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Environmental performance of Kesterite monograin module production in comparison to thin-film technology

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ABSTRACT

Kesterite-based structures are being extensively studied for solar module productions due to their earth abundant and nontoxic nature, high absorption coefficient, and a wide variety of scalable deposition methods. Kesterites are mostly manufactured using thin-film technology. However, in the last decade, the monograin approach has gained further attention, providing a third alternative to mono-crystalline wafer and thin film methods. This is due to its high throughput, low-cost deposition techniques, flexibility, and light weight. Despite the technical advancements in the monograin technology, their environmental impacts have not been studied in the literature. This paper, for the first time, presents a cradle to gate environmental life cycle assessment of CZTS monograin module production. The analysis is designed to identify the environmental hotspots associated with materials, energy usage, and manufacturing processes. The results were compared to CZTS thin-film and the commercially available CIGS technologies. The analyses suggested that the front contact accounted for the majority of impact in all categories due to the use of silver. The normalisation results showed that the marine aquatic ecotoxicity impact category dominated the overall impact results. A comparison of CZTS monograin and thin film production demonstrated that monograin outperformed the thin film technology when silver was substituted with alternative materials and was proximate to CIGS even considering their higher achieved efficiency. The analysis presents considerable environmental benefits associated with the monograin technology. Further savings in emissions could be achieved with improved conversion efficiency and usage of renewable energy sources in the manufacturing stages.

1. Introduction

Currently, climate change and energy supply security are two of the most global pressing issues. Photovoltaic technology can meet a considerable portion of the world's energy needs, however, the impact from these devices need to be reduced to avoid environmental burden shifting from one phase to another. The photovoltaic industry currently relies mostly on single-crystal and polycrystalline silicon wafers (roughly 95%), while thin-film technology accounts for only 5% of the global photovoltaic module market [1]. The most commonly used thin-film solar cell, cadmium telluride (CdTe) and copper indium gallium diselenide (CIGS), contain resource-limited elements (Te, Ga, and In). These materials are up to ten times more expensive than other metals, posing limitations for future very large scale applications. To realise the benefits of this technology in the future, new unconventional solar cell materials are required that are earth-abundant, nontoxic, and much less expensive to produce lowering the overall cost of solar photovoltaics and reducing their environmental burden [2–5].

Kesterite materials Cu_2ZnSnS_4, $Cu_2ZnSnSe_4$, and their mixtures $Cu_2ZnSn(SSe)_4$ (CZTS) are promising candidates that are being extensively studied for solar module production. They are considered as the most promising next generation solar module materials. This is due to their earth abundant, low price and non-toxic nature, a near optimum direct bandgap energy of 1.0~1.6 eV, and high absorption coefficient (>104 cm^{-1}) [6,7]. The highest reported efficiency until now is 12.6% [8,9] while the theoretical power conversion efficiency (PCE) limit reaches 32% [7]. In addition, CZTS provides a wide range of scalable deposition methods, with different architectures for various types of

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solar modules [10].

Kesterite solar modules are primarily manufactured using thin-film technology [11]. The monograin approach however, in the last decade gained further attention for solar module production, providing a third alternative to mono-crystalline wafer and thin film methods [5]. The monograin membrane approach combines the advantages of high throughput, low-cost deposition techniques primarily from the printing industry with the versatility of a flexible, lightweight, thin film solar module. In general, powder technologies are the least expensive methods of creating materials [12,13].

In this context, studies [5,6,14,15] showed that the production of CZTS powders with an improved crystal structure is comparatively simple, inexpensive, and convenient. This enables significantly more cost-effective and energy efficient material production with negligible waste flows in the manufacturing process.

Several studies have assessed the advancement in the technical performance of CZTS monograin modules in several ways. For example [6], have studied methods to decrease interface recombination of the modules [14,16,17]. have assessed the powder production of CZTS and CTZSe via isothermal crystallisation from initial binary compound precursors in molten potassium iodide. Further analysis was performed to test the efficiency of CZTS monograin modules [18]. Other studies presented by Ref. [19] have examined the improvement in CZTS efficiency through changes in thermal temperature treatment, the influence of order-disorder, Cu–Zn disordering of crystals, and changes in band gap [20]. demonstrated that utilising an oxidative chemical treatment prior to the formation of the heterojunction enhanced the performance of CZTS monograin modules by up to 9.4%. The phenomenon was attributed to the creation of a SnO_2 passivation layer on the surface of the CZTS. Based on a numerical simulation [21], proposed a potential high efficiency Kesterite solar module with multiple CZTS layers and efficient band offset alignment. The performance of CZTS monograin have reached PCE of up to 10.17 % (certified by Freiburg ISE CalLab [22]).

CZTS has demonstrated considerable long-term advantages due to its non-toxic and abundant nature especially when compared to other types of chalcogenide solar modules, such as CIGS and CdTe, although its efficiency is still extensively lower. One of the primary causes for CZTS devices' comparatively lower efficiency (e.g., as compared to CIGS) is a lack of open circuit voltage (VOC). This drop in VOC is expected to be due to material defects [19,23,24]. Recently, a VOC of 784 mV was reached using a CZTS monograin layer by carefully regulating the annealing and the Cu–Zn ordering [13]. Despite the fact that all existing studies have made considerable progress, the primary challenges have remained.

Limited studies [25–29] have assessed the environmental performance of CZTS solar modules in general. Despite significant technical advancements in monograin technology, to date, a comprehensive Life Cycle Assessment (LCA) of the technology has not been reported to the best of the authors' knowledge. This paper, for the first time, presents a comprehensive cradle to gate LCA of CZTS monograin module to identify the environmental hotspots associated with materials, energy usage, and manufacturing processes. The results were compared to CZTS thin-film and the commercially available CIGS solar module technologies.

2. Life cycle assessment methodology

LCA is a methodology that systematically assists in qualifying material and energy flows, as well as the environmental impacts created by products and services throughout their life cycle from raw material extraction to disposal or recycling [30,31]. This method consists of four major phases: goal and scope definition, life cycle inventory, life cycle impact assessment, and results interpretation. These phases should include all of the inputs and outputs necessary to complete the LCA properly. The LCA performed in this paper considered all phases from raw material extraction to the production gate, following a cradle to gate system boundary. This was mainly due to two reasons: 1) the majority of environmental impact is associated with the manufacturing stage of solar modules [28]; and 2) the uncertainties associated with the use and end of life phases due to the emerging nature of the technology. Moreover, there is currently no recycling process dedicated to the disposal of kesterite panels. The present LCA study was carried out in accordance with the international standards ISO 14040 [32] and ISO 14044 [33].

2.1. Goal and scope definition

The environmental impact of producing CZTS monograin modules was evaluated in this study. The material composition used in the analysis is based on a CZTSSe kesterite configuration but referred to as CZTS in the following sections. The LCA study included materials for both the substrate and active layers of the solar module, as well as process auxiliaries such as water and electricity. Inventory data used to calculate environmental impact results were compiled using manufacturers process data, with the impact assessment performed using GaBi ts 9.2 LCA software [34]. GaBi has been extensively used to conduct academic LCA research showing adequate competency [35,36]. The functional unit (FU) selected for this study was 1 kWh of generated electricity over the whole lifetime of the module. This FU provided a common basis for comparison with other more established thin film solar modules due to the difference in operating parameters. The FU was also used as a reference flow.

The assessment considered a cradle-to-gate approach, accounting for all impacts from raw material extraction till the production gate. This was dictated by the emerging nature of the technology, and a lack of sufficient information about its use and end of life phases. Appropriate recycling techniques need to be developed to provide a more comprehensive representation of the environmental impact of the technologies under investigation [37–39]. An efficiency of 10.17% was subsequently used to determine the module area required to attain the functional unit (1 kWh). This was calculated as 5 cm^2 according to the equation below:

$$A = \frac{Lifetime\ Output\ (kWh)}{SI\ *\ PR\ *\ E\ *\ LT}$$

where,

A = Area (m^2)
SI = Solar radiation (kWh/m^2)
PR = Performance ratio (%)
E = Efficiency (%)
LT = Lifetime (yr)

The United Kingdom is taken as the geographical location for this study, with an average solar radiation of 850 kWh/m^2 [40]. The sensitivity of the results to this choice was further analysed in section 3.4. Operational lifetime was set to 30 years and performance ratio, defined as the ratio of actual electricity generated to theoretical expected values, was assumed to be 80% in compliance with the international energy agency guidelines [41]. Fig. 1 shows the system boundary used for the LCA study. It indicates the different process steps involved in the manufacturing of the CZTS monograin module, categorised into the production of CZTS semiconductor powder and the roll-to-roll processing of the module.

The monograin module for the current research was prepared by Crystalsol, a European-based Company that develops novel types of flexible photovoltaic technologies. It is formed by $Cu_2ZnSn(SSe)_4$ monograins synthesized in the sealed quartz ampoules at 740 °C from high purity (5 N) Cu, Sn, ZnS, Se and S precursors in molten KI as a flux material [42]. The salt is required to enable the formation of CZTS powder grains as it acts as a solvent for the metal ions. The monograin treatments after synthesis included the following process steps: removal

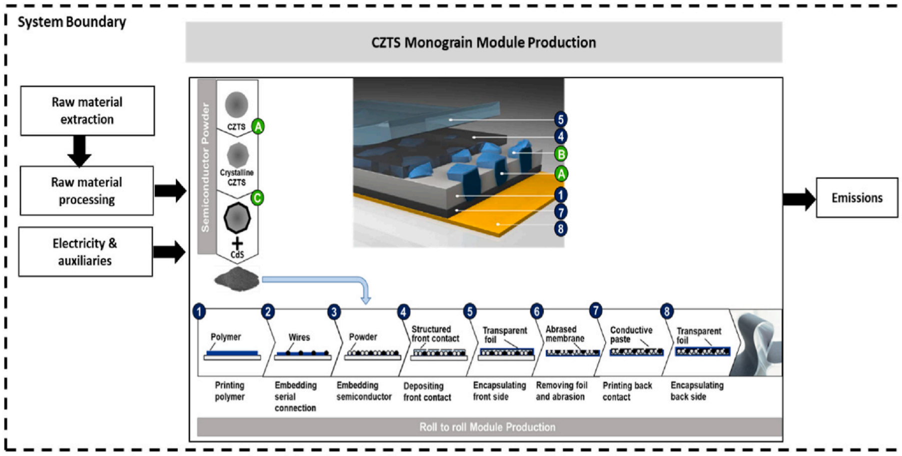

Fig. 1. System boundary considered for the LCA of CZTS monograin module production.

of the flux with water; etching the CZTS grains with 0.2 % Br_2 in methanol and 2% KCN in 0.2% KOH water solution to remove secondary phases and impurities from the surface; followed by an additional short-time thermal treatment at 740 °C to have homogeneously ordered p-type CZTS with perfect tetrahedral shaped crystals [43]. A CdS buffer layer of about 30 nm thick is then deposited around the crystals in a chemical bath deposition technique. This buffer layer is needed for the formation of the p-n junction and to enable the crystals to be transported in open air without complications [43]. After final drying, the CZTS/CdS monograins are sieved to separate the grains in a size range between 16 and 100 μm to several narrower fractions with grain diameter difference of 10–20 μm for the good quality monograin membrane making.

After the CZTS powder is manufactured and transported to the assembly site, the second step commences, involving the roll-to-roll assembly of the CZTS monograin module beginning with the printing of a thin polymer layer. This step also involves embedding the metal wires for the cell series connection and the produced CZTS powder within the roll-to-roll production (as presented in step 3 of Fig. 1). At this stage, the CZTS crystals can function as mini solar modules as p-type CZTS crystals have already been coated with n-type buffer layer. The resulting deposited CZTS active layer has a thickness of 0.045 mm. Mechanical abrasion occurs afterwards before the front contact is placed, with the back contact layered subsequently. Flexible non encapsulated modules are produced for use in buildings and other applications. Encapsulation is needed if the modules are to be placed outside [43].

2.2. Life cycle inventory

Life cycle inventory involves an assessment of all materials and energy inputs and outputs of all process steps involved in the production of the assessed CZTS monograin module. The data used to compile the inventory were obtained from the manufacturer and when not available from literature. As mentioned previously, the CZTS monograin module production consist of two steps, the CZTS semiconductor powder production and the roll to roll processing step, where the powder is embedded into the module. Table 1 shows the inventory for the CZTS monograin production including embedding the produced powder into the solar module. Energy consumption data for all production stages were assumed to be from the average of the 28 countries in the European Union (EU) from the database of GaBi. Chemicals required for this step when not available in GaBi were synthesized from literature assuming the reaction occurring at 100% efficiency. As the energy needed for these synthesis was minimal compared to the total energy requirements, it was not added to the overall energy load. The full comprehensive life cycle inventory is in the supplementary materials (see Table SM.1).

Table 1
Life cycle inventory of the CZTS monograin production process including the powder production step embedded in the roll-to-roll process capable of producing 1 kWh of energy (Compiled by authors using manufacturer's data).

S/N	Processing Steps	Energy and Material inventory
1	Printing polymer	Electricity, Epoxy resin, Polyethylene terephthalate (PET) foil
2	Embedding serial connection	Wire (Cu–Ag), Nylon thread
3	Embedding semiconductor (the CZTS powder produced in earlier stages)	Production of CZTS powder: Electricity, Copper, Potassium Iodide, Sulfur, Tin, Zinc Sulphide, Selenium, Nitrogen Post treatment of CZTS Powder: Electricity, Nitric Acid, Bromine, Methanol, Potassium Cyanide, Potassium Hydroxide, Addition of buffer layer: Electricity, Ammonia, Thiourea, Cadmium Acetate
4	Depositing front contact	Electricity, intrinsic zinc oxide (i-ZnO) target, Aluminum-doped Zinc Oxide (AZO) target, Ag nano wires
5	Front side stabilization	Electricity, Fiber glass sheet, Epoxy resin
6	Removing foil and abrasion membrane	Aluminium polishing paste, polish wool merinowool
7	Printing back contact	Electricity, graphite paste, silver paste
8	Encapsulation	Ethylene tetrafluoroethylene (ETFE), PET, Epoxy resin, Polyvinyl fluoride, Aluminium, Silicone

2.3. Life cycle impact assessment

Life cycle impact assessment results are presented using a selection of thirteen impact categories based on the CML 2001 impact characterisation [44] and cumulative energy demand methods [45] (Table 2). CML 2001 method is a widely used impact characterization method for analysing the environmental impacts of solar modules considering a wide range of metals and the depletion of critical resources used in the production processes [27]. Renewable energy technologies such as solar modules are manufactured for green energy generation, therefore alongside the selected impact categories, the Energy Pay-Back Time (EPBT) as a key environmental parameter is analysed. It is defined as the time needed for the technology to produce the same quantity of energy used in its manufacture [46]. EPBT can be obtained from the total embodied energy and energy output of the module as shown in the equation below;

$$EPBT = \frac{E_{EMB}}{E_{GEN}}$$

Table 2
Life cycle Impact categories analysed with the CML 2001 and cumulative energy demand methods (Dreyer et al., 2003, Frischknecht et al., 2015).

Category	Abbreviation	Unit	Method
Abiotic depletion	ADP	kg Sb eq	CML 2001
Abiotic depletion (fossil fuels)	ADPF	MJ	
Global warming	GWP	kg CO_2 eq	
Ozone layer depletion	ODP	kg CFC-11 eq	
Human toxicity	HT	kg 1,4-DB eq	
Fresh water aquatic ecotoxicity	FWE	kg 1,4-DB eq	
Marine aquatic ecotoxicity	MAE	kg 1,4-DB eq	
Terrestrial ecotoxicity	TE	kg 1,4-DB eq	
Photochemical oxidation	POP	kg C_2H_4 eq	
Acidification	AP	kg SO_2 eq	
Eutrophication	EP	kg PO_4 eq	
Primary energy non-renewable resource	PENRT	MJ	Cumulative energy demand
Primary energy renewable resource	PERT	MJ	

where,

E_{EMB} = Total embodied energy of the module (MJ)
E_{GEN} = Annual energy generated by the module (MJ/yr)

The selected environmental impact categories are not directly comparable to one another due to their different units (Table 3). Therefore, a normalisation step is carried out, where the different characterised impact scores are related to a common reference, to give the environmental impact results more context and to provide a common basis for comparison across impact categories. This helps to better understand the relative magnitude of each of the environmental impact categories, and enables researchers to compare and combine them to estimate a single "total" impact score. This can be calculated using normalisation techniques built into existing life cycle impact assessment methods such as CML (European method), TRACI 2.1 (US method), and ReCiPe (global method). Normalisation involves dividing the results of impact categories by a reference value (e.g. the total impact for a given geographical region or reference year) [47]. In this study, the normalisation factors shown in Table 4 for the CML 2001–jan 2016, EU25 + 3 factors from the GaBi database have been used.

The limitation of this approach is that different results and interpretations can be found depending on the normalisation method and factors used, which are region-specific. Moreover, it is difficult to interpret the absolute values from the normalisation work (Celik et al., 2016). In order to overcome this limitation, in this study, CIGS solar module is used as a reference point which allows for a direct comparison between technologies. To aid this comparison we also normalised the results of CIGS for each of the selected impact categories. The results of CIGS and CZTS thin-film technologies were obtained from the study of Resalati et al. [27] in order to provide a common basis for comparison based on a CML 2001 impact characterisation method.

Table 3
Normalisation factors for the impact assessment used in the CML 2001 characterisation method.

S/N	Category	Abbreviation	Normalisation Factors
1	Abiotic depletion	ADP	6.20E-09
2	Abiotic depletion (fossil fuels)	ADPF	2.85E-14
3	Global warming	GWP	1.92E-13
4	Ozone layer depletion	ODP	9.80E-08
5	Human toxicity	HT	2.00E-12
6	Fresh water aquatic ecotoxicity	FWE	4.78E-12
7	Marine aquatic ecotoxicity	MAE	2.23E-14
8	Terrestrial ecotoxicity	TE	8.65E-12
9	Photochemical oxidation	POP	5.79E-10
10	Acidification	AP	5.95E-11
11	Eutrophication	EP	5.41E-11

2.4. Scenario definitions and sensitivity analysis

Four scenarios were considered in addition to the baseline case described above (Table 4). These scenarios involved changes to energy supply, efficiency, geographical location, and material use in the deposition of the front contact. Scenario 1 (S1) involves changing energy supply from the use of more fossil based fuel (EU28 electricity grid mix) to renewables (solar modules) from the database of GaBi (ts 9.2). Scenario 2 (S2) assumes that the geographical location of the study was shifted from the United Kingdom to the Sahara Desert as a reference location where a higher solar insolation (2190 kWh/m^2) is available [48]. Scenario 3 (S3) assumes a change in the materials used in the front contact, mainly for the use of silver. Silver is originally used as part of the materials in the front contact as nanowires and in the back contact as silver-paste and is a costly material with high environmental impact [49, 50]. This necessitated the manufacturer to shift away from the use of silver to achieve scalability because of its costly nature and environmental impact (Inventory data for the developed CZTS monograin module can be found in the supplementary materials Table SM3). Scenario 4 (S4) is similar to S3 but considers additionally the change in the electricity mix to renewables (solar modules). The modelling of scenarios S3 and S4 using GaBi considered that all process steps for the roll-to-roll processing of the solar module remain the same except for a change in materials used in the front contact.

3. Results and discussion

This section presents a breakdown of the environmental impact assessment of manufacturing CZTS monograin modules categorised into the two primary stages of the semiconductor powder production and the roll-to-roll processing of the solar modules. The manufacturing stages as well as the materials and energy contributing to the dominant impact categories are identified for each step in order to assist the manufacturers in exploiting the results of the analyses.

The results associated with the CZTS powder production are presented in Section 3.1 due to its significance and dominance of energy and material requirements. This step is however embedded in the roll-to-roll production (step 3 of Fig. 1), presented in Section 3.2 and in the later sections. The CZTS monograin module production is used in this study to represent the semiconductor powder production and the roll-to-roll process as a whole.

Section 3.3 presents the energy payback time based on total energy use for the CZTS monograin module production. A sensitivity analysis, presented in Section 3.4, is also conducted to assess the environmental impacts of key parameters and assumptions to identify alternative scenarios and solutions. The results of the CZTS monograin production technique as well as the best alternative scenario are then compared with CZTS thin-film and the commercially available CIGS technology as presented in Section 3.5.

3.1. Environmental impact of CZTS semiconductor powder production

Table 5 demonstrates the environmental impact of the three production steps for the manufacturing of CZTS semiconductor powder. The initial production of the CZTS powder accounted for the majority of the impacts mainly due to consuming 98% of the overall energy demand required to produce the CZTS semiconductor powder.

The environmental profile of materials used indicate that alongside copper needed for the synthesis of the powder, bromine used in the post

Table 4
Different scenarios analysed for the life cycle assessment of CZTS monograin module.

Scenarios	S0	S1	S2	S3	S4
Description	Standard	Electricity supplied from renewable sources	Different geographical location	Different material composition used for depositing the front contact	Different material composition used for depositing the front contact in addition to Electricity supplied from renewable sources
Solar radiation (kWh/ m^2)	850	850	2190	850	850
Performance ratio (%)	0.8	0.8	0.8	0.8	0.8
Efficiency (%)	10.17%	10.17%	10.17%	10.17%	10.17%
Lifetime (yr)	30	30	30	30	30
Electricity Source (Extracted from GaBi ts 9.2 database)	EU-28 electricity grid mix	Solar modules (EU-28 technology mix of CIS, CdTE, mono and multi crystalline)	EU-28 electricity grid mix	EU-28 electricity grid mix	Solar modules (EU-28 technology mix of CIS, CdTE, mono and multi crystalline)
Materials usage in the deposition of front contact	Zinc oxide, Aluminium-Zinc oxide, Silver	Zinc oxide, Aluminium-Zinc oxide, Silver	Zinc oxide, Aluminium-Zinc oxide, Silver	Zinc oxide, Dopant salt, Citric acid, and ZnO NP ethanol dispersion (2.5 wt%)	Zinc oxide, Dopant salt, Citric acid, and ZnO NP ethanol dispersion (2.5 wt%)

Table 5
Life cycle impact assessment results for the production of CZTS semiconductor powder (impact per kWh).

Impact Category	Unit	Production of CZTS Powder		Post treatment of CZTS Powder		Addition of buffer layer	
		Value	Percentage Contribution	Value	Percentage Contribution	Value	Percentage Contribution
ADP	kg Sb eq	5.94E-07	93.00%	2.40E-08	3.76%	2.07E-08	3.24%
ADPF	MJ	7.38E-02	55.42%	5.76E-02	43.27%	1.75E-03	1.31%
GWP	kg CO_2 eq	6.52E-03	59.06%	4.45E-03	40.31%	6.93E-05	0.63%
ODP	kg CFC-11 eq	2.37E-16	93.19%	1.61E-17	6.32%	1.25E-18	0.49%
HT	kg 1,4-DB eq	4.68E-04	81.62%	1.02E-04	17.86%	3.00E-06	0.52%
FWE	kg 1,4-DB eq	1.91E-05	55.04%	1.54E-05	44.51%	1.56E-07	0.45%
MAE	kg 1,4-DB eq	8.38E-01	88.17%	1.07E-01	11.30%	5.06E-03	0.53%
TE	kg 1,4-DB eq	1.37E-05	76.76%	4.01E-06	22.49%	1.32E-07	0.74%
POP	kg C_2H_4 eq	1.02E-06	48.96%	1.05E-06	50.55%	1.04E-08	0.50%
AP	kg SO_2 eq	1.45E-05	50.31%	1.42E-05	49.34%	9.88E-08	0.34%
EP	kg PO_4 eq	1.77E-06	49.77%	1.77E-06	49.67%	2.01E-08	0.56%
PENRT	MJ	1.17E-01	65.08%	6.07E-02	33.79%	2.03E-03	1.13%
PERT	MJ	5.06E-02	91.81%	4.19E-03	7.60%	3.26E-04	0.59%

treatment step was seen to be contributing significantly to the associated impacts. The use of these materials in higher quantities, compared to the other materials, contributes to their elevated environmental impact. Copper contributed significantly to the ozone layer depletion (ODP) and terrestrial ecotoxicity (TE) impact categories while bromine was a major contributor to global warming (GWP). The environmental impact of copper could be potentially reduced using reclaimed copper which was found to have lower impact on the environment compared with refined copper [51]. Bromine, which is mainly used to produce brominated flame retardants is known to be toxic to humans when inhaled and poses a hazard to the environment when released [52,53]. This necessitated recommendations that brominated flame retardants be substituted with alternative halogen-free flame retardants due to their higher environmental impact [54]. Bromine is a critical chemical in the CZTS semiconductor powder production, therefore occupational health and safety best practices should be observed to limit workers' exposure and atmospheric release. The addition of the buffer layer was insignificant when calculating the environmental profile of the CZTS semiconductor powder production. However, due to the toxic nature of CdS it is recommended that alternative non-toxic buffer layer such as zinc sulphide could be adopted [55].

3.2. Environmental impact of CZTS monograin module production

The analysis in this section covers the environmental impact associated with the CZTS monograin module manufacturing stages, alongside the disaggregation of these processes into materials and energy contributions. In addition, a further analysis of the role of individual materials in the overall environmental performance of the monograin technique was considered in the following sections. The results were normalised to provide a comprehensive breakdown of the impacts for further analysis.

3.2.1. The environmental impact associated with manufacturing stages

The percentage contribution of each process step to the impact categories for the roll to roll production of a conventional CZTS monograin module is demonstrated in Fig. 2. A comparison of the processing steps showed that depositing the front contact contributed to the majority of assessed impact categories. The highest contribution (about 80%) was observed for the abiotic depletion (ADP) impact category. The front contact also contributed more than 54% and 48% to freshwater ecotoxicity (FWE) and human toxicity (HT), respectively. The main impact from ADP is that acidifying substances cause a wide range of impacts on

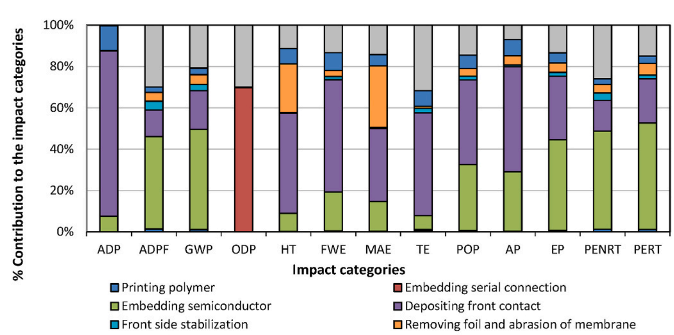

Fig. 2. Contribution of each process step to impact categories for the roll to roll production of CZTS monograin module.

soil, groundwater, surface water, organisms, ecosystems, and materials while FWE examines how emissions of chemicals that are hazardous to air, water, and soil affect fresh water and ecosystems. The main concern of the HT impact category are the effects of toxic substances on the human environment [47]. Embedding semi-conductor (which includes the CZTS powder production) also had a significant impact on different impact categories, the highest contribution was observed for primary energy renewable resources (PERT) (51.4%) and global warming potential (GWP) (48.5%). Embedding serial connection was the major contributor to the ozone depletion (ODP) category (69.7%) mainly due to the use of copper wires, while printing polymer and front side stabilization steps had the lowest contribution (<10%) to all impact categories. Copper offers higher relative electrical conductivity hence its wide use as electrical wires over other options such as aluminium. Aluminium wires however, offer lower environmental impact compared with the copper alternatives [56]. This, coupled with the long term high price of copper has made aluminium wires a viable alternative. The technical advantages from the use of copper wires need to be weighed against their environmental and economical shortcomings. Analyses such as the one conducted here will contribute to identifying the environmental hotspots associated with each manufacturing step and therefore assist manufacturers and researchers in the field to improve the environmental performance of the technology through informed material substitution and process optimisation techniques.

3.2.2. *Disaggregating materials and energy contributions*

Fig. 3 demonstrates the contribution to the impact categories of materials usage and energy required to manufacture the CZTS monograin module. The analysis demonstrates that material usage had a significantly higher environmental burden than process energy for the production of the CZTS monograin module. The material contribution was the highest for ADP and ODP impact categories, and larger than 60% for the other impact categories. Process energy has a relatively low influence on all impact categories; except for PERT that demonstrated a higher contribution for process energy (56%). The total energy required for the whole module production is around 0.1925 MJ and the estimated material embedded energy is around 0.2965 MJ. This shows that process energy is significantly lower than materials embedded energy suggesting that the material choice and composition optimisation needs to be considered more carefully if the environmental impact of the technology is to be reduced. The analysis also further highlights the efficiency of the roll-to-roll processing as a deposition technique. This will be further analysed in the following section breaking down the total impact into the contribution of individual materials.

3.2.3. *Associated impact from individual materials*

Fig. 4 shows a detailed breakdown of the contribution of materials to impact categories for the roll to roll production of CZTS monograin module. The analysis demonstrates that the highest contributor to the majority of the impact categories was silver, which was mainly used in the deposition of the front contact. This is partly due to the higher

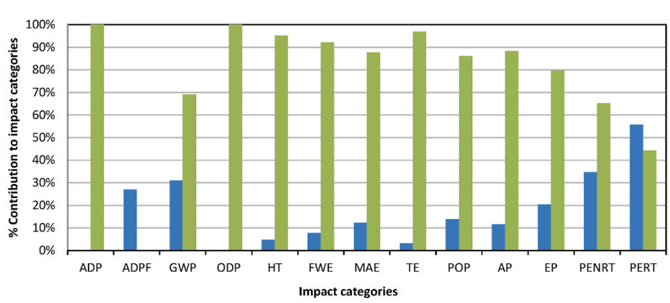

Fig. 3. Process energy and materials contributions to impact categories for the production of CZTS monograin module.

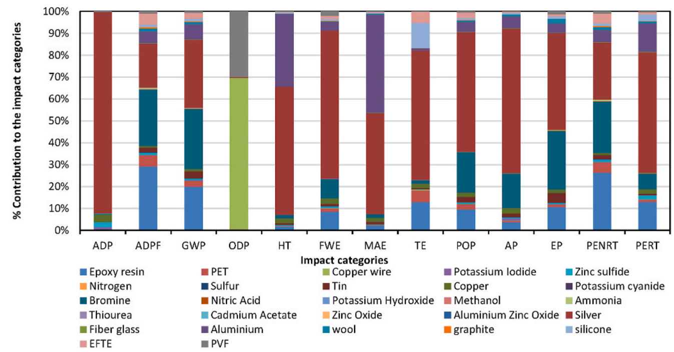

Fig. 4. Contribution of material usage to impact categories for the roll to roll production of CZTS monograin module.

quantity of the metal used in manufacturing the CZTS monograin module in comparison to other toxic compounds such as potassium cyanide and cadmium acetate. Apart from direct silver mining and processing, pure silver can be extracted as a co-product in the mining of other metals such as copper [57]. This method was found to have lower environmental impact than direct mining and processing of silver ores [58,59]. The extraction of silver from copper anode slimes can be achieved through three different processes, namely the pyrometallurgical process, hydrometallurgical process, and a hybrid of the two processes [57]. In the pyrometallurgical process reducing agents such as ammonia and hydrazine hydrate are commonly used in the final silver separation step after a series of pyrometallurgical process steps. This is problematic as hydrazine hydrate is both toxic and costly. The pyrometallurgical process also has other disadvantages such as high energy demand and low silver recovery [60]. Historically, the very harmful chemical cyanide was used as a leaching agent in the hydrometallurgical process, however this has largely been replaced with other chemicals such as ammonia, sulphuric acid, and nitric acid [57]. Although the hydrometallurgical method is considered to be more advantageous than the pyrometallurgical process, large amounts of leaching agents and harmful gases (including SO_2) are emitted into the atmosphere [57]. Sodium thiosulfate which has been proposed recently as an efficient silver leaching agent was determined to be environmentally friendly when compared with the commonly used ammonia as the leaching agent [60]. Due to the high environmental impact of silver observed here steps should be taken to reduce its footprint. Highly conductive metals such as copper with lower environmental impact could be used as a substitute [50]. However, where silver cannot be replaced in the manufacturing of the front contact, care should be taken to ensure that the source of the metal deployed is environmentally sustainable.

Apart from silver, the use of bromine, epoxy resin, and aluminium also had a significant environmental burden in terms of material usage for the production of CZTS monograin module. Epoxy resin defines a class of materials that contains one or more epoxide groups in their molecular structure [61]. It is known to be harmful to both animals and humans due to their release of smoke and toxic gases such as carbon monoxide and nitric oxide [62]. The use of bio-based epoxy resin were found to be more environmentally friendly than petroleum based ones [63–65]. As for aluminium, regardless of its primary or secondary (recycled) production, the majority of the impact comes from undertaking electrolysis, and the associated electricity demand in this process. The use of scrap aluminium however, reduces the impact of electrolysis and the overall energy demand [66]. It is worth mentioning again that copper wires in the embedding serial connection manufacturing step contributed highly (70%) to the ODP impact category. Materials identification from environmental assessment informs the product development researchers to substitute materials with high impacts with lower impact ones.

Further analysis was conducted to identify the impacts arising from the use of electricity. The breakdown of the total cumulative energy demand (CED) is shown in Fig. 5. The CED was about 0.49 MJ per functional unit (1 kWh), equivalent to 997.5 MJ per m^2. The CED values are extracted from GaBi as the summation of PENRT and PERT impact categories. It involves both the direct processing energy or electricity used during the manufacturing of devices and the energy embedded in the materials. A comparison of the processing steps demonstrated that embedding the semiconductor was the major contributor (48%) to the total CED mainly because of the materials' embodied energy in this process. Encapsulation also contributed to 24% of the total cumulative energy demand, with other processes not having a significant contribution.

3.2.4. Normalisation of the results

A comparison of environmental impact categories associated with the processing steps for the production of conventional CZTS monograin module is demonstrated in Fig. 6. The results are normalised per kWh using CML 2001 method described in Section 2.3. The MAE impact shows an exception to the general order of impacts contributing 62% to the total impact. By definition, MAE refers to the effects of toxic compounds on the marine ecosystem. It is caused by the air emissions of electricity production and non-ferrous metals [67]. In this study, MAE impact is largely affected by the use of silver and aluminium (in depositing front contact, removing foil and abrasion of membrane and encapsulation) and electricity usage. The ADP impact was also a significant contributor (22%), mainly due to the use of silver (>90 %) in depositing the front contact layer. This category's primary focus is on how the extraction of minerals and fossil fuels, which are system inputs, affects human and ecosystem health. The abiotic depletion factor is calculated for each extraction of minerals and fossil fuels. Based on concentration reserves and pace of de-accumulation, this indicator operates on a global scale [47].

Other impact categories such as ADPF, GWP, HT, POP and AP had much lower environmental impacts (Fig. 6). The deposition of front contact had the highest contribution (45%) to the overall environmental profile, followed by the removal of foil and abrasion of membrane process step (20%), embedding of semiconductor (15%) and encapsulation (12%), with the other processing steps having low weighting.

3.3. Energy payback time on a total energy use basis

The EPBT is the amount of time needed for a solar system to produce as much energy as was used during its construction and decommissioning. The concept is used as a key reference to better assess the

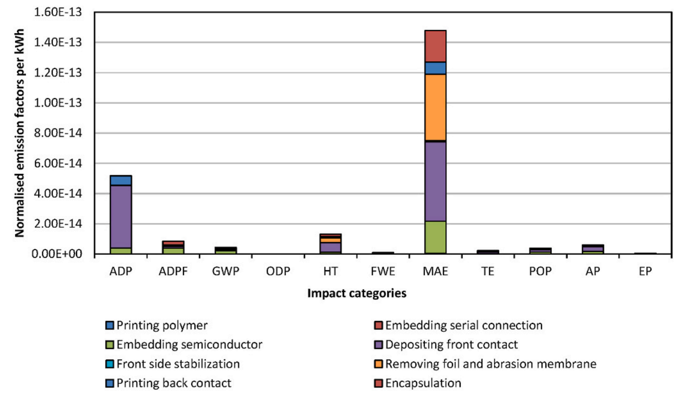

Fig. 6. Comparison of environmental impact categories and processing steps for the production of Conventional CZTS monograin module when normalised per kWh using CML 2001 method.

performance of energy sources. Due to their similarity to economic payback times, EPBT is appealing as an indicator of energy performance. Because the EPBT is additive, the total EPBT of the solar system can be calculated by simply adding the EPBT values for each component of the solar system. The limitation of the EPBT is that it does not take into account energy gains over the course of the remaining economic lifetime [46,68]. The EPBT was estimated in this study to be around 4.13 years, similar to silicon based solar modules with a range of 2.56–4.88 years [69]. This was lower than CIGS (6.12 years) but higher than thin film CTZS (1.5 years) solar modules. The lower value for thin-film CZTS is mainly attributed to the much lower embodied energy of the materials used in the manufacturing despite its higher electricity demand [27]. The calculated EPBT for CIGS in this study was similar to the reported range of 2.52–6.23 years by Ref. [70].

3.4. Impact results under different sensitivity scenarios

A sensitivity analysis using five alternative scenarios as presented in section 2.4 was conducted while varying key parameters and assumptions such as solar irradiance, energy mix, and material usage. Fig. 7 presents hotspot results that could provide guidance for manufacturers and researchers on how to minimise the effects of conventional CZTS monograin module production (scenario S0), which was used as the reference scenario.

The influence of other electricity sources was considered in this analysis. As expected, changing the EU28 energy mix to renewable

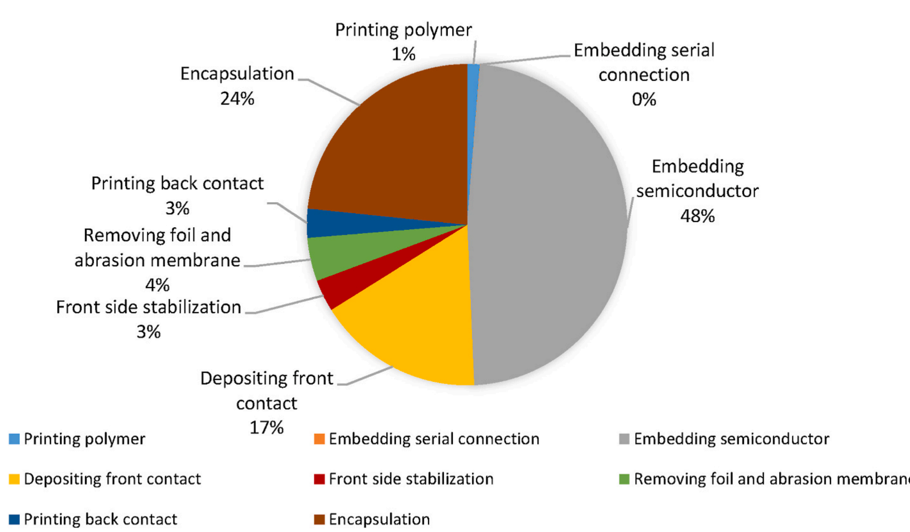

Fig. 5. Contribution of each process step of the CZTS monograin production to the total cumulative energy demand (0.49 MJ per functional unit).

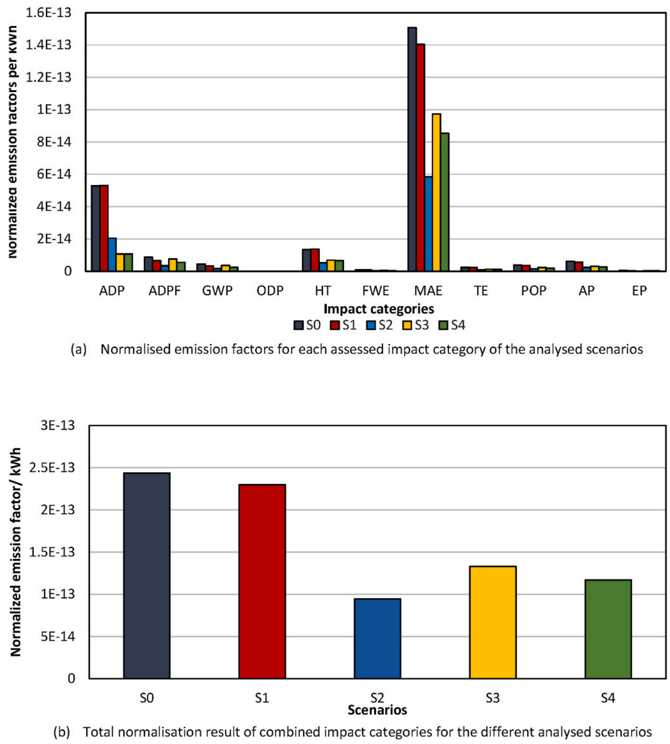

Fig. 7. Normalisation results under different scenarios
S0: Baseline scenario for the conventional CTZS monograin production
S1: The use of renewable energy (EU-28 solar module)
S2: Change of location to the Sahara Desert
S3: Improved CZTS monograin module with material substitution
S4: Improved CZTS monograin module with material substitution coupled with the use of renewable energy mix.

energy EU28 solar module mix in scenario S1 did not have a significant contribution to the reduction of the overall impact results (6%). The primary reason for this was that for all impact categories studied, the main contribution was attributed to the usage of specific materials rather than the electricity usage as demonstrated in Section 3.2.1. Therefore, scenario S3 investigated the impact of changing specific materials used in the front contact (e.g. silver) that were highlighted by this analysis as environmental hotspots. The manufacturer, informed by the analysis, substituted silver with dopant salt and citric acid to decrease the overall impact of the technology without compromising the conversion efficiency. The overall impact was reduced by 45% (S3) and was further reduced by 52% when combined with the use of renewable energy mix (S4). The breakdown of the materials utilised during manufacturing of the developed CZTS monograin module is shown in Figure SM1 in the Supplementary Materials.

The geographical location analysis (Scenario S2) demonstrated that the placement of the solar modules in places with higher solar irradiance such as Sahara Desert (as a reference point) could contribute significantly to the reduction of the overall impact (a reduction of 62%), in comparison to the other assessed scenarios. This is mainly attributed to the availability of incident solar radiation and therefore lower area of panel required to achieve the 1 kWh functional unit defined for the analysis.

3.5. Comparative analysis of CZTS monograin and thin-film solar module technologies

Thin-film and monograin techniques are the two dominant manufacturing techniques for CZTS material composition. Given that this study presents the LCA results associated with the monograin technology for the first time, the generated results are compared with CZTS thin-film and CIGS as a commercially available thin-film technology (Vidal et al., 2021, Ibn-Mohammed et al., 2017).

Fig. 8 demonstrates the normalisation results per kWh for these technologies. The production of CZTS monograin module performs better than thin-film when silver is substituted with a new material composition (as presented above). Silver is commonly used as an electrode mainly due to it being one of the best metal electrical conductor [71]. However, eliminating its use in the front contact significantly improve the overall environmental performance of the monograin technology in comparison to the other assessed solar module technologies. The conventional CZTS monograin was outperformed environmentally by CIGS due to its higher efficiency.

Marine aquatic ecotoxicity and abiotic depletion tend to dominate the impact results for all studied technologies. Similar trend was observed when comparing the environmental performance of the assessed solar technologies by impact category (Fig. 8a) to the overall performance presented in Fig. 8b, whereby CIGS demonstrated the lowest impact. This was mainly due to the higher efficiency (20%) [27]. This is worth noting that the monograin technique is offering just over 20% additional environmental impact compared to CIGS with a 50% lower efficiency, suggesting promising future for the technology. Similar to the results presented in this paper [28], demonstrated that CIGS performed better than CZTS thin-film. For all impact categories assessed, the absorber layer accounted for more than half of the impact in CIGS and CZTS thin-film. Layers in the substrate, back contact, and buffer contributed to the remaining impact. In CIGS, the absorber layer was responsible for over 90% of the ecotoxicity.

The efficiency of solar modules has an inverse relationship to its environmental impact. An increase in the efficiency of a solar module tends to lead to a subsequent decrease in its environmental impact. Resalati et al. [27] demonstrated that if CZTS thin-film solar modules can rival CIGS in efficiency, there would be an environmental advantage to their adoption over the well-established CIGS technology. A further analysis of the results suggests that the efficiency of standard CZTS monograin modules need to be above 21.87% in order to rival CIGS' environmental performance (calculated on the basis of a total

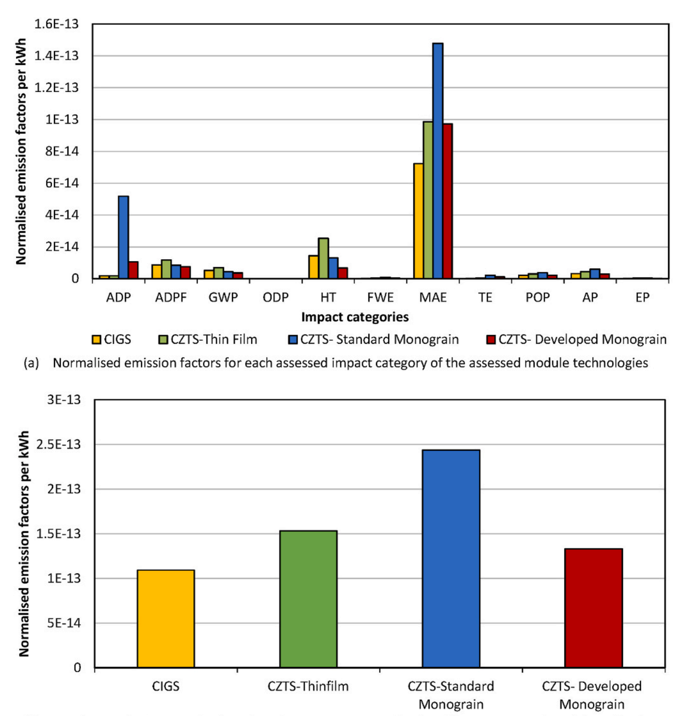

Fig. 8. Normalisation results of the different assessed solar module technologies.

normalised score as presented in Fig. 8b). This is technically feasible as this efficiency is below the Shockley-Queisser limit of 32% for CZTS solar modules [7]. However, only a 1.95% increase in efficiency of the developed CZTS monograin (based on silver substitution), will lead to the technology matching the environmental performance of CIGS. When the environmental impact of standard CZTS monograin are compared to that of its thin-film counterpart, a module efficiency of 15.60% is required to match the thin-film performance.

4. Conclusions

This study, for the first time, evaluated the environmental impacts of CZTS monograin modules using primary manufacturer data. A comparison of environmental impacts was made with CZTS thin film and the commercially available CIGS solar technology. The results showed that the front contact in the monograin production contributed to the majority of assessed impact categories. Under the assumption that the impact categories are equally weighted (normalised values), marine aquatic ecotoxicity and abiotic depletion tend to dominate the impact results. The highest contribution (about 80%) was observed for marine aquatic ecotoxicity.

Overall, material usage had significantly higher environmental burden than process energy for the CZTS monograin module production. The estimated embedded energy of the material is around 0.2965 MJ, while the total energy needed to manufacture the entire module is approximately 0.1925 MJ. This demonstrates that the process energy is substantially lower than the energy incorporated in the materials suggesting that material composition optimisation could be performed to improve the associated environmental performance. The highest contributor to the majority of the impact categories was silver, which was mainly used in the deposition of the front contact. The calculated distribution of the cumulative energy demand (CED) in the production steps indicated that the embedding semiconductor process step was the major contributor (48%), followed by the encapsulation (24%), mainly due to the embodied energy of the materials. The CED was about 0.49 MJ per functional unit (1 kWh), which delivers an EPBT of 4.12 years. The EPBT of the CZTS monograin module was similar to silicon based solar modules and lower than the commercially available CIGS thin film technology.

The results of the sensitivity analysis demonstrated that the absolute values of environmental impacts are significantly influenced by the geographical location of installation, which can contribute to the reduction of the overall impact (62%) of solar devices. In addition, eliminating the use of silver in the front contact was found to improve the modules overall environmental performance substantially (by 45%), making its impact comparable to CZTS and CIGS thin film technologies. Further LCA studies on CZTS monograin modules should be conducted to assess the environmental impacts of the usage and end-of-life stages as well as considering future improvements such as increased module efficiency, lower energy and material consumption, and recycling and recovery of materials, which are predicted to decrease the overall impacts.

CRediT authorship contribution statement

Amani Maalouf: Writing – original draft, Software, Methodology, Formal analysis. **Tobechi Okoroafor:** Writing – review & editing, Writing – original draft, Software, Formal analysis, Data curation. **Stefan Gahr:** Validation, Resources, Data curation, Conceptualization. **Kaia Ernits:** Writing – review & editing, Validation, Conceptualization. **Dieter Meissner:** Validation, Methodology, Conceptualization. **Shahaboddin Resalati:** Writing – review & editing, Supervision, Methodology, Funding acquisition, Conceptualization.

Declaration of competing interest

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

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.solmat.2022.112161.

Abbreviations

Solar modules

CIGS	Copper indium gallium selenide
CZTS	Copper zinc tin sulphide
CdTe	Cadmium telluride
CZTSSe	Copper zinc tin sulphide selenide

Nomenclature

EU-28	European Union
FU	Functional unit
LCA	Life cycle assessment

Impact categories

ADP	Abiotic depletion
ADPF	Abiotic depletion (fossil fuels)
GWP	Global warming
ODP	Ozone layer depletion
HT	Human toxicity
FWE	Fresh water aquatic ecotoxicity
MAE	Marine aquatic ecotoxicity
TE	Terrestrial ecotoxicity
POP	Photochemical oxidation
AP	Acidification
EP	Eutrophication
PENRT	Primary energy non-renewable resource
PERT	Primary energy renewable resource

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