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Lu Wang, Lars Oberbeck, Mathilde Marchand, Paula Perez-Lopez. Modelling recycling for the life cycle assessment of perovskite/silicon tandem modules. EPJ Photovoltaics, 2024, 15, pp.14. 10.1051/epjpv/2024010 . hal-04559847

HAL Id: hal-04559847

<https://hal.science/hal-04559847>

Submitted on 25 Apr 2024

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Modelling recycling for the life cycle assessment of perovskite/silicon tandem modules

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Received: 15 September 2023 / Accepted: 29 January 2024

Abstract. With the massive growth of the global capacity of photovoltaics (PV) over the last decade, the PV waste is expected to increase dramatically in the near future. Having potential to reduce the use of raw materials and preserve natural resources, PV recycling is attracting more and more attention. This being said, the environmental impacts over the life cycle of PV technologies, including the end-of-life (EoL) stage, should be evaluated carefully. Life cycle assessment (LCA) is currently the most common methodology to assess the potential environmental impacts of a product throughout its entire life cycle. However, the modelling of recycling in LCA has always been a challenge and no consensus has yet been reached, since the treatment of recycling does not only involve an EoL management of waste, but also the production of recycled material. Perovskite on silicon tandem is a widely investigated emerging PV technology having the potential to overcome the power conversion efficiency (PCE) limit of the single-junction crystalline silicon technology. The EoL modelling seems more challenging in the case of emerging technologies for which the EoL is more uncertain than for established technologies. In this article, six common and important approaches of EoL modelling in LCA were applied to future perovskite/silicon tandem modules to analyze the effect of the different EoL modelling approaches on the LCA results. The aim was to identify the most suitable methodological approaches to account for recycling, when modelling the life cycle of PV modules. The environmental performance of perovskite/silicon tandem modules was assessed over their life cycle and expressed in terms of impacts per m² of module. After testing the six EoL modelling approaches and comparing the LCA results, the EoL modelling choice was found to lead to non-negligible differences. For example, in terms of climate change, the impact of the tandem modules ranges from 45 to 59 kg CO₂-eq/m². Among the six EoL modelling options, the approaches of simple cut-off and cut-off with economic allocation are more oriented towards the promotion of high rates of recycled material integrated as an input to the assessed product among industrial actors, while the approach of closed-loop allocation provides incentives to maximize the ratio of recycling at the EoL, regardless the initial ratio of recycled content within the product. Some approaches such as the circular footprint formula (CFF) tend to provide both incentives to increase the content of recycled input material in the manufacturing of the product and the recycling ratio at the EoL of such product. After applying the different alternatives, a set of recommendations to select the relevant EoL modelling approaches are provided: 1) the CFF is recommended as a representative approach due to its wide applicability, tending to provide an intermediate result and reflecting the characteristics of materials; 2) sensitivity analysis should be applied to check the robustness of the results, 3) the cut-off approach and the closed-loop allocation should be used at least for the sensitivity analysis.

Keywords: Photovoltaic / end-of-life / sustainable / renewable energy / emerging technology

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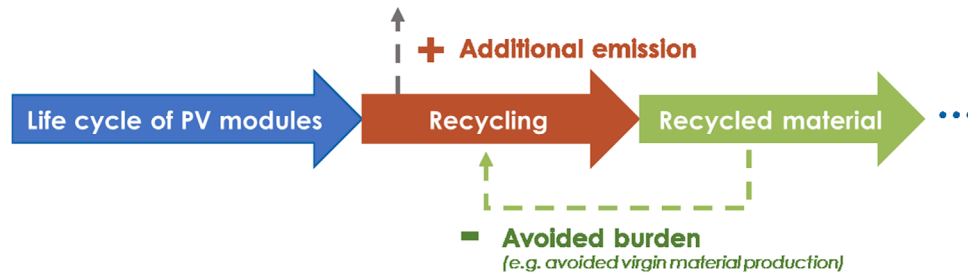


Fig. 1. The double function of recycling: the EoL stage of a product and the production of recycled material.

1 Introduction

Photovoltaics (PV) remains the powerhouse for growth for renewable electricity. In 2022, the global capacity of installed PV increased by more than 25% compared with the previous year, and exceeded 1185 GW by the end of the year [1]. Considering an average lifetime of 30 years for the panels, the cumulative PV panel waste is forecasted to reach around 70 million tons by the year of 2050 [2]. Such outlook brings both challenges and opportunities. In particular, the European Union (EU) has pioneered to include PV waste into the Waste of Electrical and Electronic Equipment (WEEE) directive and to specify the regulations including the PV collection, recovery and recycling targets [3]. Accordingly, an appropriate EoL management of PV including recycling is gaining more and more interest.

From an environmental perspective, recycling is typically considered to have the potential to reduce the use of raw materials and the amounts of waste and avoid the shortage of mineral resources. However, the overall environmental impact of the PV, including the EoL recycling and disposal, should be carefully evaluated to avoid the burden-shifting between stages of a product's life cycle [4,5]. Life cycle assessment (LCA) is currently the most common methodology to assess the potential environmental impacts of a product or a system, with a multi-criteria perspective, throughout its entire life cycle [4,5]. According to previous studies applying LCA, the environmental impact on climate change from the generation of 1 kWh of electricity by a PV system ranges from 50 to 200 g CO₂-eq/kWh and the Cumulative Energy Demand (CED) from 0.4 to 1.5 MJ/kWh mainly depending on the different PV technologies, the location and year of fabrication [6–8]. Some emerging PV technologies can even reduce the impact on climate change with a result lower than 20 g CO₂-eq by improving the PCE, eco-fabrication and recycling [9,10].

Despite the widespread use of LCA, certain methodological choices in LCA still lack consensus, which may lead to comparability issues between different studies and potential loss of credibility [11]. This is the case for the LCA modelling of the recycling of materials from one product to another, which raises an issue of allocation. The recycling treatment process may lead to additional emissions, while being also the production process of material, which has the potential to reduce the need for primary production of material [12] (cf. Fig. 1).

To date, different guidelines have been published on how to model recycling in LCA. Among them, ISO 14040 and 14044 in their 2006 versions [4,5] have stated the principles dedicated to the allocation issues for recycling. ISO 14067, the guidelines providing specific requirements for conducting LCA under the context of Greenhouse Gas (GHG) emission, has identified two modelling approaches for recycling: the closed-loop allocation (Sect. 2.2.1) and open-loop allocation (Sect. 2.2.1) [13]. A more specific document on the methodological guidelines for PV systems has been developed and is systematically updated by the Photovoltaic Power Systems Programme (PVPS) Task 12 of the International Energy Agency (IEA) [14]. PVPS Task 12 guidelines recommend the cut-off approach with economic allocation (cf. Sect. 2.2.1) and the closed-loop allocation (also known as the end-of-life approach in Task 12, cf. Sect. 2.2.1) for the modelling of recycling of PV products [14]. In parallel, the EU recommended a specific approach for the modeling of recycling in their Product Environmental Footprint (PEF) in 2018: the circular Footprint Formula (CFF). The CFF takes into account relatively comprehensive aspects of recycling such as quality degradation of material due to recycling and the balance between supply and demand for individual recycled materials, this resulting in an approach that is quite complex [15,16]. Other international guidelines can also be mentioned, such as the Greenhouse Gas (GHG) protocol, and Publicly Available Specification (PAS) 2050, which propose their own solutions for the modelling of recycling [17,18]. Despite all these efforts, these recommendations in different guidelines are not totally in line with each other [12]. As previously indicated, the choice of EoL modelling approaches may have non-negligible effects on the LCA result of products which use recycled material as an input to the manufacturing phase, and products which can be totally or partially recycled after the use phase [12]. Therefore, a clear justification of methodological choices related to EoL modelling is essential to ensure the representativeness of the LCA results.

Perovskite on silicon tandem is a promising technology to overcome the Shockley-Queisser limit of the current crystalline silicon single-junction PV technology and is widely investigated [12]. Being an emerging technology, studies dedicated to tandem module recycling are rare, but processes and assumptions to conduct LCA studies could be based on existing recycling options for commercial crystalline silicon PV modules [19]. Over the past few years, new commercial and demonstration-scale recycling options

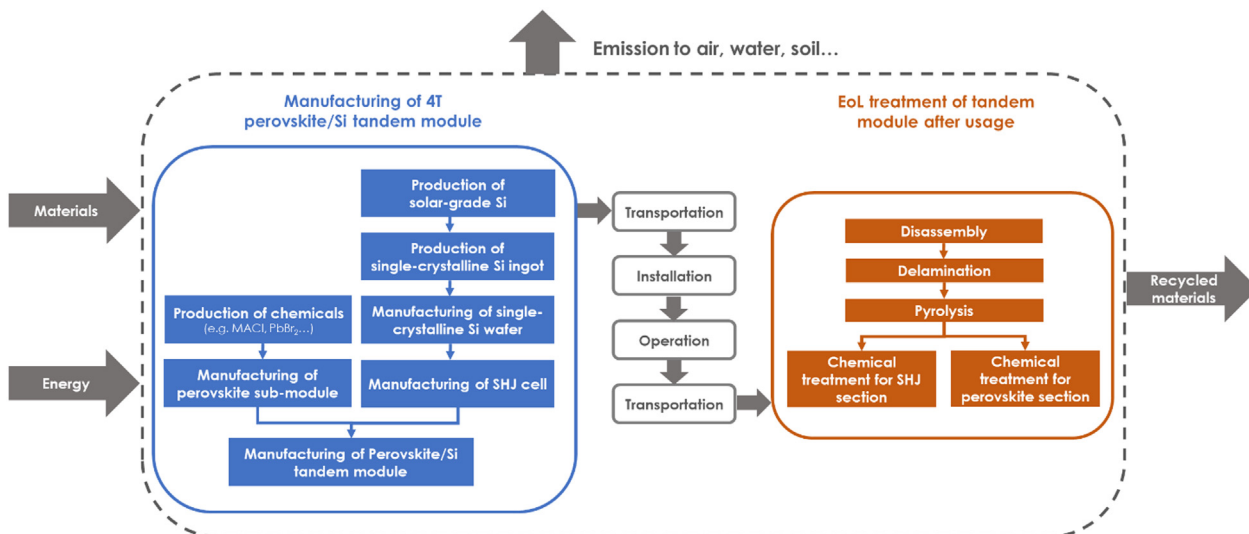


Fig. 2. The system boundary of perovskite/Si tandem modules in this study.

have emerged [20] and several recycling strategies have been demonstrated for perovskite PV cells to recover lead and valuable components at the lab-scale [21,22].

The most common processes for the recycling of PV modules, including crystalline silicon modules, begin with the mechanical disassembly of aluminum frame and junction box [20,23]. Following this, the individual layers of the module laminate (glass, encapsulant, silicon cells, backsheets or encapsulant and rear glass) are separated. The delamination can be done using (i) a mechanical method, such as the water jet process and the diamond wire cutting process to separate the glass/backsheets structure, (ii) a thermal method, such as pyrolysis, hot knife and flashlight, or (iii) solvent and ultrasonic separation [24–26]. Among these technologies, flashlight technology looks very promising for separating the glass/glass structure of the perovskite/silicon tandem modules and recovering the unbroken glass sheets [26]. In the subsequent stages, different processes are used to extract various materials such as silver, silicon or lead from the remaining components.

- Several approaches stop after the delamination process for the silicon cells and recover lower-purity Si (ferro-Si) with a minimum Si content of 75%. Then, a standard hydrometallurgical treatment with acid leaching followed by precipitation and filter press is used to further extract and separate the different materials. With such chemical treatment, Ag could be recovered from the cells with a recovery rate of more than 99% [27], and Si could be recovered at the purity of 5N (99.999%) or even solar-grade level (99.9999%) [28].
- For the perovskite cell, after mechanical or thermal delamination to separate the encapsulant and glass, the perovskite layers can be treated with chemical solvents. The various layers can be removed one by one with different solvents, or all layers removed at once using a universal solvent and then elements such as lead can be recovered from the mixed solution [21,22].

The different recycling technologies are undergoing rapid improvement. Yet, as mentioned previously, further clarification of EoL modelling approaches in LCA for PV modules would also be necessary. In this study, six typical approaches of EoL modelling from important guidelines of LCA were applied to the perovskite/silicon tandem modules developed by the Institut Photovoltaïque d’Île de France (IPVF) to analyze and compare their influence on the LCA results, so to provide some practical recommendations regarding the choice of modelling options.

2 Methodology

2.1 Life cycle assessment

This section presents the general methodology used in this study, i.e. the life cycle assessment. The LCA was applied to assess the environmental impact of the tandem modules throughout their entire life cycle. The four phases of LCA defined by ISO 14040 and 14044 [4,5] standards were applied as described below.

2.1.1 Goal and scope

In this study, the environmental impact related to the manufacturing and EoL treatment of 1 m² of perovskite/Si tandem modules was assessed. The product system includes the perovskite/Si tandem modules with their encapsulants, frame and other items such like wiring and junction-box (J-box) and aluminum frame, the inverters and mounting systems being excluded. The system boundary has been defined using a cradle to grave approach, thus, covering the raw material extraction and transformation and the energy production, the manufacturing of PV modules, transportation, installation, operation and EoL stages, including recycling of materials and final disposal (cf. Fig. 2). The installation

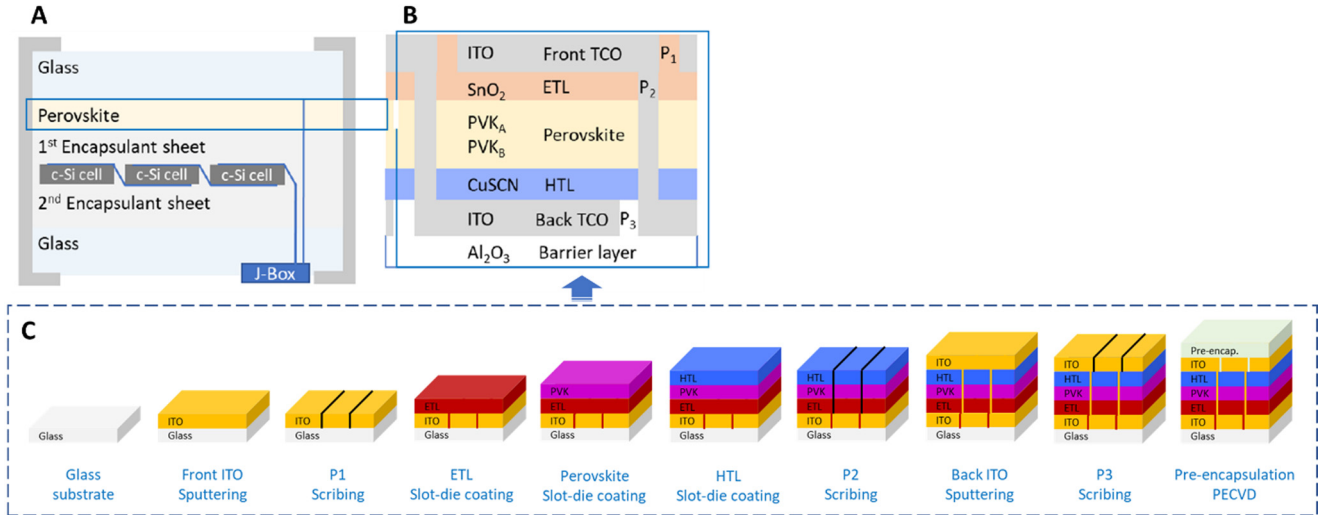


Fig. 3. Schematic drawing of the 4T perovskite/Si tandem module and the process routes of the architecture of the perovskite section [29,30]. (A: Architecture of the 4T2 perovskite/Si tandem module, with bottom and top sub-modules as well as module elements including encapsulant, frame, and items like ribbons, junction box (J-Box) and wiring. B: ITO (indium tin oxide) is the front and back TCO (transparent conductive oxide) electrodes; SnO_2 (tin oxide) is the ETL (electron transport layer); PVK_A , $\text{Cs}_{0.17}\text{FA}_{0.83}\text{Pb}(\text{I}_{0.83}\text{Br}_{0.17})_3$, is the perovskite absorbers used in 1-step perovskite deposition; PVK_B , $\text{Cs}_{0.05}\text{MA}_{0.4}\text{FA}_{0.55}\text{Pb}(\text{I}_{0.96}\text{Br}_{0.04})_3$, is used in 2-step perovskite deposition; HTL (hole transport layer) is made of CuSCN (Copper(I) thiocyanate); Al_2O_3 (aluminum oxide) serves as barrier layer; P1, P2 and P3 represent the laser scribing patterns. C: Step-by-step architecture of the perovskite section).

and operation of the modules are ignored due to their low contribution [29]. The modules were assumed to be produced and installed in France in 2030 and recycled in France in 2050. Based on this assumption, the transportation of modules is also ignored due to the limited distance of transport within the country. The software used to perform LCA is Activity Browser 2.7.4. This study is conducted to compare and analyze the impact of the choice of EoL modelling approaches on the LCA results. The intended audience are LCA practitioners, PV R&D researchers and strategy makers. Following the specific objective of this study, six modelling approaches of recycling were applied, which are detailed in Section 2.2.

2.1.2 Life cycle inventory

The life cycle inventory (LCI) phase in LCA consists of compiling data to quantify the use of resources and emissions for each process in the defined system. In this section, the processes related to the manufacturing of tandem modules and their EoL treatment are described.

2.1.2.1 Structure and manufacturing of perovskite/Si tandem modules

The perovskite/Si tandem modules under consideration are based on a four terminal two wires configuration (4T2), which is developed by researchers at the IPVF based on experimental data and scaling-up with expert opinion. The representative schematic of the tandem module is depicted in Figures 3a and 3b, which is among the most promising technologies from an industrialization perspective. It includes the top sub-module, which is composed of n perovskite cells connected in series in m parallel blocks, and

Table 1. Mechanical parameters of perovskite/Si tandem module [29].

Dimensions (mm)	2094 × 1038
Surface (m^2)	2.17
Active surface (m^2)	2.12
Weight (kg)	26.19
Weight (kg/m^2)	12.05

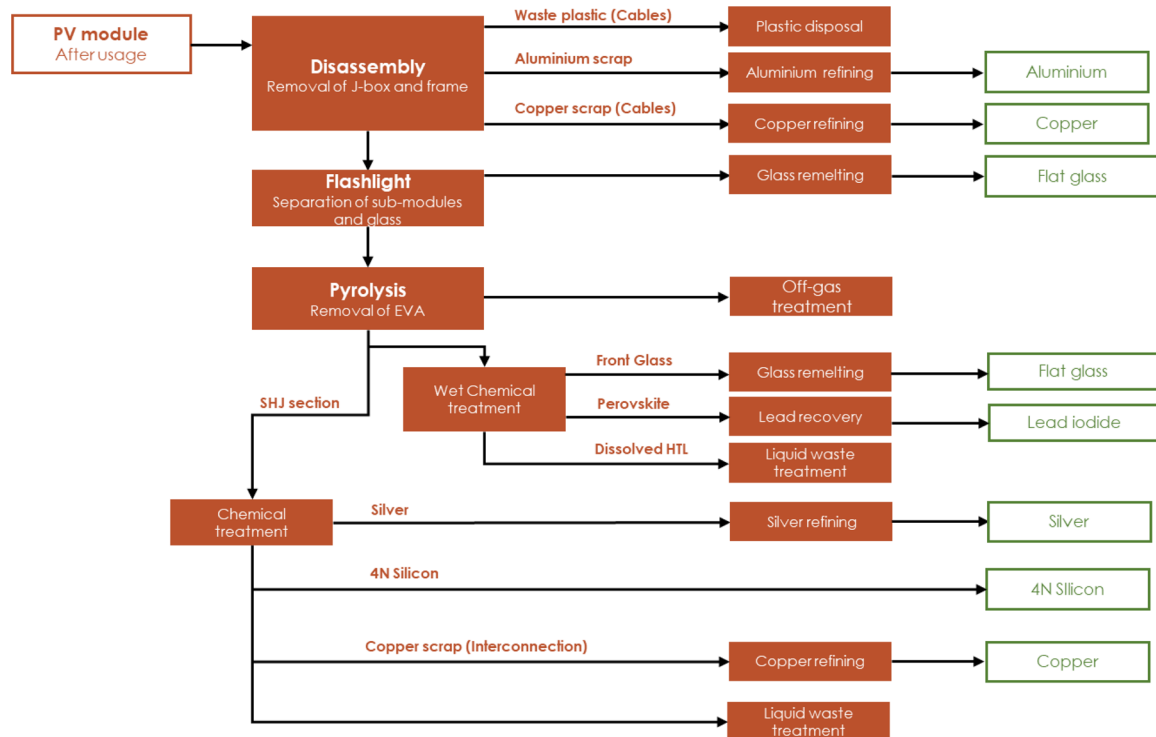
Table 2. Composition of perovskite/Si tandem module [29].

Composition	Fraction	Mass (kg/m^2)
Glass (front + rear)	84.7%	10.21
Al (frame)	7.9%	0.95
EVA	3.5%	0.42
Si	2.6%	0.31
Polymer from cables	0.7%	0.08
Cu (cables)	0.4%	0.05
Cu (interconnection)	0.1%	0.01
Ag	0.03%	0.003
Pd (perovskite)	0.0%	0.0002
Others (Sn,In)	0.1%	0.02
Total	100.0%	12.05

the bottom sub-module composed of 72 bifacial Silicon Heterojunction (SHJ) cells. The mechanical parameters and the composition of the tandem module are shown in Tables 1 and 2, respectively.

Table 3. Main references of LCI data of perovskite/Si tandem manufacturing [29].

Process block	Main references
Production of chemicals (perovskite section)	Alberola et la (2018) [33–35], IPVF confidential data
Manufacturing of perovskite sub-module	IPVF confidential data
Production of solar-grade silicon	Task 12 [31], with adjustment based on ITRPV [36]
Production of single-crystalline silicon	Task 12 [31], with adjustment based on ITRPV [36]
Manufacturing of single-Si wafer	Task 12 [31], with adjustment based on ITRPV [36]
Manufacturing of SHJ wafer	Task 12 [31], Louwen et la (2015) [37], ITRPV [36]
Manufacturing of perovskite silicon tandem module	Task 12 [31], with adjustment based on ITRPV [36]

**Fig. 4.** Simplified process route of EoL treatment of tandem modules.

The process route of manufacturing the perovskite section, including the materials and coating technologies are shown in Figure 3c. For reasons of confidentiality, the inventory data for the manufacturing of the perovskite stack will only be partially reported in this article. It should be noted that the environmental burden related to this part is limited with the perovskite stack (without front glass) contributing only 4% to the carbon footprint (CFP) of the whole module [29]. Therefore, it is not expected to have a significant impact on the comparison of the EoL modelling approaches and the conclusion in this study.

The inventory data for the other components of the module was mainly based on the IEA-PVPS Task 12 [31] with the input background flow from Ecoinvent 3.6 [32]. Several important adjustments have been made to take into account the bifacial structure and SHJ technology.

The main references of each process block for the tandem manufacturing are set out in Table 3, the adapted adjustments listed in Appendix A.

2.1.2.2 EoL treatment of perovskite/silicon tandem modules

Based on the above-mentioned tandem structure, a specific recycling process has been developed by IPVF based on a combination of technologies described in industry reports and academic literature [21,22]. The recycling processes assumed in this study are consistent with the general recycling routes of PV modules described in the introduction (cf. Sect. 1), including mechanical disassembly, delamination, pyrolysis, then extraction of the separated submodule components by chemical treatment (cf. Fig. 4).

After the separation steps, some materials may require further purification. After removing the Al frame and J-box, the flashlight technology consists in the separation of the module laminate using a high-intensity and low-energy light source to flash the module. Light absorbing layers are thereby heated to temperatures of around 600 °C within microseconds, followed by a rapid cool-down close to room temperature [26]. This process leads to a separation of the silicon cells from the encapsulants and the front glass with perovskite layers can be separated from the laminate. The silicon cells together with encapsulants then undergo a pyrolysis process to ensure a complete removal of the encapsulant. The encapsulant-free silicon cell is then treated with acid and alkaline etching, and related inventory data is adapted from ROSI Solar’s LCA study [38]. For the perovskite sub-module, it is assumed that the perovskite layer stack remains on the rear side of the front glass. Using a treatment of dimethylformamide (DMF), the perovskite can be dissolved completely, then the lead is extracted from the solution using carboxylic acid cation-exchange resin [21]. Once the various layers have been removed, the front glass can be recovered. The inventory of the EoL stage and key assumptions are detailed in Appendix B. The recovered materials are aluminum, copper, glass, silver, silicon and lead.

Inventories for other processes, including the production of raw materials, energy supply and the refining of the recycled materials, are based on Ecoinvent 3.6 database (cf. Appendix B).

2.1.3 Impact assessment

Impact assessment is the “transformer” from the LCI list to certain environmental impact. Different methodologies allow the impacts on different environmental categories to be modelled, so to achieve a multi-criteria evaluation that avoids burden-shifting from one environmental category to another. In this article, the number of categories has been restricted to four in order to focus the discussion on EoL modelling. Yet a more extensive selection should be applied for a more comprehensive analysis. The environmental impact on climate change has been selected as a representative impact category to illustrate the application of the approach, due to its extended use in the LCA of PV systems and its connection with the objective of the energy transition. The indicator “CED, non-renewable” was also chosen to quantify the amount of the direct and indirect non-renewable energy use throughout the life cycle of the PV modules. Avoiding the shortage of mineral resources is typically considered as a benefit of recycling, but the impact on resources depletion needs to be carefully checked. The impact on ecotoxicity is chosen since the lead toxicity in perovskite cells is a hot spot. The impact assessment methods recommended by the IEA-PVPS Task 12 guidelines on LCA of PV were used [12].

2.1.4 Interpretation

Interpretation is the final phase of the LCA, aiming to evaluate and communicate and at making recommendations based on the LCA results. During this phase, the LCA

practitioner considers the assumptions, limitations and uncertainties associated with the study. In this research, a sensitivity analysis was performed by applying different options of EoL modelling. The LCA results were compared and analyzed so to provide recommendation on the choice of modelling approaches.

2.2 EoL modelling approaches

As a main focus of the present study, the EoL modeling in LCA is subject to a dedicated presentation in this section. In 2020, Ekvall and his collaborators [12] published a report to summarize the existing EoL modeling approaches in LCA (12 in total) and evaluate each approach on different criteria. Thanks to this comprehensive work, different EoL modelling approaches are described in a uniform way to facilitate comparisons. Of the 12 methods mentioned, we have chosen 6 that are the most widely used and referenced by the guidelines on LCA (cf. Tab. 4). This section will therefore provide description regarding the 6 EoL modelling approaches applied in this study, including description taken from the guidelines.

2.2.1 Description of the six EoL modelling approaches applied in this study

The general schema related to the life cycle of one material in a product is set out in Figure 5. The manufacturing of a product requires virgin and/or recycled material. The share of recycled material in the product is noted as R_1 , so the share for virgin material is equal to $1-R_1$. E_V in this study refers to the specific emissions and resources related to the production of virgin material, and E_{Rin} corresponds to any specific emission or resource related to the recycling process to produce the recycled material used as an input for the manufacturing of the product. The recycled material may be related to its upstream recycling process E_{Rin} but may also be related to E_V depending on the different approaches. After usage of the product, in the EoL stage, some waste may be sent to recycling with recycled material being produced. R_2 is the proportion between the output recycled material and the total material used for the manufacturing of the product. In this study, we mainly focus on the material recycling, while the energy recovering by combustion is considered as a final disposal without energy recovering. Therefore, $1-R_2$ is the rate of material for the final disposal. E_{Rout} refers to the specific emissions and resources consumed arising from the recycling process at EoL. E_V^* represents the specific emissions and resources related to the production of virgin material assumed to be substituted by the recycled materials. E_D refers to the specific emissions and resources related to the disposal of waste material at the EoL of the analyzed product. The sum of the allocated emissions and resources related to the production of virgin material (E_V and E_V^*), recycling (both E_{Rin} and E_{Rout}) and final disposal (E_D) of the studied material is noted as E .

The simple cut-off is described as the easiest approach in the Ekvall’s report [12]. The main characteristics are its 100/0 allocation: 100% of recycling is allocated to the downstream recycled material, and 0% to the EoL of

Table 4. EoL modelling approaches used in this study.

EoL approach	References	Comments
Simple Cut-off	Ekvall et al. (2020) [12] Environmental Product declarations (EPD) International 2017 [39] PSA 2050 (Recycled content method) [18] GHG Protocol (Recycled content method) [17]	Simple cut-off was selected due to its widespread use and easy application in practice.
Cut-off with economic allocation	Ekvall et al. (2020) [12] Dutch Handbook on LCA [40] IEA-PVPS T12 (Cut-off approach) [14]	Cut-off with economic allocation was selected due to recommendation by specific guidelines relating to PV and its easy application in practice.
Closed-loop allocation	ISO 14044:2006 [4], ISO 14067:2018 [13] Ekvall et al. (2020) (Allocation to material losses) [12], PSA 2050 (Closed-loop approximation) [18] GHG Protocol (Closed-loop approximation) [17] IEA-PVPS Task 12 (End-of-life approach) [14]	Closed-loop allocation was selected due to its widespread use, easy application in practice and its market incentive. Usually, it provides incentive to promote EoL recycling.
50/50 method	Nordic Guidelines on LCA [41] Ekvall et al. (2020) [12]	50/50 method was selected, since it provides an intermediate solution via a simple way (50/50 allocation).
Open-loop allocation	ISO 14067:2018 [13] Ekvall et al. (2020) (Market price-based allocation) [12]	Open-loop allocation referenced in ISO series was selected, since it can provide a way of allocation reflecting the specific characteristics of the material based on the market price.
Circular Footprint Formula	PEF [15] Ekvall et al. (2020) [12]	CFF recommended in the PEF guide selected, since in this approach, elements are taken into account in a comprehensive manner. It is not so complex in practice due to the data availability of some allocation factors and quality degradation factors provided in the PEF. It provides a compromising solution between two market incentives and reflects the market features of specific material.

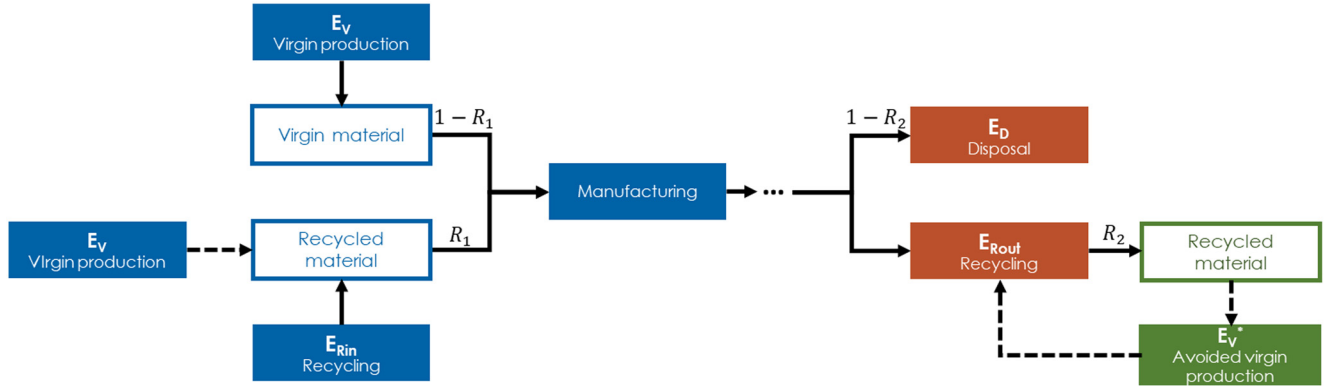


Fig. 5. General scheme of the life cycle stages of a material involved in a product.

the upstream product [17,18,39]. The total allocated emissions and resources of the production of virgin material, recycling and final disposal (E) are modeled as follows (cf. Eq. (1)):

$$E = (1 - R_1) \times E_V + R_1 \times E_{Rin} + (1 - R_2) \times E_D. \quad (1)$$

Instead of a 100/0 allocation, the recycling in the approach of cut-off with economic allocation is allocated using an economic allocation factor [12]. The allocation factor is calculated on the basis of the cost of treatment of recycling and the price of the output recycled material [14,40]. The examples are provided in the Dutch Handbook on LCA and IEA-PVPS Task 12 to explain the calculation of the economic factor [14,40]. E is modeled according to the following equation (cf. Eq. (2)):

$$E = (1 - R_1) \times E_V + (1 - A_e) \times R_1 \times E_{Rin} + (1 - R_2) \times E_D + A_e \times R_2 \times E_{Rout}. \quad (2)$$

A_e is the allocation factor in the approach of cut-off with economic allocation.

The closed-loop allocation is also called 0/100 allocation [12]. The main characteristic of this approach is an additional credit $-E_V^*$ that is given to the studied product systems due to the output recycled material. In other words, the burden related to the production of virgin material is attributed a “100%” to the product life cycle where the material is not recycled and, hence, lost from the technosphere [12]. The equation of closed-loop allocation is as follows (cf. Eq. (3)):

$$E = E_V + R_2 \times E_{Rout} + (1 - R_2) \times E_D - R_2 \times E_V^*. \quad (3)$$

When it comes to the 50/50 method, the main characteristics are its equal allocation within the production of virgin material, recycling as well as final disposal [12,41]. The equation for this modelling is as follows (cf. Eq. (4)):

$$E = (1 - R_1) \times E_V + 0.5R_1 \times E_V - 0.5R_2 \times E_V^* + 0.5[(1 - R_1) + (1 - R_2)] \times E_D + 0.5(R_1 \times E_{Rin} + R_2 \times E_{Rout}). \quad (4)$$

In the open-loop allocation, a material-specific factor is introduced to allocate the production of virgin material. This factor depends on the market price of scrap or recycled material and the virgin material [12,13]. The expression of open-loop allocation is as follows [12] (cf. Eq. (5)):

$$E = (1 - R_1 + A_{open-loop} \times R_1) \times E_V - A_{open-loop} \times R_2 \times E_V^* + R_1 \times E_{Rin} + (1 - R_2) \times E_D. \quad (5)$$

$A_{open-loop}$ is the allocation factor in the approach of open-loop allocation, which is equal to the ratio between the global market value of scrap material or recycled material and the global market value of virgin material [13]

Finally, in the CFF approach, more thorough considerations are taken into account, including a material-dependent factor which can reflect the market reality of demand and supply of recycled materials. Such factor is used to allocate the production of virgin material, recycling process and the production of virgin material that could be avoided [12,15]. In addition to this, the quality degradation of material due to recycling is also taken into account within the modelling. A list of factors of allocation and quality of some commonly used recycled material are available in Annex C of the PEF guidelines [15]. The equation of CFF can be found below (cf. Eq. (6)):

$$E = (1 - R_1) \times E_V + R_1 \times \left[A_{CFF} \times E_{Rin} + (1 - A_{CFF}) \times E_V \times \frac{Q_{Sin}}{Q_P} \right] + (1 - A_{CFF}) \times R_2 \times \left[(E_{Rout} - E_V^* \times \left(\frac{Q_{Sout}}{Q_P} \right)) \right] + (1 - R_2) \times E_D. \quad (6)$$

A_{CFF} is the allocation factor in CFF, depending on the balance between the total supply and demand for the recycled material on the market, provided by the PEF guidelines.

Q_{Sin} is the quality of the input recycled material.

Q_{Sout} is the quality of the output recycled material at the EoL stage.

Q_P : is the quality of the virgin material.

The parameters used in this study are set out in [Appendix C](#). Different EoL modelling approaches may result in different LCA results with different impact on production decisions [12]. Some EoL modelling approaches are more oriented towards the promotion of increased content of recycled materials to be included in the assessed product, while others provide incentives to maximizing the amount of final waste sent to recycling at the EoL.

2.2.2 EoL modelling in guidelines

This sub-section provides a summary of general principles applicable to the EoL modeling. It is also intended to remind the recommendations with respect to the usage of the above six EoL modelling approaches in the guidelines.

The modeling of recycling shall comply with some basic principles of allocation in ISO 14044. Among such basic principles, “the sum of the allocated inputs and outputs of a unit process shall be equal to the inputs and outputs of the unit process before allocation” [4]. As far as recycling is concerned, this principle means actually that the sum of the inputs and outputs related to the product to be recycled and the recycled material is equal to the whole system before allocation [12]. Other basic principles state that a sensitivity analysis shall be made when there are a number of alternative allocation approaches. In present study, if the “most appropriate approach” is not likely to be identified, several approaches should be applied for sensitivity analysis to check the robustness of the results. In any case, the modelling shall follow a clear allocation approach that shall be documented and explained to ensure transparency [4,42].

Under the above principles, ISO 14044 [4] and ISO 14067 [13] identify two applicable approaches for recycling and reuse: the closed-loop allocation and the open-loop allocation. The closed-loop allocation is recommended to be applied to closed-loop systems, i.e. systems where the recycled material recovered at the EoL stage of a product system is reused for the same product system again. It also can be used to open-loop systems where there are no changes in the inherent properties of the recycled material. The open-loop allocation is applicable to open-loop systems where the recycled material has changed its inherent properties and is used in other product systems.

The British standards, called PAS 2050 [18] and the American standard, namely the GHG protocol [17], both recommend to use the closed-loop allocation (namely, closed-loop approximation, in both guidelines) for closed-loop system and open-loop system with recycled material maintaining the same intrinsic properties as virgin material. This is in line with the recommendation in the ISO series. However, for an open-loop system with changes in terms of inherent properties of the material, these two guidelines recommend using the simple cut-off approach (namely, recycled content method, in both guidelines) [17,18]. Besides, in most of these cases, the GHG protocol recommends using both approaches to check the robustness of the results. The GHG protocol is also open to the options proposed in other guidelines such as the open-loop allocation proposed in ISO 14067 [17].

The 50/50 method is recommended by the Nordic Guidelines on LCA [12]. Its feature of “equal share” fits well with the closed-loop recycling systems [41].

IEA-PVPS has published its own methodological guidelines on LCA in the field of PV [14]. Two EoL modelling approaches are recommended in these guidelines: the approach of cut-off with economic allocation (namely, cut-off, in IEA’s guideline) is generally set as a default option and closed-loop allocation (namely, end-of-life approach, in IEA’s guideline) is proposed for sensitivity analysis [14].

At the European level, attempts have been made for the EU to find a harmonized approach aiming at different types of products. The solutions include the approach of CFF proposed in the PEF guidelines [43], which can be applied to different EoL scenarios, including material recycling, energy recovery and final disposal. Being suitable for both closed-loop systems and open-loop systems in a consistent and reproducible way [43], the CFF also allows changes in the quality of the material after recycling to be accounted for and using specific allocation factors to reflect and balance the market demand and supply of materials. Its broad applicability and comprehensiveness lead to a quite complex equation (cf. Eq. (6)) [15,16].

3 Results and discussion

By applying six EoL modelling approaches, the environmental impact on climate change ranges from 45.3 to 59.3 kg CO₂-eq/m², the result for the Cumulative Energy Demand (CED) indicator from 569.2 to 715.4 MJ/m² and the impacts on abiotic depletion potential and ecotoxicity from 6.3×10^{-3} to 1.3×10^{-2} kg Sb-eq/m² and 5.7×10^3 to 7.1×10^3 CTUe/m² respectively (cf. [Fig. 6](#)). The LCA results of this study are clearly lower than those obtained from other references, the latter results in terms of impact on climate change ranging from 200 to 250 kg CO₂-eq for the manufacturing of 1 m² of perovskite/Si tandem modules [9,10,44]. Several reasons explain the differences between these results and those from other studies. The inventory applied in this study for the perovskite/Si tandem module has been adjusted with the support of experts in IPVF. As it is shown in [Table A1](#) in [Appendix A](#), the weight of aluminum frame, EVA and glass were adjusted to be much lower (nearly 1/2) compared to IEA-PVPS Task 12 to fit the specific design of the tandem module in this study (cf. [Tab. 2](#)). The Si consumption to produce 1 m² of wafer was adjusted to be 0.51 kg according to ITRPV (2021) [36], compared to 0.6 kg in Task 12 and 1.07 kg in eocinvent 3.6. Furthermore, the production of Si wafer and manufacturing of tandem are assumed to be in Norway and France, which further reduces the CFP of the tandem module by around 84 kg CO₂-eq/m² compared to modules produced in China. For more details, please refer to [Appendix A](#).

Regarding the comparison and analysis of the results of the different EoL modelling approaches, [Figure 6a](#) shows that the result for cut-off with economic allocation (59.3 kg CO₂-eq/m²) is the highest, followed closely by the simple cut-off (57.0 kg CO₂-eq/m²). The former is 31% higher than

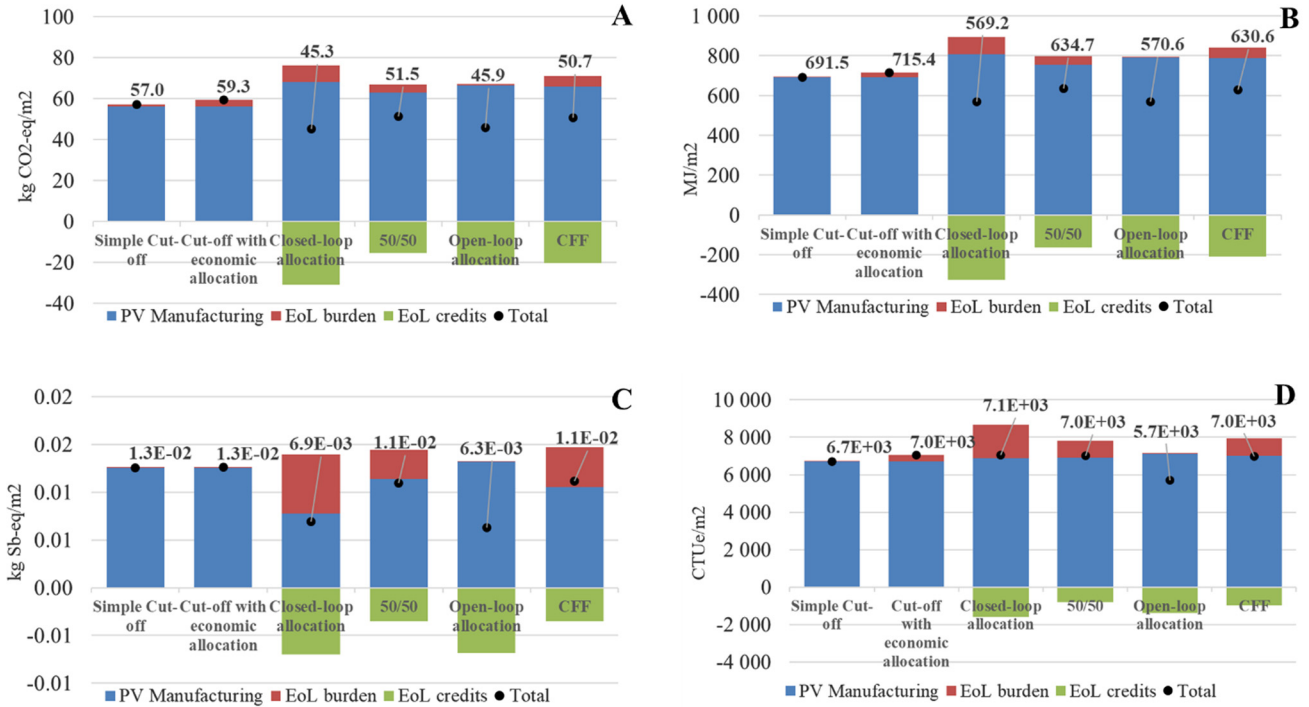


Fig. 6. LCA results of 1 m² of perovskite silicon tandem modules by applying the six selected EoL modelling approaches. A: climate change; B: cumulative energy demand (non-renewable); C: abiotic depletion potential (metals/minerals); D: ecotoxicity (fresh water).

that of the closed-loop approach (45.3 kg CO₂-eq/m²), which is the lowest within the climate change category. The other three EoL modelling methods lead to intermediate results. The result of CED (non-renewable, cf. Fig. 6b) shows same trends within the six EoL modelling approaches, and the result for the cut-off with economic allocation is 26% higher than that of the closed-loop allocation. The results for abiotic depletion potential and ecotoxicity show different trends from the former two impact categories (cf. Figs. 6c and 6d). The cut-off with economic allocation still provides the highest result, but it is the open-loop allocation which provides the lowest, in terms of impact of abiotic depletion potential. The closed-loop allocation represents the highest and the open-loop allocation the lowest impact for ecotoxicity.

The breakdown of the results has been made to facilitate the analysis. Firstly, the environmental impacts related to the EoL stage including the burden and credits are analyzed in Figure 6:

- The simple cut-off and cut-off with economic allocation only allocate partially the EoL burden to the PV module and do not allocate any EoL credits to the PV module. The EoL credits reflect, to some extent, the potential benefits of the recycling. Therefore, although in this study, a specific EoL treatment was designed to recover a maximum amount of valuable materials, i.e. to increase the recovery rate of material at the EoL (cf. R_2 in Tab. C1 in Appendix C), few benefits were passed on to the result. The increase of R_2 would even increase the impact when the cut-off with economic allocation is used.

- For the closed-loop approach, we can find that a high EoL burden is given to the PV module, but at the same time, a high credit which is equal to $-R_2 E_V^*$ is also given to the product (cf. Eq. (3) and Fig. D3). Combining the impact of burden and credits, when the environmental burden related to the EoL recycling (E_{Rout}) is clearly less than the potential burden avoided by the output recycled material (E_V^*), the increase of R_2 would lead to a decrease of the impacts in the closed-loop allocation. That is the case for the impact on climate change and CED.
- The 50/50 method, open-loop allocation and CFF approaches provide partial allocation of both the EoL burden and the EoL credits. As described in Section 2.2.1, the 50/50 method provides a half/half allocation to all types of materials, whereas the open-loop allocation and CFF approaches introduce different material-specific allocation factors. The allocation factors of materials used in this study can be found in Appendix C.

The modelling approaches not only have impact on the EoL stage, but also on the manufacturing when recycled material is contained in the product. The environmental burden of input material may be impacted by R_1 , the share of recycled material in the product.

- For simple cut-off and cut-off with economic allocation, the input recycled material only takes (partially) the burden of the upstream recycling process E_{Rin} (cf. Figs. D1 and D2). When the E_{Rin} is less than the burden of the production of virgin material E_V , the usage of recycled material, i.e., increasing R_1 would decrease the

impact of manufacturing, and consequently the total impact of PV modules. That is the case for the impact on climate change and CED.

- For the closed-loop allocation, according to equation (3), R_1 has no impact on the LCA results, which means that although the recycled material is used for the manufacturing, the environmental burden taken is still the same as for the virgin material E_V (cf. Fig. D3). Therefore, when the E_V is higher than E_{Rin} , e.g. the case for climate change and CED, the closed-loop allocation provides the highest impacts compared with other approaches in terms of manufacturing.
- The 50/50, open-loop and CFF approaches provide a partial allocation of both E_V and E_{Rin} to the input recycled material (cf. Figs. D4–D6). Therefore, in terms of manufacturing, these three approaches tend to provide intermediate results.

For the impacts on climate change and CED (cf. Figs. 6a and 6b), the burden related to EoL recycling E_{Rout} is much lower than the potential burden avoided by output recycled material E_V^* ; at the same time, the burden of the upstream recycling process to produce the recycled material in the product E_{Rin} is much lower than that of the production of the virgin material E_V . In the above cases, we can summarize that:

- The simple cut-off and cut-off with economic allocation are quite sensitive to R_1 , and the increase of R_1 can lead to a dramatical decrease of the result. The result is not sensitive to R_2 , as the burden related to EoL is quite low compared with the manufacturing.
- The closed-loop allocation is quite sensitive to R_2 , and the increase of R_2 can lead to a dramatical decrease of the result. R_1 has no impact on the result in closed-loop allocation.
- The result obtained using the 50/50 method, open-loop allocation and CFF is sensitive to both R_1 and R_2 , thus the increase of R_1 and R_2 leads to a decrease of the environmental impact.

Several important assumptions have been made in this study. The R_1 of each material was assumed based on the market mix in the Ecoinvent [32], which reflects the level of market average. The R_2 was assumed based on the specific recycling process in this study, which represents the best that can be technically achieved currently (cf. Tab. C1 in Appendix C). Therefore, the R_2 of each material is higher than the respective R_1 . Under the above assumptions, the closed-loop allocation leads to the lowest impacts, the cut-off with economic allocation and simple cut-off lead to the highest impact. However, if a different assumption would be made, e.g. R_1 of each material set as 100% and R_2 set as 0%, the simple cut-off and cut-off with economic allocation would provide the lowest impacts, the closed-loop allocation would provide the highest. In both cases, the closed-loop allocation and the two cut-off approaches tend to provide extreme results. The 50/50, open-loop and CFF approaches tend to provide the intermediate results.

In practice, the CFP and CED of the production of virgin material is usually higher than that of the recycling processes, especially for some common recyclable material such like aluminum, copper and glass. Therefore, in this

context, applying one of the cut-off approaches and the closed-loop allocation together seems to be enough to provide the whole range of results. This fits with the recommendation set in the GHG protocol [17], PSA 2050 [18] and PVPS Task 12 [14] in which these EoL modelling approaches are recommended simultaneously.

However, it is important to be prudent, especially when the impact related to the recycling is close or higher than the production of virgin material. For example, due to the usage of chemical solvent in the recycling processes of PV modules, E_{Rout} is higher than E_V^* in the impact of ecotoxicity. In such cases, the results of the different approaches do not depend only on the allocation of E_V^* and E_V , but also on the allocation of E_{Rin} and E_{Rout} , and the related allocation factor and quality degradation factor. The trend in these approaches varies on a case-by-case basis. Therefore, simply applying the cut-off approach and closed-loop allocation may not always provide us with an accurate range of LCA results, as was previously suggested. A more prudent way would be to ask for employing additional approaches for the sensitivity analysis.

While one specific EoL modelling approach cannot be identified as the best one to model the life cycle impacts of emerging PV modules in every context and for all impact categories, we recommend a representative approach that 1) can be suitable for a wide variety of material recycling systems, including closed-loop and open-loop, 2) avoids providing extreme results and 3) reflects, at least to some extent, the characteristics of the material. The CFF approach fits with all the above expectations. In addition, the CFF takes into account, in a comprehensive way, elements such as the material quality degradation in recycling and a material-specific allocation factor. The allocation factor and quality-degradation factor, as well as the R_1 and R_2 of some commonly used recycled material have been provided in Annex C of the PEF guidelines and reviewed by the EU and subject to continuous update [15]. The allocation factor defined in the CFF is a market-based factor which can balance, to some extent, the market demand and supply of the recycled material. For example, in the case of recycled aluminum, of which the demand is higher than the supply, the given factor will lead to a formula more sensitive to R_2 , i.e. promoting recycling of aluminum waste. Nevertheless, it is still important to recognize the weakness of the CFF. The different elements involved in CFF lead to a certain complexity and a balance needs to be found between the representativeness of the LCA results and the complexity of the work. Besides, not all materials and parameters are listed by PEF [15,16]. In such cases, the accurate parameters of the materials are hard to get and using the default values will lead to a partial loss of representativeness of the results. Nonetheless, we believe that the CFF provides a comprehensive modelling framework for recycling and that further development and improvement would be valuable. Some practical recommendations are given regarding the application of the six EoL modelling approaches. We recommend to apply some market average data if specific R_1 and R_2 data are unclear. They may be found in the Annex C of the PEF guidelines [15], as well as in the data of market mix in Ecoinvent database [32]. The economic factor in the approach

of cut-off with economic allocation needs additional information on the price of treatment of recycling and the output recycled material. The allocation factor in the open-loop allocation needs additional information on the price of the recycled material and virgin material. As mentioned previously, the allocation factor and quality-degradation factors of some material can be found in the Annex C of the PEF guidelines [15].

In this study, the potential impact avoided by the output recycled material at the EoL stage is directly counted as credit into or partially into the life cycle of the product in some modelling approaches to reduce the impact of the PV modules. As it has been discussed previously, these approaches would motivate the producers and recyclers to recycle more PV panels. However, whether such credits could be accounted for in the products is still under discussion, especially for the products having a long lifetime such as buildings [12,45]. One key critical point is whether the material used in the product can go immediately to recycling and then back to the market, which is not the case for PV modules. This effect is more evident when the lifetime of a product, in our case, the PV module, is longer. In Task 12, the closed-loop allocation (also named as end-of-life approach in Task 12) is one of the recommended methodologies for PV systems, but it is specified that the EoL burden and credits shall be reported separately [14,23]. Such reporting ensures the transparency of the study, but at the same time reduces the comparability of the results. An option could be to add these credits to the product to clearly incentivize the EoL recycling but introducing a lifetime-dependent factor to differentiate between products recycled within one year and those recycled after thirty years. However, to our knowledge, there is no dedicated study exploring this approach.

4 Conclusions

In this study, six different EoL modelling approaches were applied in the LCA of perovskite / Si tandem modules and PV recycling processes and their results were analyzed. These results demonstrate that a clear justification of the methodological choices related to the EoL modelling is essential to ensure the comparability and reliability of the LCA results. Based on this work, the following recommendations are made, for EoL modelling as part of an LCA of PV modules including the recycling stage:

- To choose the CFF approach as the baseline representative EoL modelling that best reflects the characteristics of the recycled material and avoids extreme results;
- To perform a sensitivity analysis to check the robustness of the results;

- To use at least a cut-off method and the closed-loop allocation in parallel for the sensitivity analysis, and to introduce additional approaches for the sensitivity analysis if the burden of recycling is high compared with the burden of production of virgin material.

These recommendations are intended to raise awareness among practitioners and share thoughts that will enable them to refine their work. Ultimately, this study should contribute to a more reliable evaluation of the benefits and challenges of the tandem module recycling, which may help to increase the competitiveness of such technology within the energy transition context.

This study is performed based on a case study of perovskite/Si tandem modules, in which the environmental impact of electricity is greatly reduced due to the location of the Si water production and manufacturing of the tandem modules. As a next step, further work will be done by introducing different scenarios and other sources of uncertainty to study the impact of the EoL modelling approaches at a global level.

Acknowledgments

The authors would like to thank Matthew Hull, George Wong and Lian Duan for insightful discussions. This research was supported at IPVF by the French Government in the frame of the program of investment for the future (Programme d'Investissement d'Avenir-ANR-IEED-002-01).

Funding

This research was funded by TotalEnergies and ANRT (Association Nationale de la Recherche et de la Technologie) with CIFRE (Contrat Industriel de Formation par la Recherche Convention N°2022/0344) and was supported at IPVF by the French Government in the frame of the program of investment for the future (Programme d'Investissement d'Avenir - ANR-IEED-002-01).

Conflicts of interest

The authors have nothing to disclose.

Data availability statement

The data reported in this article are openly available and can be found in the list of reference. Some data associated with this study cannot be disclosed due to legal/ethical/other reason.

Author contribution statement

Lu WANG and Paula PEREZ-LOPEZ contributed to the design and implementation of the research, Lu WANG contribute to the analysis of the results and to the writing of the manuscript, all authors commended and reviewed on the manuscript.

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Cite this article as: Lu Wang, Lars Oberbeck, Mathilde Marchand Lasserre, Paula Perez-Lopez, Modelling recycling for the life cycle assessment of perovskite/silicon tandem modules, *EPJ Photovoltaics* **15**, 14 (2024)

Appendix A: Adjustments of the LCI to IEA-PVPS (2020) to fit the design of SHJ and bifacial structure of tandem

Table A1. Key adjustments of the LCI to IEA-PVPS (2020) to fit the design of SHJ and bifacial structure of tandem.

Process block	Input	Adjusted value	Original value	Unit	Comment
Tandem module manufacturing	Rear glass – solar grade	5.10 [36]	8.10 [14]	kg/m ²	The thickness of the glass was adjusted from 4 mm to 2 mm [36].
	EVA	0.42 [36]	0.88 [14]	kg/m ²	The thickness of the encapsulant has been adjusted from 940 um to 450 um [36].
	Aluminium frame	0.95 [36]	2.13 [14]	kg/m ²	The weight of aluminum frame has been adjusted [36].
	Backsheet	Without	With		The bifacial structure of perovskite/Si tandem module does not contain the backsheet structure. The inventories of related components (polyvinylfluoride film, polyethylene terephthalate, PET polyethylene, HDPE) were removed.
	Electricity	14.00 [14]	14.00 [14]	kWh/m ²	Change for an electricity mix in France
SHJ cell manufacturing	Silver (front+rear)	3.40 [29]	7.30 [36]	g/m ²	The silver consumption related to SHJ single junction [36] is 7.3 g/m ² . In the tandem configuration, the bottom SHJ sub-modules receive less irradiation which implies a lower current. The silver consumption is adjusted to 3.4 g/m ² [29].
	ITO	1.90 [29]	3.10 [36]	g/m ²	The ITO consumption related to SHJ single junction [36] is 3.1 g/m ² . We applied a reduction of 40% following an opinion from an expert in PV [29].
	Electricity	11.90 [37]	17.70 [14]	kWh/m ²	The consumption of electricity has been adjusted based on Louwen (2015) [37]. Change for an electricity mix in France
Single-Si wafer manufacturing	Single-Si	0.51 [36]	0.60 [14], 1.07 [32]	kg/m ²	The consumption of single-Si for manufacturing of 1m ² of wafer has been adjusted to 0.51 kg/m ² [36]. This adjusted value is much lower than the one in Ecoinvent 3.6 [32] ‘single-Si wafer production, photovoltaic – RER – single-Si wafer, photovoltaic’: 1.07 kg/m ² .
	Electricity	4.76 [14]	4.76 [14]	kWh/m ²	Change for an electricity mix in Norway
Single-Si ingot production by Czochralski	Electricity	32.00 [14]	32.00 [14], 85.6 [32]	kWh/kg	This adjusted value is much lower than the one in Ecoinvent 3.6 [32] ‘silicon production, single crystal, Czochralski process, photovoltaics’: 85.6 kWh/kg. Change for an electricity mix in Norway
Solar-grade Si production	Electricity	49.10 [14]	49.10 [14], 109.89 [32]	kWh/kg	This adjusted value is much lower than the one in Ecoinvent 3.6 [32] ‘silicon production, solar grade, modified Siemens process’: 109.89 kWh/kg. Change for an electricity mix in Norway

Appendix B: LCI of the EoL treatment of perovskite/Si tandem modules

Table B1. LCI of the EoL treatment of perovskite/Si tandem modules.

EoL treatment of 1 kg of perovskite/Si tandem modules								
Process block	Input/Output	Value	Unit	Input technosphere / biosphere flows inecoinvent			Database	Comment
				Product	Activity	Location		
Delamination	Electricity	3.6E-02	kWh	Electricity, medium voltage	Market for electricity, medium voltage	FR	Ecoinvent 3.6 Cut_off	Delamination based on the flashlight technology from FLAXRES [26]
	Electricity	7.9E-02	kWh	Electricity, medium voltage	Market for electricity, medium voltage	FR	Ecoinvent 3.6 Cut_off	
Pyrolysis	Nitrogen	6.2E-03	kg	Nitrogen, liquid	Market for nitrogen, liquid	RER	Ecoinvent 3.6 Cut_off	The inventory data of pyrolysis is from ROSI [38], including pyrolysis and the waste gas treatment.
	Natural gas	4.9E-03	Nm ³	Natural gas, high pressure	Natural gas, high pressure, import from NL	FR	Ecoinvent 3.6 Cut_off	
	Air	3.6E-01	kg					
	NaOH	1.9E-01	kg	Sodium hydroxide, without water, in 50% solution state	Chlor-alkali electrolysis, membrane cell	RER	Ecoinvent 3.6 Cut_off	
	Ca(OH) ₂	1.3E-03	kg		Market for lime	RER	Ecoinvent 3.6 Cut_off	
SHJ section chemical treatment	KOH	1.4E-01	kg	Potassium hydroxide	Market for potassium hydroxide	GLO	Ecoinvent 3.6 Cut_off	The chemical treatment process from ROSI is based on p-type Si wafer [46]. The inventory of treatment for SHJ wafer was adjusted based on Sah (2022) [47] and Zhang (2021) [48] with the support of expert from IPVF.
	H ₂ SO ₄	1.5E-01	kg	Sulfuric acid	Market for sulfuric acid	GLO	Ecoinvent 3.6 Cut_off	
	Deionised water	5.0E-01	kg	Water, deionised	Market for water, deionised	Europe without Switzerland	Ecoinvent 3.6 Cut_off	
Perovskite section	HNO ₃	2.0E-04	kg	Nitric acid, without water, in 50% solution state	Market for nitric acid, without water, in 50% solution state	RER	Ecoinvent 3.6 Cut_off	Chen (2021) [21]
	DMF	5.1E-03	kg	N,N-dimethylformamide	Market for N,N-dimethylformamide	GLO	Ecoinvent 3.6 Cut_off	
	Resin	1.6E-03	kg	Cationic resin	Market for cationic resin	RoW	Ecoinvent 3.6 Cut_off	
	DCB	3.2E-04	kg	O-dichlorobenzene	Market for o-dichlorobenzene	RER	Ecoinvent 3.6 Cut_off	
Second treatment	Ag refining	2.6E-04	kg	Silver	Treatment of precious metal from electronics scrap, in anode slime, precious metal extraction	SE	Ecoinvent 3.6 Cut_off	The value is calculated by the quantity of Ag in tandem and the recovery rate of 100% [38] (cf. Tabs. 2 and C1).
	Al refining	7.6E-02	kg	Aluminium, cast alloy	Treatment of aluminium scrap, post-consumer, prepared for recycling, at refiner	RER	Ecoinvent 3.6 Cut_off	The value is calculated by the quantity of Al in tandem and recovery rate of 97% [38] (cf. Tabs. 2 and C1).
	Cu (cables)	3.6E-03	kg	Copper	Treatment of used cable	GLO	Ecoinvent 3.6 Cut_off	The value is calculated by the quantity of Cu in cables and recovery rate of 100% [38] (cf. Tabs. 2 and C1).
	Cu (interconnection)	8.4E-04	kg	Copper	Treatment of copper scrap by electrolytic refining	RER	Ecoinvent 3.6 Cut_off	The value is calculated by the quantity of Cu in interconnection and recovery rate of 76% [38] (cf. Tabs. 2 and C1).
	Glass remelting	7.9E-01	kg	Please refer to Table B3				The value is calculated by the quantity of glass in tandem and recovery rate of 93% [38] (cf. Tabs. 2 and C1).
Waste treatment	Waste plastic	-7.0E-03	kg	Waste plastic, mixture	Market for waste plastic, mixture	FR	Ecoinvent 3.6 Cut_off	The value is calculated by the quantity of polymer in cables (cf. Tab. 2).
	Waste water	-6.5E-01	kg	Waste water, average	Treatment of wastewater, average, capacity 4.7E10l/year	CH	Ecoinvent 3.6 Cut_off	The sum of wastewater from pyrolysis [38] and chemical treatment [21]

Table B2. Avoided material from EoL treatment of 1 kg of perovskite/Si tandem modules.

Avoided material from EoL treatment of 1 kg of perovskite/Si tandem modules							
Material	Value	Unit	Technosphere flows in ecoinvent				Comment
			Product	Activity	Location	Database	
Ag	2.6E-04	kg	Silver	Silver-gold mine operation with refinery	RoW	Ecoinvent 3.6 Cut_off	The value is calculated by the quantity of Ag in tandem and the recovery rate of 100% [38] (cf. Tabs. 2 and C1).
Al	7.6E-02	kg	Aluminium, cast alloy	Aluminium ingot, primary, to aluminium, cast alloy market	GLO	Ecoinvent 3.6 Cut_off	The value is calculated by the quantity of Al in tandem and recovery rate of 97% [38] (cf. Tabs. 2 and C1).
Cu (cables)	3.6E-03	kg	Copper	Copper production, primary	RER	Ecoinvent 3.6 Cut_off	The value is calculated by the quantity of Cu in cables and recovery rate of 100% [38] (cf. Tabs. 2 and C1).
Cu (inter-connection)	8.0E-04	kg	Copper	Copper production, primary	RER	Ecoinvent 3.6 Cut_off	The value is calculated by the quantity of Cu in interconnection and recovery rate of 76% [38] (cf. Tabs. 2 and C1).
Glass	7.9E-01	kg	Flat glass, uncoated	Flat glass production, uncoated	RER	Ecoinvent 3.6 Cut_off	The value is calculated by the quantity of glass in tandem and recovery rate of 93% [38] (cf. Tabs. 2 and C1).
Pb	1.2E-05	kg	Lead	Primary lead production from concentrate	GLO	Ecoinvent 3.6 Cut_off	The value is calculated by the quantity of lead in tandem and recovery rate of 99% [21] (cf. Tabs. 2 and C1).
Si (2N)*	2.5E-02	kg	Silicon, metallurgical grade	Silicon production, metallurgical grade	RoW	Ecoinvent 3.6 Cut_off	The value is calculated by the quantity of silicon in tandem and recovery rate of 98% [49,28,38,50] (cf. Tabs. 2 and C1).

*According to the report from ROSI [38], the quality of recovered Si can reach a quality of 5N. However, the downstream value chain of recovered 5N-Si is still unclear. Due to the data availability, at this stage, the credits of 2N-Si was assumed in this study.

Table B3. Remelting of 1 kg glass cullets to produce flat glass.

Remelting of 1 kg glass cullets to produce flat glass								
Process block	Input/Output	Value	Unit	Technosphere flows in ecoinvent				Comment
				Product	Activity	Location	Database	
Glass remelting	Glass cullets	1	kg					
	Heat	4.86	MJ	Heat, district or industrial, natural gas	Heat and power co-generation, natural gas, conventional power plant, 100MW electrical	FR	Ecoinvent 3.6 Cut_off	Fabrication of glass from glass cullets can save 30% of heat compared with primary glass [51].
	Electricity	0.11	kWh	Electricity, high voltage	Market for electricity, high voltage	FR	Ecoinvent 3.6 Cut_off	

Appendix C: Key parameters and assumptions applied in EoL modelling of perovskite/Si tandem modules

Table C1. Key parameters and assumptions applied in EoL modelling of perovskite/Si tandem modules.

Material	R_1^a	R_2^b	Cut-off with economic allocation ^c A_e^{ca}	Open-loop allocation $A_{open-loop}^d$	CFF ^e		
					A_{CFF}	Q_{sin}/Q_p	Q_{sout}/Q_p
Al	74%	97% ^{ba}	0.73 ^{ca}	0.8	0.2 [15]	1 [15]	1 [15]
Glass	0%	93% [38]		0.5	0.2 [15]	1 [15]	1 [15]
Cu (cables)	20%	100% [38]		0.5	0.2 [15]	1 [15]	1 [15]
Cu (interconnection)	20%	76% [38]		0.8	0.2 [15]	1 [15]	1 [15]
Ag	8%	100% [38]		1	0.2	1 [15]	1 [15]
Si (2N)	0%	54% ^{bb}	1	0.5	1	1	
Lead	55%	99% [21]		0.8	0.5	1	1

^aThe recycled content ratio R_1 means the share of recycled material in each input material. The value of each material is assumed based on the data of market mix in the Ecoinvent database [32], which reflects the level of market average.

^bThe recovery rate of R_2 is the ratio between the recovered material and the material used for the manufacturing. The value of each material is assumed based on the PV recycling process in this study. The recovery rate represents the best that can be technically achieved currently.

^{ba}The R_2 of Al is assumed to be 97%. The Al frame could be fully recovered, but the secondary treatment of Al scrap leads to a final recovery rate of 97% for Al.

^{bb}In this study, around 98% of Si in the modules was assumed to be recovered, i.e. 0.025 kg of Si per kg of modules. Due to the loss of Si in the manufacturing, the Si consumed to produce 1 kg of tandem modules is 0.047 kg. Therefore, the R_2 for Si was assumed to be 54%.^c In the EoL treatment processes, the secondary treatment processes such like the Al refining are more related to the production of recycled material. The chemical treatment of the SHJ section and perovskite could be also considered as a secondary treatment of the silicon waste and glass waste respectively. Therefore, in this study, these treatment processes were excluded from the EoL stage of the tandem modules when modelling recycling by the approach of cut-off with economic allocation. The disassembly, delamination and the pyrolysis processes were allocated between the EoL of modules and the recyclable material, i.e. Al waste, copper scrap, waste glass and ferro-Si.

^{ca}The allocation factor 0.73 is the share allocated for the PV modules. It was calculated based on the information in Table D1.

^dThe allocation factor of open-loop allocation in this study is the ratio between the market price of the recycled material and virgin material. The value of $A_{open-loop}$ varies as the market price changes. In this study, the approximate values were assumed depending on the publicly available information [52–54]. The price of the recycled glass from PV after remelting is unknown due to the lack of downstream value chain. Whether the recycled glass could be reused as solar glass is not clear, and due to the traces of metal (e.g. Ti) doped in the glass, the usage of recycled glass would be limited. Therefore, a value of 0.5 was given to the recycled glass. The downstream value chain for the recycled Si is also unknown. However, as described in Appendix B, the purity of Si is higher than 2N, so the recycled Si could be sold at least at the same price as for 2N-Si. Therefore, a value of 1 was assumed for the recycled Si.

^eThe A_{CFF} , Q_{sin}/Q_p , Q_{sout}/Q_p of Al, Cu and glass in the approach of CFF could be found in the Annex C of the PEF guide. The A_{CFF} of Ag is assumed to be 0.2 according to the description of factor in PEF. A default value of 0.5 was assigned to silicon and lead. Q_{sin}/Q_p and Q_{sout}/Q_p are assumed to be 1 for Ag, silicon and lead, as no specific quality degradation was found.

Table C2. Mass and price used to calculate the allocation factor of the approach of cut-off with economic allocation.

Process/Material	Mass kg	Price €/kg	Allocation factor –
PV modules EoL treatment	1.00	0.275 [23]	0.73
Al scrap	0.08	0.700 [23]	0.15
Cu scrap	0.004	4.000 [23]	0.04
Waste glass	0.85	0.020 [23]	0.05
Ferro-Si	0.03	0.409 [28]	0.03

Appendix D: Schema of description of six EoL modelling

The description of the six EoL modelling approaches are shown in [Figures D1–D6](#).

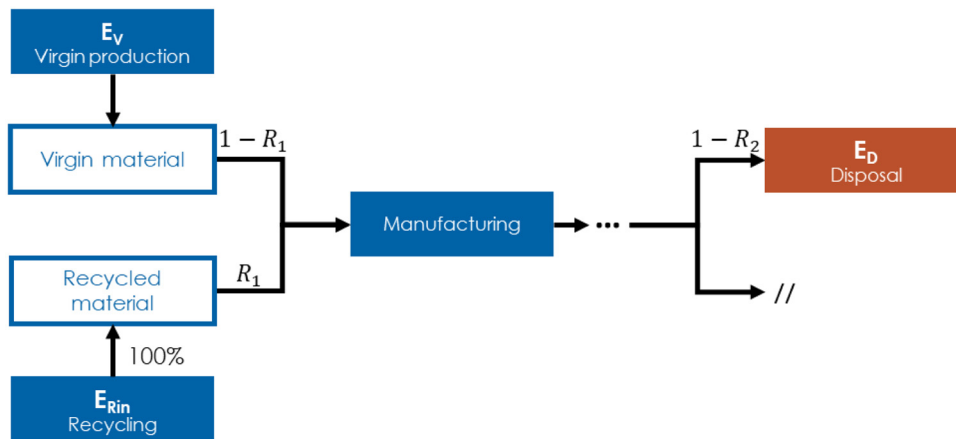


Fig. D1. Schema of simple cut-off.

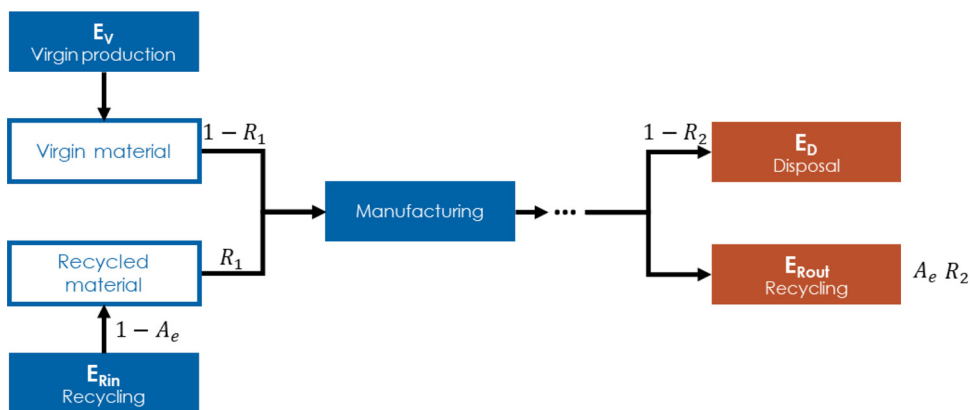


Fig. D2. Schema of cut-off with economic allocation.

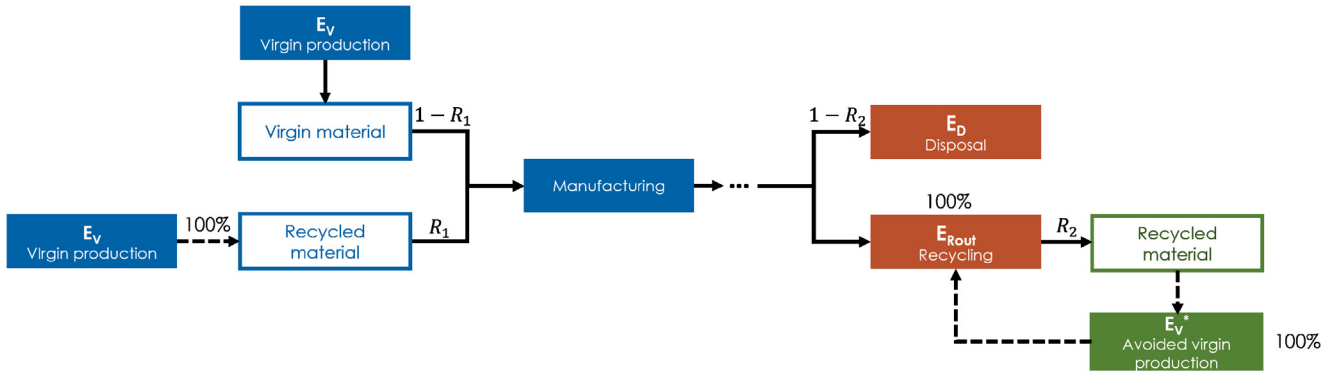


Fig. D3. Schema of closed-loop allocation.

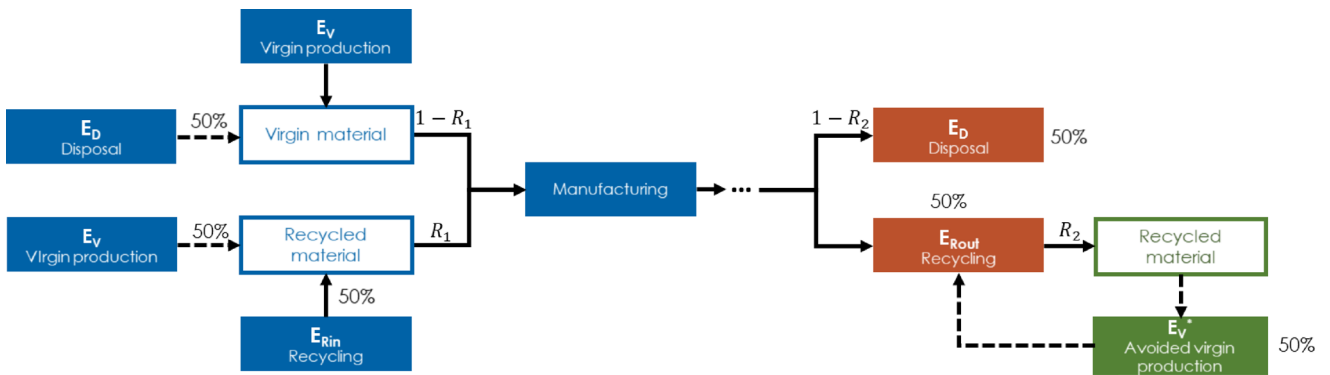


Fig. D4. Schema of 50/50 approach.

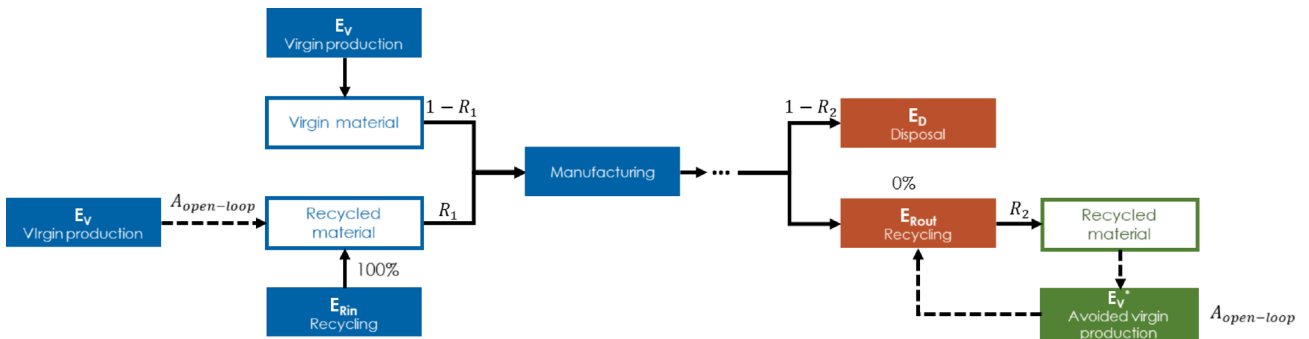


Fig. D5. Schema of open-loop allocation.

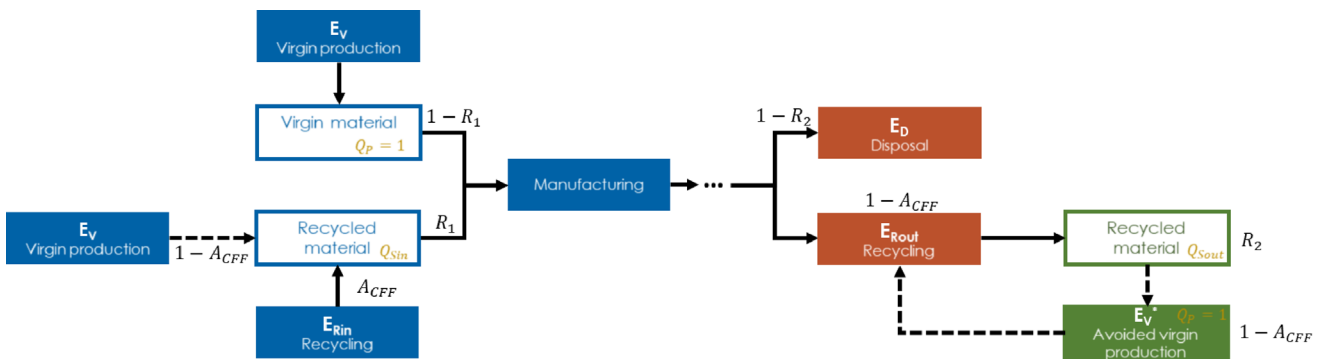


Fig. D6. Schema of CFF.