mature, suitable for recycling a low-grade feed and a relatively low throughput (Mezei et al., 2008). Marwede et al. (2013) name the disadvantages of the hydrometallurgical processes as the many necessary separation and concentration steps and the required adaptation of the chemical process steps to target materials.

4.2.3.1 Wet-chemical methods

One example of how these processes can be implemented is described by Nieland et al. (2012). They discussed wet-chemical solutions for retrieving the pure components for further use by comparing different methods of separating Ag, Pb and Al. They suggested etching Ag from the front and back sides of c-Si by using hydrogen peroxide in combination with organic and non-organic catalyzers. An alkaline bath process was preferred for the etching of Al.

Tao and Yu (2015) state that the front and back contact of Si cells can be recycled. They are made of Ag and Al, respectively. Al can be removed by using a fresh solution of aluminium chloride and water. At the same time, poly-aluminium-chlorides can be recovered, which can be used for wastewater treatment. The silver that remains on the front contact can be dissolved with nitric acid.

4.2.3.2 Thermal, chemical and mechanical combination

Park et al. (2016) describe research for recycling of c-Si PV modules carried out by Korea Electronics Technology Institute. It is a combination method of thermal, chemical and mechanical processes. First, the module is heated to 480°C while increasing the temperature for 15°C every minute. In this way, the Si cells are recovered undamaged. Subsequently, the Si cells are etched by nitric acid and Ag is separated. Mechanical grinding is carried out to remove the anti-reflective coating, emitter and p-n junction. Then, potassium hydroxide is applied to remove the Al electrode on the back side of the cell. The recovered Si wafers can be recycled into Si cells. Their efficiency is nearly identical to that of the initial Si cell.

Klugmann-Radziemska et al. (2010) have formulated a suitable composition and concentration of the etching solution, as well as the optimal temperature range for the chemical reactions required.

4.2.3.3 Leaching

For the recovery of valuable metals leaching is proven to be effective (Tao and Yu, 2015). By this process, metals are solubilized and converted into other chemical types of bonds. For converting the compounds back to metals additional steps are required. Several acids in combination with oxidizing agents have been tested, including nitric acid, sulfuric acid, hydrochloric acid, sodium hydroxide and a solution of ferric chloride/hydrochloric acid. The disadvantages of leaching are the high usage of chemicals and a possible generation of acidic fumes. In general, the control of the chemical reactions necessary is complicated with leaching.

Cd and Te are also recovered by leaching followed by ion-exchange in sulphuric acid media (Marwede et al. 2013). Cd can be recovered by electro-winning afterwards while Te can be precipitated using sodium carbonate and sodium sulfide. Another possibility is to electrolyse the leachate. In this way, Te is recovered while further evaporation of the effluent solution is needed to obtain CdO that is suitable for recycling.

4.3 Overview of recycling pathways

4.3.1 Technologies for whole c-Si panels

International Energy Agency (2018) states that the processes for c-Si PV module recycling can be roughly divided into two groups – delamination and recovering the metals from the Si cells. Thermal, mechanical and chemical methods can be used for delamination. For recovering metals from Si cells, chemical methods such as etching can be used or a direct treatment by a metal refinery company is possible.

The comprehensive patent filing study for c-Si modules done by International Energy Agency (2018) gives a good overview of what kind of separation technologies are mostly used or being researched globally. Mechanical processing accounts for the largest part of all patents filed with 40%. The next most widely researched fields are chemical processing with 19% and thermal processing with 15% of total patents. Most thermal methods were developed for the thermal decomposition of the encapsulant. One patent used an optical method to separate the glass.

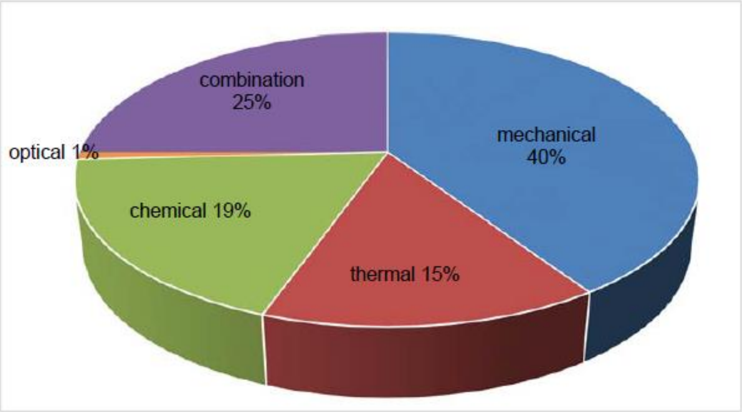


Figure 13: Share of patent filings for c-Si PV module recycling according to the processing method (International Energy Agency, 2018)

Regarding the combination of the methods, the most frequently used combination was chemical and thermal technology mixture, followed by a combination of mechanical/chemical and mechanical/thermal approaches. When a mechanical method was involved, mostly glass and solar cells were recovered. When chemical methods were used, the recovered materials included glass, solar cells and metals. Many thermal methods are used for the treatment of encapsulants, thus resulting in glass and solar cells as recyclables.

The thermal approach is typically an exothermic combustion process or a cracking process. The recyclables derived from the thermal approach are glass, Si cells and electrode ribbons. If certain conditions are met, glass and Si cells can be recovered without breakage. Tao and Yu (2015) state that wafer recovery is considered the best method of c-Si panel recycling.

Yu and Yang (2013) suggest that the commonly used delamination technologies of Si modules degrade or break the Si cells. This is a huge disadvantage because the manufacturing of Si wafers is a highly energy-consuming process. Si cell production takes up 80% of the total energy consumption for the production of c-Si PV panels (Bechník et al., 2015). Therefore, delamination methods that avoid cell damage should be prioritized. Solar cells are unchanged at the end of their life from the material point of view. There is already some practical experience gained with their recycling (Strachala et al., 2017). International Energy Agency (2018) explains that flawed cells, e.g., edge chipping or micro-cracks, typically cannot be recycled into intact wafers. However, they can be allocated as Si raw material.

The thermal approach requires a mass treatment to increase both its economy and efficiency. Another disadvantage of the thermal approach is the fluorine gas that is generated during the treatment because backsheets are often fluoride-based. Countermeasures need to be implemented in these cases.

Mechanical methods include cutting the encapsulation layer, scribing non-glass layers or glass itself and crushing. The first two technologies can recover glass without breakage that is necessary to get a higher quality and recovery rate. Glass cullet of larger particle size has the potential to improve the recovery rates in comparison to smaller particle sizes. The mechanical technologies need to be combined with a post-treatment to separate Si chips and other metals from the remaining mixture. It is typically done by chemical treatment such as etching with acid or alkali hydroxide. Proper treatment for the resulting chemical waste needs to be included. The metal recovery can also be done directly by a metal refinery company.

Chemical methods use chemical reactions for separating the components by immersing panels in solvents. The chemical approach typically requires more time than the thermal approach, but it also results in a higher yield of the recovered Si cells since they are recovered without damage.

A schematic overview of the above-described technologies used for recycling c-Si-based solar panels is provided in Figure 14.

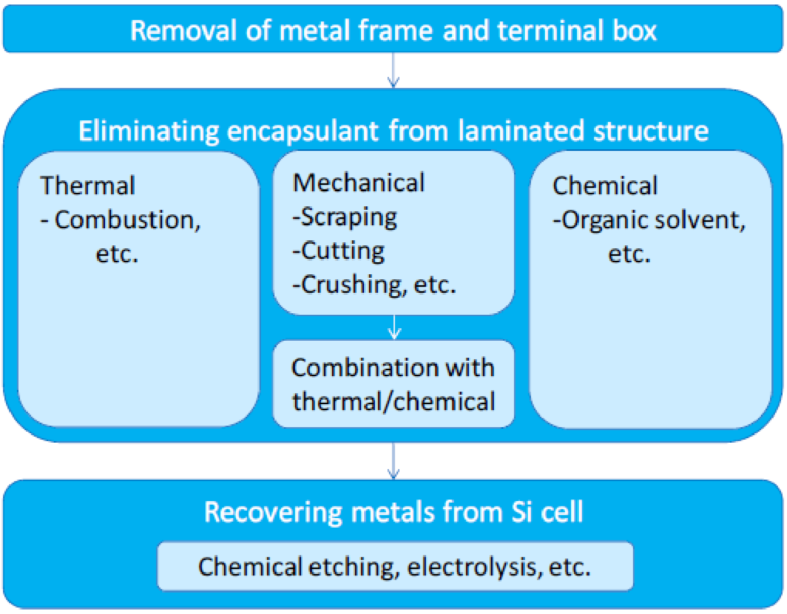


Figure 14: Recent R&D for c-Si PV module recycling (International Energy Agency, 2018)

According to International Energy Agency (2018), the commercially used approach for end-of-life c-Si panels currently in Europe is the mechanical approach, namely crushing and sorting. By additional processes, metals will be recovered from the crushed fraction. Chapter 4.2.1.7 describes in more detail how the combination of these methods leads to the following output fractions – glass, Ag and aluminium. The biggest disadvantages are the low purity of glass and the loss of Si wafers. Preparing for future high waste streams and the opportunity for material recovery therein, further technical innovations will be required to realize economical processes, achieve a higher recovery rate and improve the quality of the recovered materials.

4.3.2 Technologies for c-Si individual solar cells

Strachala et al. (2017) state that the demand for silicon is growing. The recycling of silicon cells should receive higher attention along with this trend. Park et al. (2015) divide the PV recycling process into two main phases, namely separation of PV cells and cleaning of the cell surface. The cleaning of the cells can be performed chemically or by laser techniques. Unwanted layers, such as anti-reflective coating, metallization and PN junction, are removed.

Franz and Piringer (2020) claim that the current silicon recovered from primary recycling of kerf and end-of-life PV modules has low quality and therefore cannot be used to produce new silicon wafers. There is a target set for research to produce secondary silicon with a purity of 99.9% with a lower carbon footprint than virgin silicon material. The recovery of pure silicon is both technological and economical one of the major challenges. It needs to be separated from adhesives, metallisation and other impurities to obtain the necessary quality required for new silicon wafers.

The most currently researched methods lead to the breakage of the Si cells. In the case of the thermal approaches, this effect is due to the thermal changes of EVA during the heat treatment (Lee et al., 2017). There are two phenomena involved. One is the expansion and the shrinkage of the EVA. The thermal deformation of the EVA on the back side of the cell applies stress to the solar cell. The other one is the generated gases that form bubbles behind the glass exerting stress on the cells.

The expansion and the shrinkage of the EVA occur when the temperature increases, which can lead to cracks in the solar cell. The different thermal expansion behaviours of the components of a PV module often result in internal stresses because the coefficient of thermal expansion of the EVA is much larger than that of Si.

4.3.3 Technologies for chalcogenide-based panels

International Energy Agency (2018) gives an overview of the compound semiconductor PV module recycling status. A combination of mechanical and chemical processes (crushing and chemical etching) is under commercial operation by First Solar. Several other technologies are under development.

The different processes can roughly be divided into those that eliminate the encapsulant from the laminate and those that recover metals and substrate glass after pre-disassembly (Marwede et al. 2013). Mechanical crushing has been proven for eliminating the encapsulant from laminated structure, however other thermal, mechanical and optical approaches have been developed as well. For recovering semiconductor metals from the substrates and substrate glass itself, chemical approaches such as etching are effective. As an alternative, mechanical scraping can be used in case the substrate is recovered without breakage. A separation of the metallic and non-metallic fractions needs to be applied. As the last preparation process for the actual material recycling, the extraction and refining of metals are carried out. A graphical overview is provided in Figure 15.

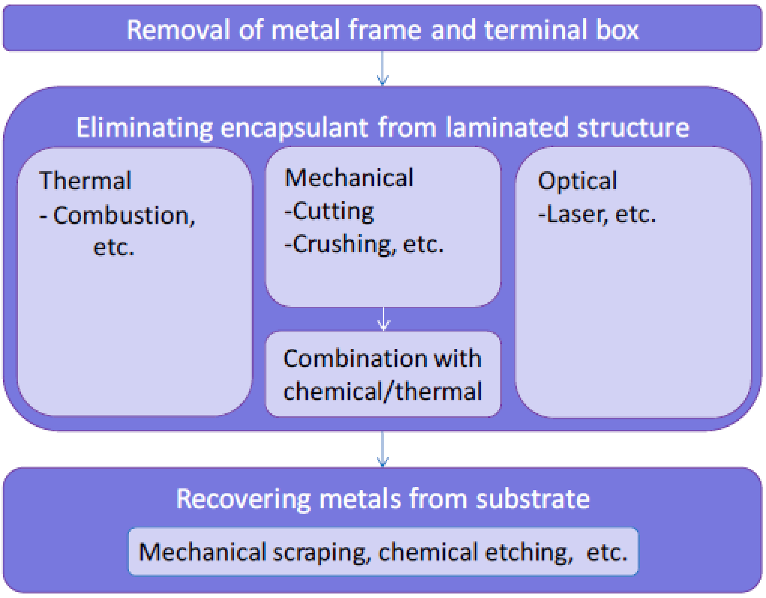


Figure 15: Recent R&D for compound PV module recycling (International Energy Agency, 2018)

4.3.4 Overview of currently operating recycling facilities

In Table 1 an overview of the researched technologies and recycling facilities is given for a better understanding of the current state in the recycling field of the waste PV panels, including the geographic location of the facility or research activities. The geographic location is important not only for logistics of waste management in case of recycling facilities but also in case of R&D activities due to the facilitated knowledge transfer with an increasing vicinity.

As it is visible from Table 1, only five operating recycling facilities for PV panels have been identified within this study. The vast majority of activity is under research and development. The list of the technologies and recycling facilities explored in this master thesis is not intended to be exhaustive.

Table 1: Overview of operating recycling facilities

Institution	Scale	Module type	Technology and output summary[1]	Status	Location
Reiling	Facility	Si	Combination of mechanical treatments for recovery of glass and possibly other materials	Active[2]	Germany
Wilhelm Geiger GmbH & Co. KG. (earlier Exner Trenntechnik)	Facility	Si	Dry mechanical processes for glass recovery	Active but PV panels are not treated anymore[3]	Germany
Green Energy and Environment Research Laboratories, Industrial Technology and Research Institute	R&D pilot plant	Si	Thermal delamination for recovery of whole glass plates, Si and Cu	Tested	Taiwan
Padua University	R&D	Si	Electro-thermal heating process for glass recovery	Tested	Italy
Solarworld GmbH	Facility	Si	Thermal delamination, manual separation and etching for recovery of glass, Ag, Cu, solar cells	Inactive	Germany

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[1] Aluminium can always be recovered if the frame is removed before the treatment

[2] Source: Reiling, 2021

[3] Source: Gögler, G, 2020

Sasil, S.p.A. et al.	R&D	Si	Infrared heater, roller mill and vibrating knife equipment followed by chemical methods for recovery of glass and metals	Tested	Europe
Mitsubishi Materials Corporation	R&D	Si	Scraping for recovery of glass and Ag	Tested	Japan
Toho Kasei Co. Ltd.	R&D	Si	Scraping and treatment by a solvent for recovery of glass, Si, other metals and encapsulation polymer	Tested	Japan
Yingli Solar et al.	R&D	Si	Crushing, refrigeration, grinding, physical separation for recovery of Si, Ag, Cu and glass	Tested	China
Veolia	Facility	Si	Shredding, magnetic separator, fine crusher, screening, non ferrous separator, optical and inductive sorting for recovery of glass	Active	France
LuxChemtech (earlier Loser Chemie GmbH)	R&D	Si	Solution of aluminium chloride and water followed by nitric acid for recovery of glass, encapsulation foil, fluor plastic foil, Si, Ag and Al from the back contacts of Si cells	Tested	Germany
Yokohoma Oils & Fats Industry	R&D	Si	Solution with neutral and alkali solvents and crushing for recovery of glass, encapsulant, Si and electrode ribbons	Tested	Japan
Korea Research Institute of Chemical Technology and	R&D	Si	Solution with organic solvent and ultrasonic irradiation for Si cell recovery	Tested	Korea

Kangwon National University					
5 N Plus	R&D	Both	Irradiation by a laser for the destruction of EVA	Tested	Canada
Suez and Fraunhofer	R&D	Si	Delamination by pyrolysis for freeing metals, glass and silicon for further electrochemical treatment	Tested	Germany
Korea Electronics Technology Institute	R&D	Si	Thermal treatment, etching and mechanical grinding for recovery of Si cells and Ag	Tested	Korea
Flaxres GmbH	R&D	Both	Plasma jet by flashbulbs separates all materials for further recovery process	Tested	Germany
Kitakyushu Foundation for the Advancement of Industry, Science and Technology	R&D	Both	Milling of the backsheet, heating and scraping for glass recovery; cyclone collector for the recovery of In und Se	Tested	Japan
Hamada Corporation and NPC Incorporated	R&D	Both	Heated cutter for recovery of glass and metals	Tested	Japan
First Solar	Facility	Thin-film	Shredding, crushing by hammermill, etching, precipitation, vibrating screen and rinsing for recovery of glass and a metal-rich filter cake (Cd and Te)	Active	USA, Malaysia, Vietnam
EU-LIFE project RESOLVED	R&D	Thin-film	For intact panels: thermal delamination, blasting of the substrate glass for glass recovery. For broken panels:	Tested	Europe

			crushing, milling, attrition, sieving floatation for recovery of glass and Te, In, Cd		
Antec Solar GmbH	R&D	CdTe/CdS and possibly Si	Physical disintegration, pyrolysis, etching, cooling precipitation for recovery of glass, Cd and Te	Tested	Germany

4.4 Design-for-recycling concepts

Summarizing the results of the separation technologies from chapter 4.2, it becomes clear that the PV panels are relatively complex to recycle. Almost all recycling approaches include a combination of different separation technologies to achieve high quality and quantity of the output fractions. The background for this issue is the focus on the efficiency and durability in the PV industry (Lunardi et al., 2018). Different types of efficient and cost-effective materials have been researched, as well as technologies of keeping them bonded and protected for years exposed to the external environment. Recycling of the materials however requires the separation of them, which is contrary to the objective of the manufacturing. Therefore, design-for-recycling or design-for-reuse should be considered during manufacturing to facilitate the reuse of the PV panels or the recovery of materials (Scherhaufer et al., 2020).

Design-for-recycling is a concept of product design with a target to improve the recyclability of the product and its components, as well as to maximise the output of the recycling process (Allesch et al., 2019). The focus of design-for-recycling lies on the ease of mechanical dismantling to facilitate shredding, regeneration and recycling (Shahbazi and Jönbrink, 2020). All products inevitably fail eventually, even if their lifetime is prolonged, or the products have been reused. This is the reason why among all the different environmental designs, design for recycling should always be considered. Nevertheless, only a few design-for-recycling concepts have been developed (Allesch et al., 2019). An important reason for this is the currently small waste PV panel quantities with no existing recycling market available.

The design-for-recycling includes information declaration on the material content and whether it is possible to separate and recover the semiconductor layer from the frame, glass, encapsulants and backsheet (Franz and Piringer, 2020). The general suggestion derived from the literature for design-for-recycling of PV modules is to leave the encapsulation material out or to substitute the commonly used materials with recyclable ones. In the following subchapters, several alternative possibilities for the protection of the cells instead of the encapsulation films are described. The list of design-for-recycling concepts is not intended to be complete.

4.4.1 Frame sealant

The frame is glued to the laminated structure to protect the PV panels from moisture ingress (Allesch et al., 2019). As Goris (2014) claims, commonly used edge sealants are DOW Corning 804 or a double-sided adhesive tape, e.g., Duplomont. By using these materials, the frame is difficult to remove, and this results in a distortion of the frame during the manual removal. Alternative frame sealants are recommended for easier removal of the frame, such as:

- O-ring
- U-profile
- Sponge rubber
- Single-sided adhesive tape (no adhesion in frame)

The O-ring, the U-profile, the sponge rubber and the single side adhesive tape were tested in 500 hours of damp-heat test at a temperature of 85°C and relative humidity of 85%. Only the PV panel type sealed with the sponge rubber showed discolouration of the copper, indicating that the other sealant types performed well in this test.

This concept is intended for an easier frame removal. Apart from the sponge rubber that led to discolouration of the copper, no disadvantages of the alternative frame sealant options have been documented.

4.4.2 Lead alternatives

In the future lead and lead-compounds from the output glass will have to be removed (International Energy Agency, 2018). Manufacturing c-Si PV panels is also possible without this toxic heavy metal. Hutchins (2018) describes the current state of lead use and the lead-free alternatives in the PV industry.

A typical commercially produced 60-cell c-Si module contains up to 12 g of Pb. Most of it is found in the ribbon coating and soldering paste that connects the cells. Currently, most PV manufacturers use a lead-containing ribbon. The substrate of the ribbon is made of Cu, the coating is made of 67% Sn and 37% Pb. During the soldering, the coating layer melts and thus contacts the Ag bus bar. Pb is used because of its melting point. It allows a lower process temperature during the stringing. The reduced temperature reduces the stress on the cells. While using Pb, soldering at 210°C is possible. Lead-free soldering requires a temperature of about 260°C. At this temperature risk for microcracking and a higher breakage rate during production arises.

Ribbon manufacturers have tested lead-free solder by replacing the Pb with Bi and using a pure Sn coating. These methods require a higher processing temperature and increase the costs.

Another possibility is electrically conductive adhesives (ECAs). These are different types of glue embedded with a metal, mostly Ag, to make the glue electrically conductive. The feasibility of ECAs as an alternative has been proven, and the manufacturing is possible without an increase in temperature. The use of Ag makes the ECAs more expensive than solder, and the Ag is also on several restricted chemicals lists because of its short supply. Nevertheless, the stringer supplier Teamtechnik has successfully developed a stringer using this method in production lines for heterojunction technology PV panels in 2018.

The manufacturer of the PV module production line Meyer Burger has developed a Smartwire interconnection technology, that replaces standard busbars and ribbons with copper wires coated with a thin low melting point alloy layer of indium-tin and supported by a polymer foil (Faes et al., 2014). An indium-free coating has been developed as well, and it consists of 58% Bi and 42% Sn. This technology lowers ohmic losses and improves light management and thus increases the overall module efficiency. The low temperature during the lamination ensures less thermo-mechanical stress. The Ag consumption is reduced by at least 85%. This technology has also been used in large-scale heterojunction technology PV panel production.

PV panel manufacturer SunPower has developed a panel design using the interdigitated back contact solar cells, thus avoiding the use of a ribbon and Pb (SunPower, 2021). On the back of the cells, a thick layer of tin-plated copper is used that holds the cells together. This technology offers the highest efficiency solar panels currently available with enhanced durability.

The disadvantages of the lead-free ribbons are the higher processing temperature and increased costs. The higher temperature induces risk for microcracking and a higher breakage rate during production. The disadvantage of other lead free concepts is the higher costs. Some concepts offers an increased overall module efficiency and/or enhanced durability though.

4.4.3 Double encapsulation module

Doi et al. (2005) have developed an improved PV module structure, which is easier to disassemble and has a recyclable rate of over 97% and a cell recovery rate of more than 90%. The common EVA film is used as the encapsulant on both sides of the cells. Beneath the EVA a layer of a non-adhesive film is inserted for an easy release during the cell recovery. This concept is called the double encapsulation module. Damp Heat tests proved the reliability of this structure. However, the power output is reduced because of the air gap between the non-adhesive sheet and the front side of the cells, which results in a worse optical incoupling.

Li et al. (2011) improved this concept by introducing an optical coupler that reduces the electrical performance loss compared to the double encapsulation module. The structure of the double encapsulation module with an optical coupler (DEMOC) is depicted in Figure 16 as a cross-sectional view. The air gap described in the previous paragraph introduces optical reflection, therefore an additional layer of optical coupler between the front non-adhesive film and the front side of the cell is used. It reduces the optical reflection.

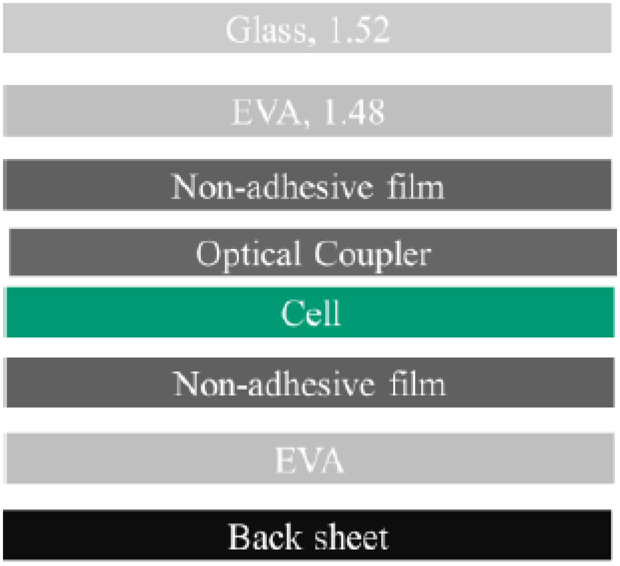

Figure 16: The structure of the DEMOC module design: a cross-sectional view (Li et al., 2011)

The non-adhesive film should be optically transparent, stable and compatible with other components. For the respective study, a commercial grade of ETFE is used.

The optical coupler should be optically transparent, stable regarding heat and optical characteristics and suitable for a lamination process. A commercial-grade of liquid silicone is selected for this study.

The disassembly of the double encapsulation module is easier than that for the DEMOC design. First, the backsheet is cut along the cell perimeter, keeping a short distance from the cell edges for both module designs. Then, the whole cells can already be removed from the rest of the panel for the double encapsulation module.

As for the DEMOC design, the optical coupler made from the liquid silicone has a weak adhesion with the front side of the Si cells, therefore it cannot be directly removed. It is possible to remove the cell together with the optical coupler from the non-adhesive layer by using a thin wire. Then, the optical coupler can be easily removed from the cells. The cell recovery rate for DEMOC design is only 50%. Further improvements could be made by tuning the liquid silicone formulation for the optical coupler and the final curing state for adjusting its surface property.

The disassembly of DEMOC is relatively complex, however 50% of the cells can be recovered.

4.4.4 Encapsulation alternatives

The elimination of the encapsulant from the laminate is one of the most difficult and important targets of the PV panel recycling process (International Energy Agency, 2018). Encapsulants must withstand very high requirements regarding durability and safety over a long time in diverse climatic and operational conditions (Wiesmeier et al., 2013). The microclimatic conditions are the reason for the degradation processes in the polymeric materials, which are influenced by other materials of the module. In independent tests, different encapsulation materials have been tested (Mūsiņa, 2014-2018). The results have shown that the encapsulation material has a great influence on the power output and the longevity of the whole panel. If the encapsulation material starts to degrade, other parts of the panel can start to corrode, making the selection of the encapsulation material and method crucial.

Currently, only thermal treatment is economically viable for the encapsulation films of PV panels according to a study by Johannes Kepler University (Wallner, 2013). The multiple-layer composition of panels requires a chemical treatment to separate the layers from each other, therefore material recycling is currently not ecologically meaningful. Moreover, PET and EVA are very cheap materials, and this questions their recycling from an economic point of view.

In the following subchapters, encapsulation materials and PV module structures that are suitable for recycling at their end of life are described.

4.4.4.1 Backsheet alternatives

The combustion of the most used backsheets made of polyvinyl fluoride under the brand name Tedlar® has severe disadvantages (Wallner, 2013). It may induce corrosion of incineration plants and are carcinogenic. Moreover, the resulting fluorinated hydrocarbons have a high global warming potential. In addition, highly toxic hydrofluoric acid can be formed. Therefore, it is suggested to use backsheets without fluoropolymers, such as polyamide or polyester. Such backsheets are available on the market, however, Tedlar® is still widely used for c-Si panels (Mūsiņa, 2014-2018). A glass plate instead of a backsheet can also be used, however, it results in slightly higher manufacturing costs of the panels and the panel is heavier. On the other side, independent damp-heat tests have shown that glass-glass panels perform better in terms of power output and longevity than glass-foil panels with Tedlar® in the long run.

The different composition of the backsheets is another impediment to their recycling (Allesch et al., 2019). This could be solved by using backsheets consisting of a single material, e.g., polyolefins or glass.

The recyclability increases and less greenhaus gas emissions and toxic acids are generated during the EoL. Glass as an alternative is slightly more expensive and the panel is heavier, however the power output and longevity increase.

4.4.4.2 Thermoplastic silicone elastomer

The most widely used material for the encapsulation of cells is EVA (Wiesmeier et al., 2013). It has been used in the PV industry for more than twenty years. Besides resulting in accelerated aging of PV panels, this material is also unsuitable for eventual material recycling.

Thermoplastic silicone elastomers as encapsulation material do not require additives for the cross-linking during the lamination process as opposed to EVA, and the cross-linking is performed via hydrogen bonds. Therefore, the modules with thermoplastic silicone elastomers are easier recyclable than the modules with EVA.

4.4.4.3 Vacuum technology

Another concept that facilitates the recycling of Si cells is the New Industrial Solar Cell Encapsulation (Allesch et al., 2019). This technology requires no lamination at all. The encapsulation of the cells is enabled through a vacuum that is established between the front and the backsheet of the panel. In this way, the cells are under pressure to enable electrical contact between the cells and the metal connections without soldering. The absence of lamination caters for an efficient and low-cost disassembly at the end of life (Einhaus et al., 2018). The panel components can be recovered as entire pieces. This ensures a high-value recycling and more reuse potential.

However, the seal tightness of this type of panels is questioned and therefore the overall efficiency of these panels is assumed to decrease. The reasons for this are the air gap and electrical connections held together by internal module vacuum pressure (Norgren et al., 2020). Moreover, the electrical efficiency would vary with changes in temperature and altitude.

4.4.4.4 TPedge technology

TPedge modules are made without encapsulation foil and lamination (Fraunhofer Institute for Solar Energy Systems, 2017). These double-glass modules have an edge sealant and have a similar construction to insulating glass windows. The Si cells are fixed between the glass plates with adhesive pins as visible in Figure 17. The pins cover only 0.02% of the cell surface (Norgren et al., 2020). Some glue-like material is still required on portions of the front glass and rear materials. Since the double-glass modules have enough mechanical stability, TPedge modules do not require a frame (Fraunhofer Institute for Solar Energy Systems, 2017). The omission of several components of a typical PV panel and the time-consuming and energy-intensive lamination process leads to a cost reduction in PV panel manufacturing.

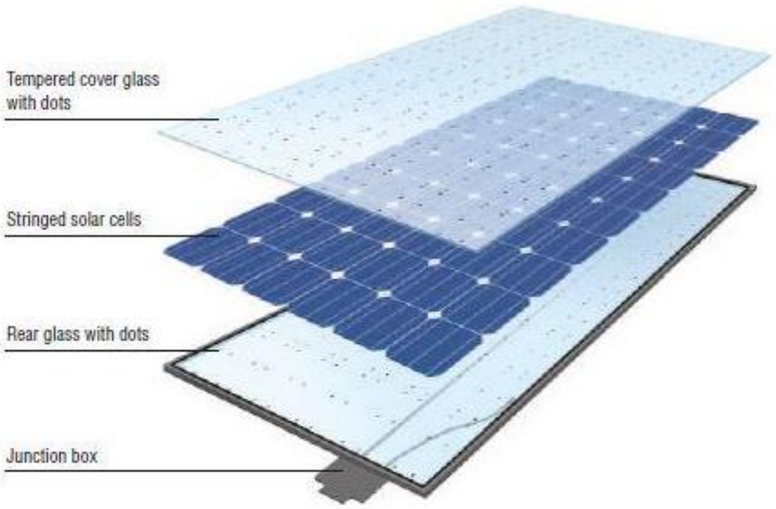

Figure 17: TPedge structure (Glaston, 2021)

Fraunhofer Institute for Solar Energy Systems together with the company Bystronic glass have developed a process to transfer this concept to industrial production. An automated production plant has been successfully commissioned. The Si cells are glued to the back glass with the help of adhesive pins. Then, the front glass plate is put in place, and the edges are sealed with a thermoplastic material at 130°C (Glaston, 2021). The space between the two glass plates is filled with gas.

Thanks to this panel construction, the aging stability of the module is considerably increased in comparison with the commonly manufactured PV panel types (Fraunhofer Institute for Solar Energy Systems, 2017). This has been proved in the damp-heat and the thermal cycle durability tests. The mechanical stability tests were also successfully passed. Tests for recyclability have not been specifically conducted, however, the design indicates that it could be higher than for the typical module types (Norgren et al., 2020).

No disadvantages of this concept have been detected, except that different production lines than the currently used ones are required.

4.4.4.5 Edge sealant alternatives

Kempe et al. (2010) summarize the necessity of edge sealing for a stable power output over the expected lifetime of PV panels. Many types of PV technologies are sensitive to moisture. Even if the front and the back sides of a module are covered by an impermeable sheet, the moisture can diffuse from the sides. Therefore, a proper edge sealing material should be used to prohibit moisture ingress in the panel throughout the expected lifetime of the panels. In the search for proper sealing material, the polyisobutylene materials have proved to be sufficient to ensure no moisture ingress for 20 to 30 years.

Flaxres GmbH is working on the replacement of the polymer films commonly used as the edge sealants with a glass paste (Heuschkel, 2021). It is possible with their flashbulb technology and would not require proof of the airtightness as it is in the case of the rubbers conventionally used as the edge sealants in the PV industry. This edge sealing type could enhance the longevity of the PV modules - possibly up to 100 operational years. Moreover, the glass paste can be applied as a thin layer, whereas the synthetic materials need to be applied as a thicker layer taking away the space of the solar panels that could otherwise be used for electricity generation.

Usually, the edges of the c-Si PV modules are sealed with silicon-based materials and degrade in independent damp-heat tests, indicating that the water vapour enters the laminate over time (Mūsiņa, 2014-2018). Constant temperature change also degrades the edge sealant. If the edges are insulated with a material of higher durability, e.g., polyisobutylene-rubber, the power remains stable in the damp-heat tests over the same time as compared to the conventional silicon-based materials. The exact transition of the laboratory conditions used in the tests to the real-life conditions is not possible, therefore the lifespan of PV panels judged from the laboratory tests remains a rough estimate.

The advantage of the edge sealant alternatives is the use of recyclable materials with no documented disadvantages.

4.4.5 Overview of design-for-recycling concepts

In Table 2 an overview of design-for-recycling concepts described in chapters 4.4.1-4.4.4 is given. Their main effects on recycling are outlined. The effects on the costs cannot be plausibly judged since most of these concepts and recycling methods are still in the development phase.

Only the latest version of the double encapsulation modules is included, namely the one with the optical coupler. Its predecessor, the double encapsulation module without the optical coupler, shows reduced power output and therefore is not likely to be well received in the efficiency-driven PV industry. Apart from it, all other concepts are included in the overview.

The improvements resulting from design-for-recycling concepts lead to at least one of the following benefits regarding recycling:

- a more efficient disassembly of the solar panels in terms of time and costs
- an increased share of recyclable materials
- less pollution
- components recovered in whole
- recyclables of higher quality because of increased material purities

This implies that the different design-for-recycling concepts improve different aspects of recycling.

Table 2: Summary of design-for-recycling concepts

Design-for-recycling concept	**Concept summary**	**Improvements regarding recycling**	**Disadvantages**
Backsheet Alternatives	• polyamide or polyester • polyolefins • glass plate	• less GHG and toxic acids • if glass or polyolefins as a substitute – increased share of recyclable materials	• glass is more expensive • glass is heavier
Encapsulation Material Alternatives	• thermoplastic silicone elastomers • instead of lamination, vacuum is established between the front and the backsheet	• Efficient and low-cost disassembly/components recovered as a whole	• seal tightness of vacuum panels is questioned • electrical efficiency varies with changes in temperature and altitude (vacuum panels)
TPedge	• absence of encapsulation foil and lamination; Si cells are fixed between the glass plates with adhesive pins	• Higher recyclability expected	• Different production lines required
Double Encapsulation Module with Optical Coupler	• Beneath the EVA a layer of a non-adhesive film is inserted for an easy release during the cell recovery	• 50% of cells can be recovered	• Disassembly relatively complex

Lead Elimination	• replacing the Pb with Bi and using a pure Sn coating • glue embedded with a metal, mostly Ag, to make the glue electrically conductive • replacement of standard busbars and ribbons with copper wires • interdigitated back contact solar cells through a thick layer of tin-plated copper	• Recyclables of higher quality	• risk for microcracking and a higher breakage rate with lead-free ribbons • higher costs
Frame Sealant Alternatives	● O-ring ● U-profile ● sponge rubber ● single-sided adhesive tape (no adhesion in frame)	• Easier removal of the frame / no distortion	• Not documented, apart from the sponge rubber that led to discolouration of the copper
Edge Sealant Alternatives	● glass paste ● polyisobutylene-rubber	• Increased share of recyclable materials	• Not documented

5. Conclusions

The findings of this study confirm the early stage of the end-of-life management of disposed PV panels. The complete legal framework, schemes and procedures for recycling implementation are still to be established. This is mostly explained by the fact that the current global waste PV panel streams are negligible compared to the typical quantities in the waste management sector. Relatively large amounts of annual waste are anticipated by the early 2030s.

End-of-life PV panels are typically treated in separate batch runs in existing recycling plants for the main components of a PV module, namely glass or metal recycling facilities. In these plants only the metal frame and glass can be recycled, however, the impurities in the glass are so high that it can be merely downcycled. In the long term, dedicated PV module recycling plants have the potential to increase treatment capacities and maximise revenues due to better quality of recyclables and the ability to recover a greater fraction of materials. In some cases, the end-of-life PV panels end up with electronic waste recyclers. These quantities are so small that the panels are shredded together with other WEEE whereas is some cases the frame and junction box are not even removed before the shredding. Merely aluminium is recovered and sometimes also glass, however it can only be downcycled because of its impurities. In cases where the glass cullet has an attached polymer layer with it, it is treated as commercial waste or is incinerated, depending on the composition. In developing countries, the end-of-life PV panels are improperly landfilled implying that no material recovery takes place, at least at this stage of the waste management.

The separation of compound materials has been identified as the biggest challenge in the technical implementation of dedicated recycling of thin-film and c-Si PV panels. In this master thesis, both established separation technologies of the compound materials used in the above-mentioned PV panel types and promising ones in the research or pilot phase have been studied. Almost all separation approaches include a combination of different separation technologies to achieve high quality and quantity of the output fractions which makes the separation process relatively complex. Design-for-recycling or design-for-reuse should be considered during the manufacturing to facilitate the reuse of the PV panels or the recovery of materials. The separation technologies have been studied by the means of a literature review and interviews by experts from the field. Design-for-recycling concepts have been either derived from the existing unconventional c-Si PV technologies or found in the literature as existing design-for-recycling concepts.

Separation technologies depend on the panel type and the manufacturing technology. They can be divided into mechanical, chemical, thermal, optical methods and combinations of thereof. After the selected separation technologies, the aftertreatment in the form of recovering methods takes place. Recovery methods are divided mainly into hydrometallurgical processes to recover metals and pyrolytic processes. An overview of the different separation technologies is provided in Figure 10.

When summarizing the findings on the recycling technologies currently available or under R&D, it seems that for obtaining a higher share and/or quality of recyclables at least one of the following is required:

- chemicals

- high energy usage
- long treatment time
- innovation

Since chemical usage creates new waste, high energy use creates additional emissions and a long treatment time is economically inefficient, expectations lie on the innovation. The current status of the flashbulbs operated by Flaxres GmbH seems to be promising for fulfilling these expectations.

Another aspect to stress is the technical feasibility of high-quality recycling of the cells. This would enable high-purity glass and silicon recyclables; however, it is mostly omitted due to economic reasons (Scherhaufer et al., 2020).

International Energy Agency (2018) observed increased R&D efforts in the mechanical recycling approaches while subsiding activities for the chemical approaches directed to delamination. The declining interest in the chemical approaches is due to the long treatment times that are not likely to be suitable for large-scale recycling. The chemical approaches may suit the small-scale on-site treatments though, e.g., at c-Si module manufacturing factories.

In general, PV panel structure hinders recycling due to the following reasons:

1. materials used for backsheets
2. encapsulation methods
3. frame and edge sealants
4. lead use

The first three points can be summarized under composite use in PV panels and lamination. A general suggestion derived from this insight would be the substitution of composite materials and the lamination process. Only a few design-for-recycling concepts have been found in this study. An important reason for this is the currently small disposed PV panel quantities with no existing recycling market available.

Five dedicated PV panel recycling facilities that are currently in operation have been found worldwide. Sener and Fthenakis (2014) estimate that only about 10% of PV modules worldwide are currently recycled. The main reason for this is the lack of regulation and the fact that recycling is a financially unfavourable option compared to landfilling if the externalities are not considered. This means that the environmentally more favourable end-of-life management option, namely recycling, should be enforced through the legislation for it to become implemented more often. The legislation regarding this is slowly emerging in the last years, however only in the developed countries.

Two of the dedicated recycling facilities are located in Europe (Germany and France) and both of them treat only Si solar panels. Three recycling facilities specialise in thin-film panels and are located in the USA, Malaysia and Vietnam as an integrated part of the thin film panel manufacturing factories. This implies that currently, high-quality recycling of Si solar panels is possible only in the central part of Europe and that of thin-film panels in the USA and Southeast Asia. This is a very poor geographical dispersion and therefore a logistics problem due to the distributed solar PV installations. Flaxres GmbH plans to create mobile recycling facilities that would ease this issue.

Due to the current conditions relevant to PV panel recycling, such as the price for the secondary materials and the currently low PV panel waste streams, an economically profitable PV panel recycling facility is generally considered to be unfeasible (Allesch et al., 2019). This view is held also by an employee of PV Cycle, with no prospects of an economically viable recycling technology at the moment (Magent, 2019). According to the statements of Flaxres GmbH, their technology should be economically viable once it gets commercialised (Heuschkel, 2021).

The currently commercialised PV panel recycling technologies show plenty of room for improvement also in terms of quality and share of recyclables. The R&D efforts in this sector are ongoing and the vast majority of activity in the recycling of disposed PV panels is under R&D. Also design-for-recycling needs to be further developed and included in the manufacturing practices. Design-for-recycling concepts can be more expensive than conventional production because of the more expensive materials. Sometimes they are not considered because of the investment needed for the existing production lines.

5.1 Outlook

The end-of-life management of PV systems will face major challenges when the first large-scale free-field plants reach the end of their lifetime in about 15 years. Even though the waste treatment requirements and financing of PV module recycling and disposal in the European Union are regulated by the WEEE Directive, some major legal, financial and technical questions are still open. One key issue is the classification of PV modules as hazardous or non-hazardous waste since the modules contain small amounts of hazardous substances, such as Pb or Cd, varying with module technology.

An easily implementable improvement for PV panel recycling would be a list of all components of each panel type that is publicly available to make sure the responsible entity dealing with the end-of-life PV panel after its long operating lifetime knows the material composition that can be crucial for recycling (Ledersteger, 2020).

Studies on life cycle assessments have been carried out to compare the various recycling technologies. However, as the recycling technologies and concepts of design-for-recycling are still under development, updated comparisons between the different options for handling the end-of-life PV panels as well as environmentally friendly material compositions of the panels need to be further performed.

Another aspect of the rapidly changing PV industry is the potential change of the required recycling technology (Allesch et al., 2019). Although the average lifespan of PV panels is 30 years, recycling technologies need to be adapted to the changing PV technology to be able to recycle them. According to Huang et al. (2015), PV panel manufacturers operate under tight profit margins well below 10% before interest and taxes, therefore the recycling costs cannot be ignored. These aspects together could lead to an unintended shift in manufacturing towards the PV technology that is relatively less costly in its treatment even if its overall environmental impact is higher. For monitoring this possibility, the comparisons regarding the environmental sustainability factors should be ongoing.

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