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THE USE OF RECYCLED SEMICONDUCTOR MATERIAL IN CRYSTALLINE SILICON PHOTOVOLTAIC MODULES PRODUCTION - A LIFE CYCLE ASSESSMENT OF ENVIRONMENTAL IMPACTS

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Abstract

To offset the negative impact of photovoltaic modules on the environment, it is necessary to introduce a long-term strategy that includes a complete lifecycle assessment of all system components from the production phase through installation and operation to disposal. Recycling of waste products and worn-out systems is an important element of this strategy.

As the conclusions from the previous studies have shown, thermal treatment provides an efficient first step in the recycling process, while chemical treatment was more advantageous in the second step.

This study aims to assess the environmental impact of recovering and recycling the valuable semiconductor silicon wafer material from photovoltaic solar cells. A comparison was made between producing new solar cells with or without recycled silicon material.

The analysis of the photovoltaic cell life cycle scenario including material recycling presented in this article was performed using SimaPro software and data combined and extended from different LCI databases. The idea is that the use of recycled materials, which were energy-consuming in the primary production stage, allows to meaningly reduce the energy input in the secondary life cycle.

All stages of the silicon cell life cycle contribute to the Global Warming Potential (GWP) and greenhouse gas emissions reductions through the use of recycled silicon material represented 42%. The total environmental impact of photovoltaic production can be reduced by as much as 58%, mainly through reduced energy consumption in the production process of high purity crystalline silicon.

Keywords: photovoltaic solar cells, recycling, life cycle analysis, silicon, metals and glass recovery

Introduction

Photovoltaic (PV) technology is considered an energy source responsible for relatively small amounts of waste, as none is generated during the lifetime of PV modules. We should not however, ignore the waste stream generated at the end of the exploitation phase of PV installations. A small stream of waste is also created at the production stage of products that are rejected by quality control, as well as during operation in the case of damaged modules, which show reduced efficiency (e.g., anti-reflective coating defects).

In the European Union, photovoltaic modules are defined as e-waste in the WEEE Directive (Directive 2012/19/EU of the European Parliament and of the Council of 4 July 2012 on waste electrical and electronic equipment). The term 'electrical and electronic equipment' or EEE is defined as equipment designed for use with a voltage rating not exceeding 1,000 V for alternating current and 1,500 V for direct current, equipment dependent on electric currents or electromagnetic fields in order to work properly, equipment for the generation of such currents, equipment for the transfer of such currents, or equipment for the measurement of such currents.

Reviews of PV life cycle assessment (LCA) have been published [1–4]. However, they do not take into account the stage involving recycling and re-use of the silicon material, the production of which, due to the high purity required, significantly impacts the environment. The reason why scrap silicon from electronic-grade silicon production has not been taken into account is because its share in the market in 2005 was only 5% [5]. In this calculations silicon from PV modules recycling was not considered as scrap from production. Some researchers have included the end of life (EoL) phase of PV modules by burying the waste in landfills or recycling them [6], but recycling covers only the glass, plastic, metal components, and other waste materials without recovery of the valuable semiconductor material.

Life cycle inventory (LCI) databases for silicon photovoltaic modules are generated with data from eleven European and American (US) photovoltaic companies participating in the European Commission's Crystal Clear project [5]. Many researchers focused on the energy payback time (EPBT) [1, 7-21]. The EPBT is between 3.5 and 5 years, depending on ambient irradiance, and for systems installed in desert areas that use crystalline silicon (c-Si) modules the EPBT is 2.5 years [22]. Most of the energy consumption of PVs is linked to the module production step [4]. However, it is worth noting that EPBT is an index dependent on insolation (localization) [23], and therefore cannot be universally applied to comparisons.

On the other hand, the environmental impacts of a polycrystalline PV module and a wind turbine using the LCA method were compared [6]. This study modelled landfill disposal and recycling scenarios for decommissioned PV modules and wind turbines, and compares their impacts to those of the other stages in the life cycles. In contrast to this analysis, another LCA was based on the recycling step focusing mainly on the glass (74.16% recycled) and aluminium frame (10.30% recycled) with the assumption that the energy input for these processes was calculated as 26% of the total energy required in the manufacturing process [24].

Some articles [e.g., 25] discuss the application of LCA methodology to the innovative process of only recycling PV waste panels. They conclude that the majority of the impacts for the recycling process are related to the transport of PV waste to the site, the plastic incineration processes, and further treatments (including sieving, acid leaching, electrolysis, and neutralization) for the recovery of metals (including silver) from the bottom ash. Environmental benefits of recycling are related not limited to the space in landfills, but also to energy savings, avoided raw materials extraction, and emissions reductions.

The operational life of photovoltaic modules guaranteed by manufacturers is typically 25 years, though there is a tendency to extend this period as their production technology improves. However, practice shows that many users exchange their PV installations before the theoretical end of life, after an average 17 years of use, in order to obtain better energy yields resulting from continuous technology improvements [26]. At the stage of manufacture and assembly, the level of waste is about 2 per cent. On the basis of the installed power in Europe and assumed life expectancy of modules (17 years), the amount of PV waste is estimated to rise to 5,500,000 tons by 2026 [27].

Crystalline PV modules have been the most desirable choice for years and accounted for approximately 85–90% of the global PV market. Si-wafer based PV technology accounted for about 95% of the total production in 2017 [28].

The ecological impact of silicon is high because the metallurgical process for silicon purification has a high energy demand and because of the use of submerged arc and induction furnaces. Some toxic compounds such as chlorine can be emitted in the process [29]. The use of highly harmful substances such as hydrogen fluoride (HF) and hydrogen chloride (HCl) is reduced in PV production. For example, in the US photovoltaic industry the quantity of these chemicals still being used is less than 0.1% of the total amount of chemicals used [5]. Adding pure silicon from the recycling process can decrease the high cost of solar grade silicon production and limit unfavourable impacts to the environment.

Silicon is the most important material recoverable from classic c-Si solar cells [30], though the value of silver increases the profitability of the recycling process [31–32].

The flow of materials in the recycling system for PV modules is shown in Figure 1.

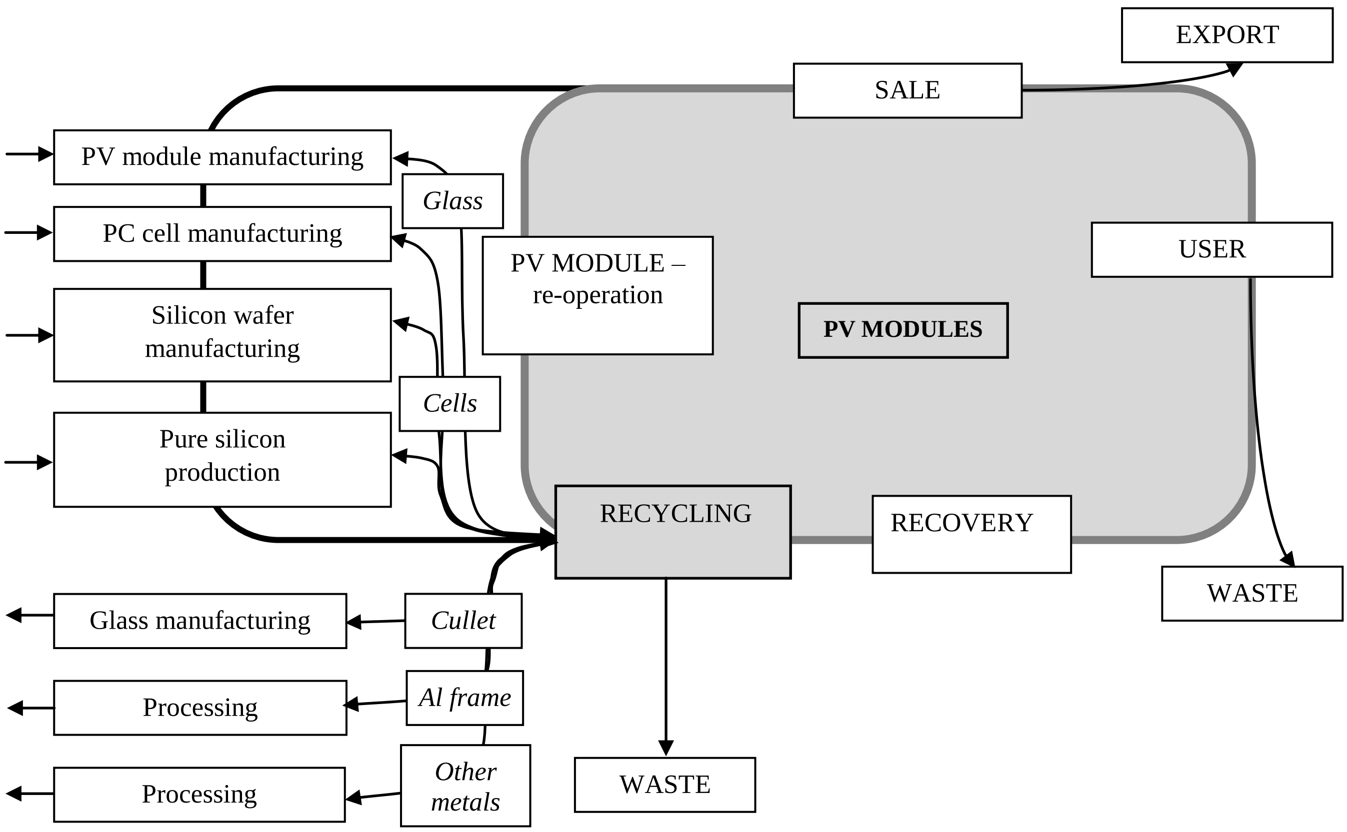

Figure 1. Material flow in the PV module recycling system [26].

Recovered materials, such as aluminium and glass, can be used in PV module manufacturing and also in any other process. Pure silicon is a valuable material and reuse in new cell production would lower the cost and environmental impact of production. This is the reason why the aim of this study is to include the environmental impact of recovering and recycling the semiconductor material from photovoltaic solar cells. Authors compare process of new solar cells production with or without recycled silicon material to confirm this assumption.

For test purposes, spent silicon solar cells made according to different standards were selected. Originally, the PV cells had dimensions of 125×125 mm^2.

The PV recycling process requires two main stages (Figure 2):

- PV solar cell separation: in thermal delamination, the EVA (Ethylene Vinyl Acetate) is removed and materials such as glass, Tedlar®, aluminium frame, steel, copper and plastics are separated;

- cleansing the surface of PV solar cells: unwanted layers (antireflection layer, metal coating and p-n semiconductor) are removed from the silicon solar cells separated from the PV modules; as a result, the silicon substrate, suitable for re-use, can be recovered.

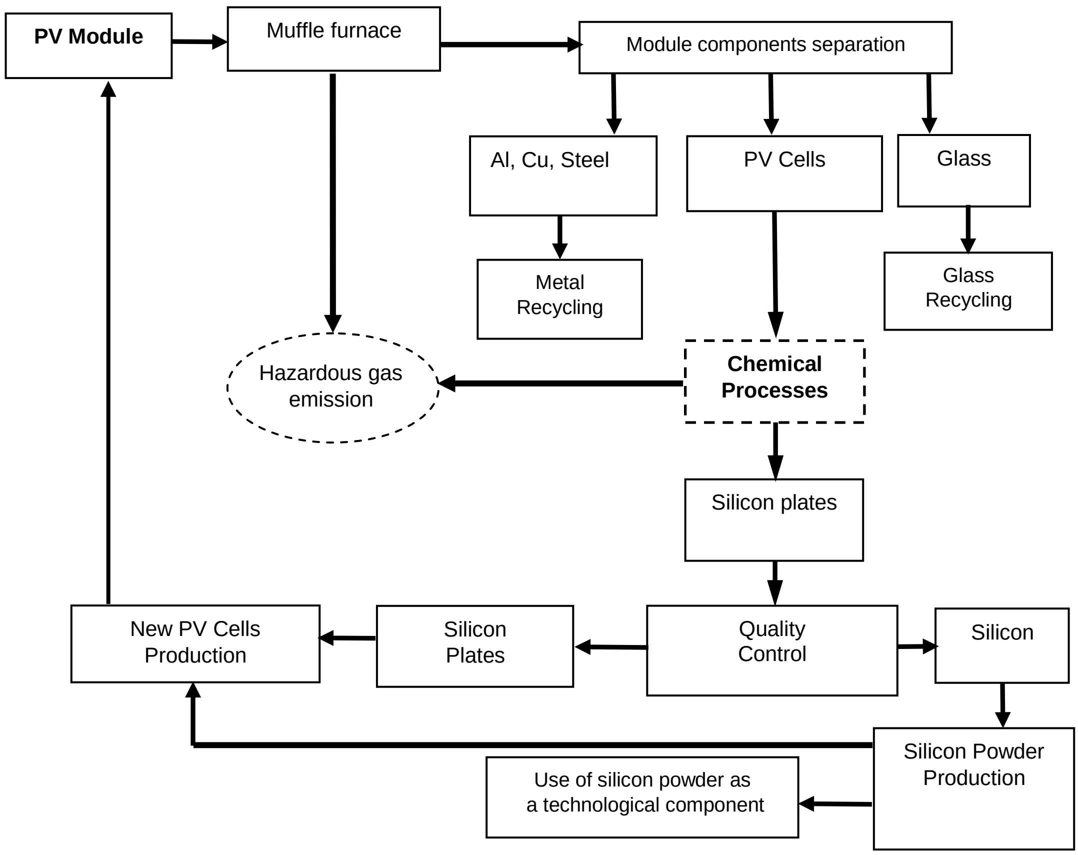

Figure 2. Thermal and chemical processes in PV crystalline cell and PV module recycling [30]

In order to separate silicon photovoltaic cells from a damaged PV module, the module was placed on a SiO_2 bed, which then was heated. After the cells have been separated from PV modules, the various layers of material applied in the production process must be removed in a specific order: front metal coating, bottom metal coating, anti-reflective coating and n-p junction. The chemical process of removing the different layers so that the silicon base could be recovered was optimised and examined.

With the recovered silicon wafers, several processes were used for preparing new silicon solar cells. Before processing, each wafer was laser-cut to dimensions of 50 mm×50 mm; this technique guarantees the quality of the edges.

The suggested technology enables the production of photovoltaic solar cells with conversion efficiencies of 16% for monocrystalline silicon wafers and 13% for multicrystalline silicon. Each of the seven basic steps of the technological process, described in detail below, was carried out with particular emphasis on the physical parameters of the base material and the finished solar cell. The cell manufacturing sequence consists of the following main steps: texturisation, emitter formation, parasitic junction removal, passivation, antireflection coating deposition, front and back contact formation.

Measurements of illuminated current-voltage characteristics for the cells produced were conducted under Standard Test Conditions: AM1.5, irradiance 1000 W/m^2, temperature 25^{o}C. Series and shunt resistance and temperature coefficients were specified as well. The new cells, despite the fact that they have no SiN_x antireflective coating, have a very good efficiency of 13-15% [40].

Methodology

Life cycle analysis (LCA) is a technique to assess environmental aspects and potential impacts associated with all stages of life of products and technologies including: mining and processing of mineral resources, production process, distribution, transport, use, recycling, and final disposal of waste. In the presented analysis, the actual methodology was used, normalization was performed, and the result was obtained according to LCA international standards and the ILCD handbook [33].

The basic standard associated with the assessment of the product life cycle is the ISO standard 14040 Environmental management - Life cycle assessment - Principles and framework. ISO

14040:2006 describes the principles and framework for life cycle assessment, including definition of the goal and scope of the LCA, the life cycle inventory analysis (LCI) phase, the life cycle impact assessment (LCIA) phase, the life cycle interpretation phase, reporting and critical review of the LCA, limitations of the LCA, the relationship between the LCA phases, and conditions for use of value choices and optional elements.

The following standards are included under the 14040 series: ISO 14040 – General Principles and Framework, ISO 14041 – Goal and Scope Definition and Inventory Analysis, ISO 14042 – Life Cycle Impact Assessment (LCIA), ISO 14043 – Life Cycle Interpretation, ISO 14047 – Technical Report, ISO 14048 – LCA Data Documentation Format, and ISO 14049 – Technical Report. ISO 14040 form International Organization for Standardization, 2006, divides LCA into four stages: goal and scope definition, LCI, LCIA, and interpretation (Figure 3).

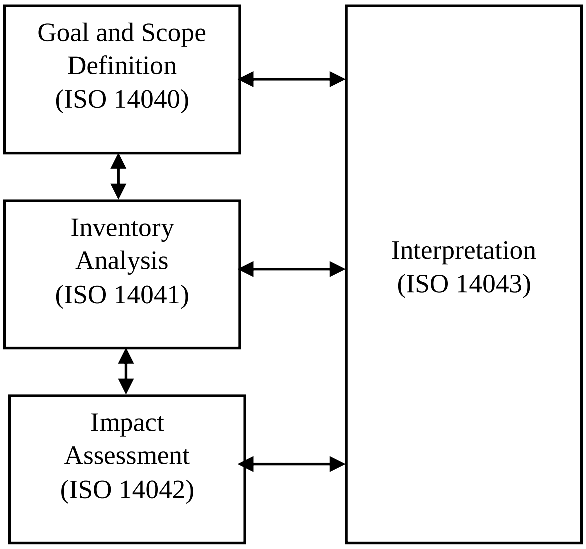

Figure 3. Relationships among LCA stages according to ISO 14040.

A LCIA consists of several elements, the classification, characterization, normalization, and weighting. Classification and characterization are mandatory elements within a LCIA, the normalization and weighting are optional elements (Figure 4).

The third phase of the LCA (characterization) is carried out using various methods, for example, Eco-indicator'99 or ReCiPe, most often implemented with LCA research software (SimaPro, GaBi). The number of available methods for LCA analyses indicates continuous development of the LCA methodology, however, in Europe the most frequently chosen method is Eco-indicator’99. Eco-indicator’99 is an end-point method, originally developed in 1995 to provide design engineers with environmental information in a simple single value format and is intended for internal use.

ReCiPe is a more recent end-point method that transforms inventory results into eighteen mid-point indicators and three end-point indicators that are weighted into a single score.

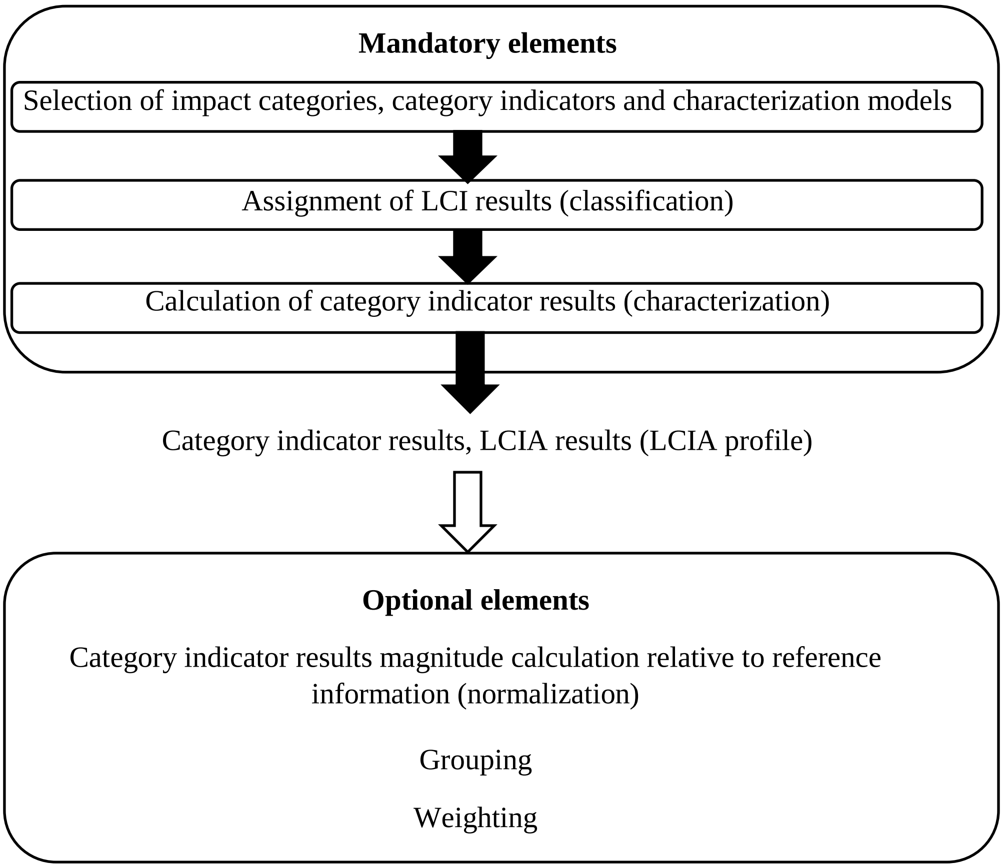

Figure 4. Elements of the LCIA phase (ISO 14040, 2006).

Assessment models of the temporal perspective of ecological effects are presented in Table1.

Table 1. Archetypes of the temporal perspective of ecological effects [34–35].

Model	**Time perspective**	**Management possibilities**	**Evidence**
Individual (I)	Short-term	Technology helps avoid many problems	Only verified
Egalitarian (E)	Long-term	Problems may cause a disaster	All

Hierarchical (H)	Balance between long- and short-term	Good politics help avoid many problems	Consensus between verified and non-verified

The type of model chosen has a large influence on the results (Table 1). The egalitarian method has a long-time perspective and includes even very preliminary scientific knowledge about the environmental impact. This approach is supersensitive to any effect - future or present. This method assumes that nature is fragile and unstable, and provides the worst possible scenario.

A short-term perspective, based on very strong proves and judgment of the environmental consequences, is presented in the individual model. This approach assumes that nature is stable and able to recover from almost any damage, and emphasises the present situation over future gains or losses. Balance between these two types of model is achieved with the hierarchical approach, which assumes that nature is in balance and any damage can be avoided with proper management. These three archetypes are based on an anthropological approach to the human nature of environmental decision-making.To make results comparable to other Europe countries Eco-indicator’99 method was chosen and very often used SimaPro software. The chosen model was hierarchical because of its balanced approach giving the view for long and short term perspective. Because of the silicon recovered from the recycling process usage in manufacturing comparison this approach seems to be the most reasonable.

Assumptions

The preparation of inventory tables for LCA requires the preparation of universal (from the point of view of different producers) data on the consumption of materials and energy during the production of solar modules without taking into account recycling processes and also including the material recycling.

Currently, the dominant semiconductor material used for the production of photovoltaic cells is silicon in the form of mono- or poly- crystalline tiles. By weight, typical c-Si PV panels contain about 76% glass (panel surface), 10% polymer (encapsulant and backsheet foil), 8% aluminium (mostly the frame), 5% silicon (solar cells), 1% copper (interconnectors), and less than 0.1% silver (contact lines) and other metals (mostly tin and lead) [36]. For each type of silicon module production the quantity of particular material is different (Table 2).

Table 2. Inputs in the production of 1m^2 of PV module in 2006 (frameless) [5]

Input	Material	Ribbon-Si	p-Si	m-Si
Components [kg]	Cell material	0.9	1.6	1.5
	Glass	9.1	9.1	9.1
	EVA	1.0	1.0	1.0
	Others	1.8	1.8	1.8
Consumables [kg]	Gases	6.1	2.2	7.8
	Liquid	2.2	6.8	6.6
	Others	0.01	4.3	4.3
Energy media	Electricity [kWh]	182	248	282
	Oil [l]	0.05	0.05	0.05
	Natural gas [MJ]	166	308	361

Also, the energy consumption in 1m^2 production varies according to the type of silicon used, but for mono- and polycrystalline silicon the difference is almost negligible. From the perspective of the recycling process, monocrystalline silicon cells are easier to reuse.

In the production process, solar cells are encapsulated for protection against climatic conditions and mechanical damage. An ethylene-co-vinyl acetate (EVA) copolymer material covers both sides of the cells, while Tedlar® is used on the rear side. The front of PV modules are covered with glass.

Table 3. Material share and recovery rate in the recycling process of silicon solar module [37]

	Glass	Aluminium frame	Solar cells	EVA, Tedlar	Ribbons	Other
Weight [kg]	10.0	1.39	0.47	1.37	0.10	0.16
Share [%]	74.16	10.3	3.48	10.15	0.75	1.16
Recovery rate [%]	90	100	90	-	95	-

An example of innovative recycling process of silicon solar cells is presented in the Figure 5. Amount of chemicals and energy is presented as help to understand importance of recycling process development and used techniques. Table 3 presents typical recovery ate for crystalline silicon module. Both (Table 3 and Figure 5) they give an idea about material and energy consumption and recovery rate in today used techniques.

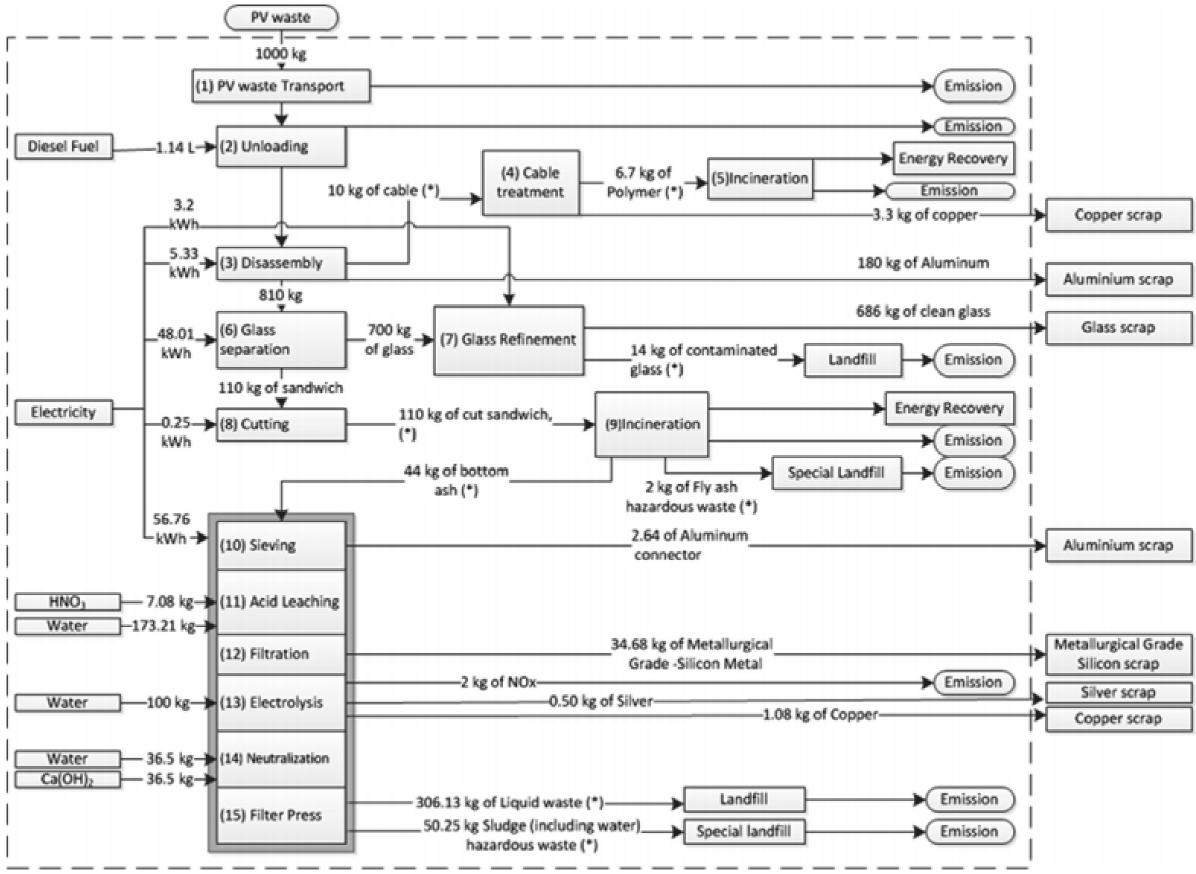

Figure 5. An example of innovative recycling process for crystalline silicon photovoltaic panels [25]

In order to recover silicon as a ‘wafer’ suitable for use as a substrate in the production of new cells, the aluminium frame and junction box should be dismantled manually and the EVA poly laminate layer must first be removed from the module by thermal or chemical processes. Then, with a mixture of alkalis or acids, the anti-reflective layer, the metallization of the front and rear surface of cell, and p-n junction have to be removed.To separate the individual cells, a thermal method was chosen. A thermal process enabling quick, simple, and economically-efficient disassembly of the module is the first stage in PV module recycling. First, the EVA-laminated cells are separated. This recycling process is energy consuming, but since up to 85 % of the recycled cells are reused, energy consumption when manufacturing new PV modules is reduced up to 70 % [38].

Optimal compositions of the mixtures and the design of technological lines for recycling of silicon photovoltaic cells have been registered in the Republic of Poland Patent Office under Patent No. 215 770 [39].

The proposed recycling process aims at separating PV wafers for their potential reuse in new panels [30]. Recovering pure silicon from damaged or end-of-life PV modules can lead to economic and environmental benefits. Cells manufactured from recycled silicon wafers had efficiencies between 15-12% [40]. With reference to cells manufactured with the same technique, in literature efficiency is 16% for monocrystalline and 13% for polycrystalline. It can be state that lost of the efficiency is almost negligible if thickness of recovered wafers is suitable. If not, silicon can be reused as pure powder in monocrystal forming process.

To enable recycling of the silicon base from PV cells, a universal chemical process for removing different layers from the cell surface has been developed. Because of the high quality requirement for the recovered silicon, chemical processing is the most important stage of the recycling process. Etching should be continued until the relevant layers have been removed, although it is essential to avoid too great a loss of silicon. For the silicon base to be suitable for incorporation into new cells, it must not be too thin. A loss of mechanical strength may cause the base to break during the processes performed on its surface. It was assumed that photovoltaic silicon cells have a standard size of 156×156 mm^2 and a thickness of 270-300 μm.

The metal coating should be removed through an etching sequence. The universal recycling procedure for all Si solar cells includes the three main steps: removing metal coatings, e.g. Al and Ag coatings, anti-reflective coatings (ARC), and n-p junctions. During this step, the silver is dissolved. Silver is recovered from the waste acids by electrolysis. From a value standpoint, silver is an expensive component per unit of mass for a c-Si panel – consuming today about 15% (including losses) of the global silver production. The amount of silver that can be recovered from the etching solution is up to 1.6 $g \cdot kg^{-1}$ of broken solar cells [32].

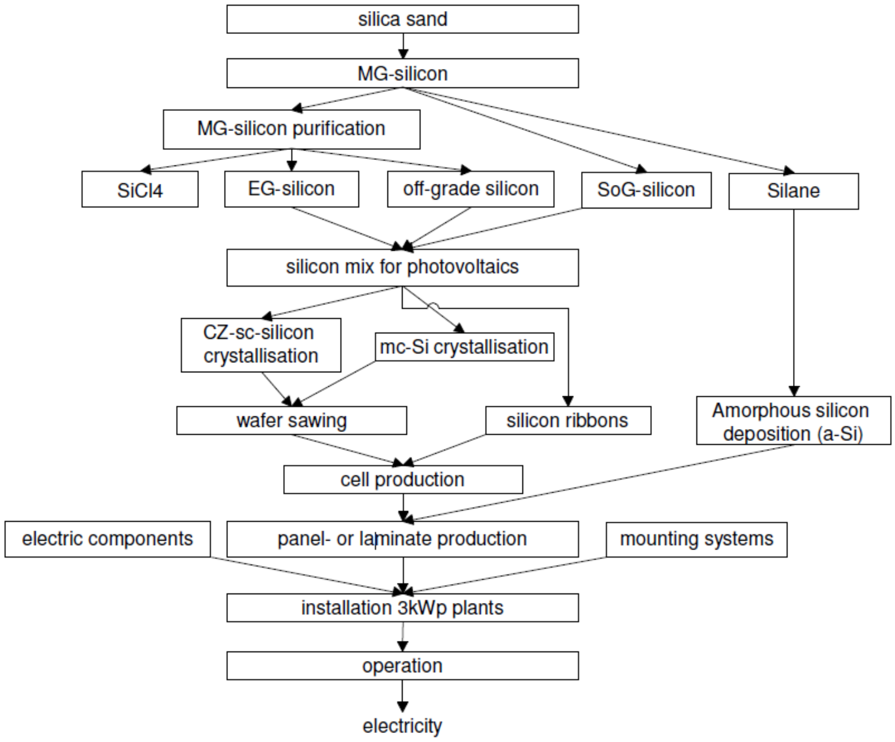

Figure 6 Supply chain of silicon based photovoltaic electricity production. MG-silicon: metallurgical grade silicon; EG-silicon: electronic grade silicon; SoG-silicon: solar-grade silicon; a-Si: amorphous silicon; CZ: Czochralsky; kWp: kilowatt peak [41]

The models proposed for analysis by different authors present in fact a similar approach to the production chain of solar modules, including recycling.

Lunardi et al. presented a process flow diagram for c-Si possible EoL scenarios and the description of each process as a compilation of the best results found by the authors [42].

Development of new material-saving technologies is expected in the coming years. By 2030 the glass content of c-Si panels is predicted to increase by 4% to a total of 80% of a panel's weight. The main material savings will include a reduction in silicon from 5% down to 3%, a 1% decrease in aluminium, and a very slight reduction of 0.01% in other metals. A reduction in the quantity of silver used is a clear target [43]. Specifically, silver consumption is expected to further decrease through adoption of improved metallization processes [44]. However, at present and for the next two decades, recycling will include solar modules produced in recent years.

The basic environmental benefits of the proposed photovoltaic module recycling technology result from the use of recycled semiconductor material. The repeated use of silicon, which has been purified in the previous cycle of cell production (Figure 6.), will significantly reduce energy consumption. The quantities of energy required in various processes during the fabrication of PV modules are listed in the Table 4. Additionally etching solutions can be reused to decrease environmental impact of recycling process. New etching methods without solvents are studied to make recovery of materials environmental friendly as much as possible. Benefits for the environment are major compared to the alternative solutions such as PV waste incineration.

SimaPro was used for the LCA. SimaPro is a professional tool for collecting, analysing, and monitoring sustainability performance data of products and services.

Table 4. Energy requirement in different processes during the production of PV modules [45].

Process	Energy requirements
Silicon purification and processing	
- Czochralski Silicon (Cz-Si) production from EG-Si	290 kWh kg^{-1} of EG-Si
- Electronic grade silicon (EG-Si) production form MG-Si	100 kWh kg^{-1} of EG-Si
- Metallurgical grade silicon (MG-Si) production from silicon dioxide (quartz, sand)	20 kWh kg^{-1} of MG-Si
Fabrication of solar cell	120 kWh m^{-2} of silicon cell
Assembly of PV module	190 kWh m^{-2} of PV module
Roof top integrated PV system	200 kWh m^{-2} of PV module

However, the data from Ecoinvent base all reflects European averages, except for the end of life waste treatment, which was based on the situation in Switzerland.

Results

This section dissects the environmental impacts calculated in the LCA carried out here. The main environmental impacts of two possible end-of-life scenarios were analysed: i) disposal and ii) recycling of a panel with the recycled semiconductor base reused.

The environmental impacts associated with the production steps of PV modules remain the same for both scenarios, and therefore this study mainly focused on analyzing traditional cell production with new raw materials versus production that incorporates recycled silicon material. Other differences of installations, such as building integrated vs. free-standing systems, as well as those associated with PV module orientation, direction, and performance of the balance of system (the balance of system (BOS) encompasses all components of a photovoltaic system other than the photovoltaic panels) all have notable effects on the LCA results.

Table 5 presents results of the comparative analysis of environmental impacts for characterisation per impact category carried out using Eco-indicator'99 method.

Table 5. LCA characterisation results for impact category

Impact category	**Unit**	**c-Si cells at plant**	**c-Si cells using recycled materials**
Carcinogens	DALY	4.2E-5	6.55E-6
Respiratory organics	DALY	1.04E-7	8.02E-8
Respiratory inorganics	DALY	8.9E-5	3.6E-5
Climate Change	DALY	5.25E-5	3.1E-5
Radiation	DALY	1.88E-6	1.72E-7
Ozone layer	DALY	3.33E-8	2.86E-8
Ecotoxicity	$PAF \cdot m^2 year$	31.4	5.96
Acidification/Eutrophication	$PDF \cdot m^2 year$	2.0	1.56
Land use	$PDF \cdot m^2 year$	1.39	0.823
Minerals	MJ surplus	1.44	0.71
Fossil Fuels	MJ surplus	272	185

The results of the comparative analysis of environmental impacts of electronic grade silicon production with and without the use of recycled material are presented in the Figure 7. This figure presents the normalization results.

Table 6. presents the final LCA analysis results in points after weighting for producing 1 kg of c-Si with all new material in comparison to using recycled material.

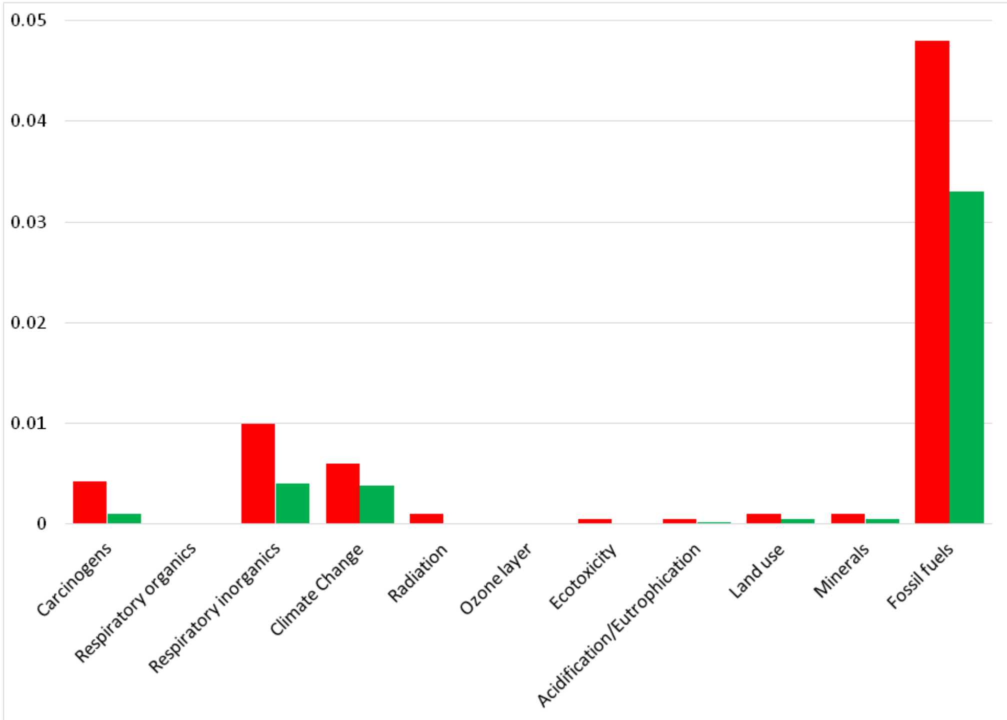

Figure 7. The results of the normalization for the production of silicon solar cell without (red bars) and with the use of recycled silicon (green bars).

Table 6. LCA results for producing 1 kg of c-Si at plant in comparison to the use of recycled material.

Impact category	c-Si cells at plant (Pt)	c-Si cells using recycled materials (Pt)
Carcinogens	0.0757	0.0636
Respiratory organics	0.00165	0.000674
Respiratory inorganics	0.66	0.339
Climate Change	0.603	0.242
Radiation	0.0016	0.00148
Ozone layer	0.000611	0.000406
Ecotoxicity	0.0226	0.0183
Acidification/Eutrophication	0.0829	0.037
Land use	0.0421	0.03
Minerals	0.0139	0.0107
Fossil fuels	3.82	1.49
Total	**5.33**	**2.23**

To achieve environmental benefits, the environmental impact of the recycling process must be smaller than the environmental impact of the material production process. Others have demonstrated that the environmental impact (expressed in environmental points – Pt) of transportation and recycling stages are of little significance in terms of the whole lifecycle [45].

Conclusions

Recycling helps to reduce the consumption of valuable raw materials, production costs, and environmental impacts. An important argument for the recycling of photovoltaic modules is the reduced energy consumption at the production stage through the use of existing purified material. All stages of the silicon cell life cycle contribute to the GWP and reduction of greenhouse gas emissions through the use of recycled silicon material represents 42%. The total environmental impact of PV production can be reduced by as much as 58%, primarily due to reduced energy consumption during the production of high purity crystalline silicon.

In the analysed recycling process, after separation of the cells from the PV module, several processes were carried out that led to the recovery of the base material, i.e., silicon wafers. These silicon wafers were examined to determine whether any inclusions remained. Following the appropriate preparation, new photovoltaic cells were produced from the recovered wafers. This approach allows for a maximum reduction of photovoltaic panel impacts on the environment through re-use of all valuable materials, including silicon substrates.

To achieve environmental benefits, the environmental impact of the recycling process must be smaller than the environmental impact of the material production process. Although the cost of storage module landfill is smaller, recycling must be economically viable. The LCA of c-Si PV cell production with and without recycling of spent semiconductor material demonstrates that the negative environmental impact of photovoltaic cell production with recycled material was nearly two times lower than the environmental impact associated with producing cells from primary materials.

The biggest difference was seen in the fossil fuels category and a relatively small difference was observed in the radiation category. Photovoltaic panel production involves the use of many chemical substances and emissions, which are not environmentally neutral. We cannot consider photovoltaics as a zero-emission technology. It must be emphasized that the coal-based energy sector is one of the least environmentally friendly. That is why the comparative analysis with clean energy mixes may indicate even lower environmental efficiency of PV cells.

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