Life Cycle Assessment of Two Experimental Recycling Processes for c-Si Solar Modules

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Abstract

In this work we calculate and compare the environmental impacts of two experimental recycling processes for c-Si PV modules using organic (toluene - C_7H_8) and inorganic (nitric acid - HNO_3) solvents. The environmental impacts are estimated following the LCA methodology using available LCI data from the Ecoinvent database (version 3.2) complemented with experimental data from the Laboratory of Corrosion, Protection and Recycling of Materials (LACOR - Brazil) and the School of Photovoltaic and Renewable Energy Engineering at UNSW Sydney. Results show that electricity from non-renewable sources has the largest contribution to the environmental impacts of both recycling routes, which might be reduced with the use of renewable sources of electricity. Although the comparative impact of the used chemicals is small, they still represent harm to humans and the environment, toluene in particular.

Keywords: PV modules, Recycling, Life Cycle Assessment, LCA.

1. Introduction

The global market of solar photovoltaic (PV) modules has grown by 40% (compound annual rate) from 1997 (114.1-MWp) to 2017 (93.9-GWp) (Mints 2018), and approximately 90% of the current PV market is captured by crystalline silicon (c-Si) technologies (ITRPV 2018). This significant growth in solar energy is critical in the search for a more sustainable economy. However, this global increase in PV installations will also produce a substantial amount of waste. Predictions show that PV will generate a significant share of global electricity and that the corresponding future waste will account for up to 8.0 million tonnes annually by 2030 (Weckend, Wade et al. 2016).

Regulations for this type of waste are still being developed worldwide. The most comprehensive laws and policies addressing the end-of-life (EoL) management of PV waste are from the European Union, which includes PV waste into the Waste of Electrical and Electronic Equipment (WEEE) directive since 2012 (Shin, Park et al. 2017). It is essential that the PV industry develops the knowledge and capability required to deal with EoL modules to maintain its position as a sustainable technology (de Wild-Scholten, Wambach et al. 2005). It is expected that increasing progress in cost-effective recycling technologies will encourage its competitiveness and reduce the recycling process' environmental impacts, to which regulations can be a facilitator (Bombach, Röver et al. 2006, Kang, Yoo et al. 2012, Tao and Yu 2015).

The main components of a standard c-Si module (apart from the junction box) are a glass cover, aluminium (Al) frame, encapsulant (normally ethylene vinyl acetate, EVA), c-Si wafers, polymer-based backsheet (commonly Tedlar®) and metal contacts and ribbons (Dias, Benevit et al. 2016). By weight, a c-Si module contains about 76% glass (front cover), 10% polymer (encapsulant and backsheet foil), 8% Al (mostly the frame), 5% silicon (solar cells), 1% copper (Cu) (interconnectors) and less than 0.1% silver (Ag) (contact lines) and other metals

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(mostly tin and lead) (Wambach, Schlenker et al. 2006, Sander and Politik 2007). Figure 1 shows a common Si-based module structure.

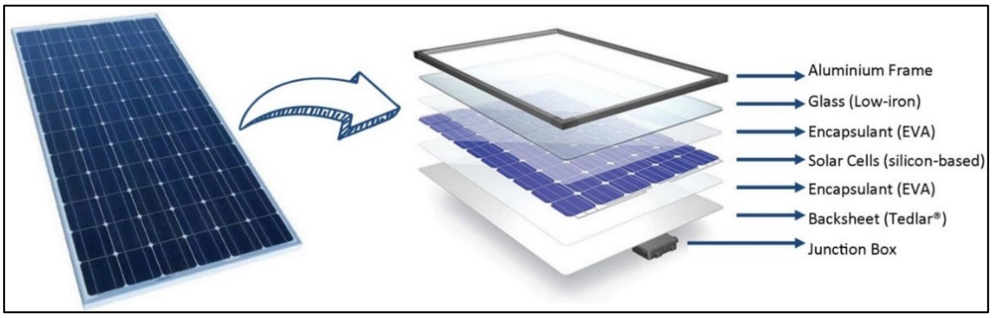

Figure 1: Common Si-based module structure and materials.

Recovering these materials is beneficial as they could be re-introduced into the supply chain of solar modules or other industries. Besides, some of the elements, particularly metals, are valuable or scarce (Stephanie Weckend 2016). Ag, for example, is the most expensive element per unit of mass of a c-Si panel (IEA-PVPS 2016), which led the solar industry to reduce the amount of Ag used in PV modules for many years. The reduction of metals and other components can be advantageous regarding cost reductions in PV modules. However, the industry hasn't been very active in reusing or recycling parts of EoL PV systems. Nevertheless, is has been shown that recycling processes for PV modules can recover glass, Si, Al, Ag, Cu and other materials at sufficient quality for sale on the world market (Stephanie Weckend 2016).

Along with the search for recycling processes for PV modules, there is also a concern with the potential environmental impacts associated with these recycling methods, such as from the use of chemicals or large amounts of energy from non-renewable sources. Hence, it is essential to analyse the materials and energy involved in PV recycling processes to evaluate their sustainability (Twidell and Weir 2015). In this study, Life Cycle Assessment (LCA) will be used for this purpose. LCA is an analytical methodology used to assess any product or process from an environmental perspective. It collates and analyses information from the whole product/process life cycle, considering inputs and outputs as energy, materials, wastes and emissions (Owens 1997). Life cycle information can be used to inform the public sector, stakeholders, manufacturers, and in the design and implementation of new policies.

This report presents two different approaches for recycling solar modules (at a laboratory scale) and the environmental analysis of these processes. The results from the experiments are analysed and discussed in order to understand the benefits of these processes in terms of reduction of environmental impacts from the recovery of materials from c-Si solar modules.

2. Method

The chosen procedures evaluated in this study are chemical-thermal separation processes using different organic and inorganic solvents (Lunardi, Alvarez-Gaitan et al. 2018). Data concerning the energy and labour input are recorded, as well as material inputs and outputs (the outcome of the recycling process), required for the LCA methodology in order to calculate the environmental impacts of both processes. The final environmental impacts are calculated using 3 endpoint indicators based on the ReCiPe method (Figure 2) (Mark A.J. Huijbregts, Zoran J.N. Steinmann et al. 2016).

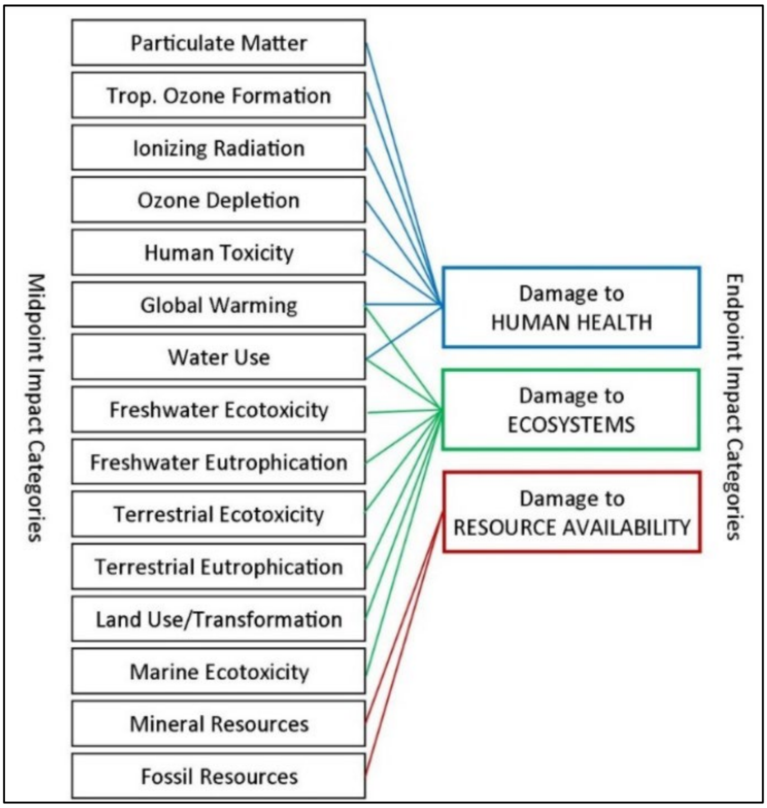

Figure 2: Overview of structure of environmental impacts using the ReCiPe method.

To estimate the environmental impacts from these processes, a life cycle inventory (LCI) is created with a mix of existing LCIs, including Ecoinvent database version 3.2 (Hedemann, König et al. 2007), experimental data from the Laboratory of Corrosion, Protection and Recycling of Materials (LACOR - Brazil) (Dias, Javimczik et al. 2016), and the UNSW SPREE laboratories (Sydney - Australia) (Zhang 2016). The inventory from our experiments is presented in Section 3. Based on these experiments, the LCI considers all materials and energy consumption, as well as the efficiency and impacts of recovering the metals and other materials from both recycling processes. The results from the LCA will provide the basis to assess both recycling processes and provide a recommendation based on the environmental impacts.

The total time, energy consumed, materials used, and solvents (Experiment 1: nitric acid - HNO_3 and Experiment 2: toluene - C_7H_8) recovered were measured for both experiments and are described in this section. However, the electricity mix considered is for China, as it remains the biggest producer and customer for Si cells and modules (Hutchins 2018) and its electricity mix is based on approximately 70% in fossil fuels (coal based) (West 2017). In 2016, generation from combustible fuels accounted for 67.3% of total world gross electricity production. Combustible fuels include coal and coal products, oil and oil products, natural gas, biofuels (solid biomass and animal products), gas/liquids from biomass, industrial waste and municipal waste (IEA 2018).

This LCA study considers one EoL PV module as the functional unit (size: 0.053 m^2). Because the module used in Experiment 1 is smaller than the module used in Experiment 2 (as shown in Table 1), all Experiment 1 inventory data has been scaled up to match the size and weight of the different solar modules.

Table 1: Mini-module specifications – Experiments 1 and 2.

	Sample dimensions (cm)	Sample area (m^2)
Experiment 1 (without frame)	22.0 x 22.0	0.048
Experiment 2 (without frame)	21.5 x 24.5	0.053

3. Experiment description and Life Cycle Inventory

The first step for experiments 1 and 2 is the preparation of the c-Si PV modules by manually removing the junction box and the Al frames.

In Experiment 1 a thermal process is first used to separate the EVA from the wafers (Zhang, Pahlevani et al. 2018). The thermal delamination process is based on separating module layers through the decomposition of EVA and backsheet under high temperature. The thermal treatment offers the potential for recovering the main components and materials of a PV module close to their original state (i.e. unbroken and undamaged cells). Then, the chemical treatment for the wafers is conducted in an ultrasonic bath with HNO_3 at approximately 40°C, to create micro-stirring and to promote the leaching of doped metal ions such as Ag, Al and Cu, in order to recover the metals from the c-Si cells and reuse them. The inventory data for Experiment 1 is shown in Table 2.

Table 2: Inventory table for Experiment 1.

<table>
<tr><th>Process Step</th><th>Material/Product</th><th colspan="2">Quantity</th><th colspan="2">Unit</th></tr>
<tr><td rowspan="7">Preparation – Frameless module</td><td colspan="5">Input</td></tr>
<tr><td></td><td colspan="2"></td><td colspan="2"></td></tr>
<tr><td>Waste Module</td><td colspan="2">266</td><td colspan="2">g</td></tr>
<tr><td>Frame</td><td colspan="2">0</td><td colspan="2">g</td></tr>
<tr><td>Junction Box</td><td colspan="2">0</td><td colspan="2">g</td></tr>
<tr><td colspan="5">Output</td></tr>
<tr><td>Waste Module</td><td colspan="2">266</td><td colspan="2">g</td></tr>
<tr><td rowspan="6">Step 1 - Thermal separation</td><td colspan="5">Input</td></tr>
<tr><td>Waste Module</td><td colspan="2">266</td><td colspan="2">g</td></tr>
<tr><td>Electricity</td><td colspan="2">32.83</td><td colspan="2">kWh</td></tr>
<tr><td colspan="5">Output</td></tr>
<tr><td>Glass (intact) + Tabbing Material</td><td colspan="2">156</td><td colspan="2">g</td></tr>
<tr><td>Silicon Wafer (broken)</td><td colspan="2">110</td><td colspan="2">g</td></tr>
<tr><td rowspan="9">Step 2 - Chemical separation (nitic acid + ultrasonic bath)</td><td colspan="5">Input</td></tr>
<tr><td>Silicon Wafer (broken)</td><td colspan="2">110</td><td colspan="2">g</td></tr>
<tr><td>Electricity</td><td colspan="2">13.78</td><td colspan="2">kWh</td></tr>
<tr><td>3M Nitric Acid (HNO_3)</td><td colspan="2">20</td><td colspan="2">ml</td></tr>
<tr><td colspan="5">Output</td></tr>
<tr><td>Silicon Wafer (broken)</td><td colspan="2">109.98</td><td colspan="2">g</td></tr>
<tr><td>Aluminium</td><td>20445.00</td><td>µg</td><td>0.02044</td><td>g</td></tr>
<tr><td>Silver</td><td>811.50</td><td>µg</td><td>0.00081</td><td>g</td></tr>
<tr><td>Copper</td><td>16.20</td><td>µg</td><td>1.62 x 10^{-5}</td><td>g</td></tr>
</table>

In Experiment 2, after the manual removal of the frame and junction box, binder clips are placed on the edges of the module to maintain mechanical pressure during the chemical reaction (EVA separation). The next step is to put the modules in a steel container covered by a glass panel. The system is filled with C_7H_8 until the modules are completely immersed and the system, which is placed on a heating plate controlled by a thermocouple, is kept at 90ºC for four days. After every 24 hours, the system is opened, the C_7H_8 is weighed and recovered, and the modules are visually inspected. After the completion of the (four days) processes, the different components are manually separated and weighed individually. The goal of this process is to separate the materials and other materials, such as glass, in order to recover them and use them in new c-Si solar cells and modules. Table 3 shows the inventory data.

Table 3: Inventory table for Experiment 2.

Process Step	Material/Product	Quantity	Unit
Preparation- Mechanical separation	Input		
	Waste Module	689.77	g
	Output		
	Frame	166.00	g
	Junction Box	10.64	g
	Waste Module S1	513.13	g
Step 1 - Chemical separation (toluene + heating plate)	Input		
	Waste Module S1	513.13	g
	Energy	25.56	kWh
	Toluene (C_7H_8)	1.96	kg
	Output		
	Waste Module S2	29.79	g
	Backsheet	18.21	g
	Superstrate (glass)	415.77	g
	EVA + cell	17.35	g
Step 2 - Manual separation	Input		
	Waste Module S2	29.79	g
	Output		
	Waste Module S3	1.18	g
	Backsheet	4.32	g
	EVA + cell	24.29	g

4. Experiment Results

As mentioned previously, the objective of Experiment 1 is to use HNO_3 to achieve a chemical leaching of metals from the solar cells, which depends on the concentration of HNO_3 used. The reaction under ultrasonic bath was rapid and strong. A significant development of bubbles occurred during the chemical reaction and there was an increase in the temperature and apparent visual changes in the surface of the samples were observed (the metal conduct lines on the top surface and Al on the back surface almost disappeared). Figure 3 shows the main results for Experiment 1.

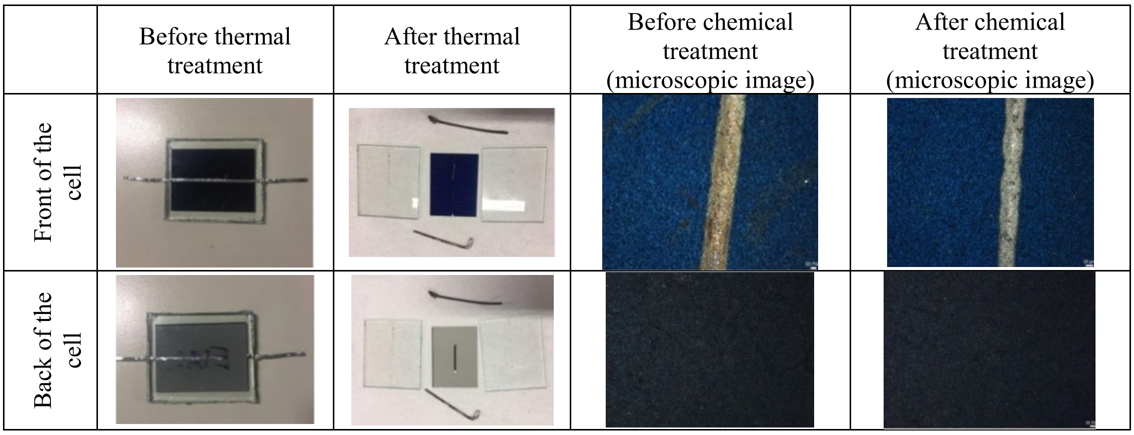

Figure 3: Results from Experiment 1, considering the thermal and the chemical treatments.

Experiment 1 demonstrated that HNO_3, with the aid of high temperature and the ultrasonic bath, can recover high amount of Ag, Al and Cu from used PV wafer (Zhang 2016). Tests with other possibilities of recycling processes were made such as under normal conditions (without ultrasonic bath) and using a hot plate instead of the ultrasonic bath. The extraction rate of all targeted metals (Ag, Al and Cu) is improved significantly using ultrasonic bath, which is the focus of this LCA study. Especially for Ag, the extraction rate with ultrasonic bath was increased by one hundred times when compared with the extraction rate without it (Zhang 2016).

The results for Experiment 2 are shown in Figure 4, which presents the different components manually separated after the last (fourth) immersion into the toluene solution. A benefit of this method is that unlike in Experiment 1, the module backsheet is also recovered, instead of being incinerated.

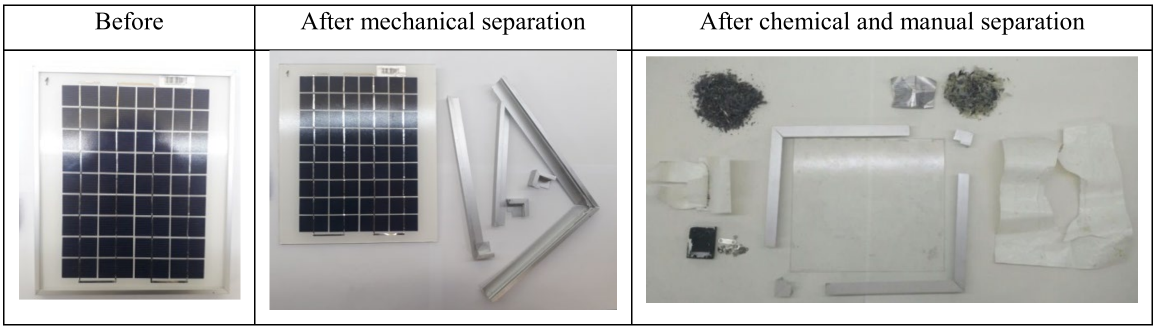

Figure 4: Different PV components separated using organic solvent (Experiment 2).

As Figures 3 and 4 show, the two experiments were conducted successfully and all the required data was collected. Experiment 1 showed good outcomes, recovering different metals from the cells through a chemical process. Experiment 2 also presented good results, being able to separate most of the materials from the solar modules. In both cases, the materials recovered could be used as raw materials in the production of new solar cells and modules, resulting in a more environmentally friendly process (Ardente, Latunussa et al. 2019).

The LCI was built with the data from both experiments, including material usage, energy consumption, emissions and wastes from each step of the process. The next step is to conduct the impact assessment using the ReCiPe methodology and compare these two experiments, so to assess which steps of these processes have the most significant impacts and how these effects can be minimised.

5. LCA Results and Discussion

This LCA uses the ReCiPe method as it is usually beneficial to aid the understanding of environmental outcomes if the target audience is not an expert in this field. The results for Experiments 1 and 2 are shown in Figure 5, where, for Experiment 1, step 1 is the thermal separation and step 2 is the chemical separation (nitic acid + ultrasonic bath), as described in Table 2.

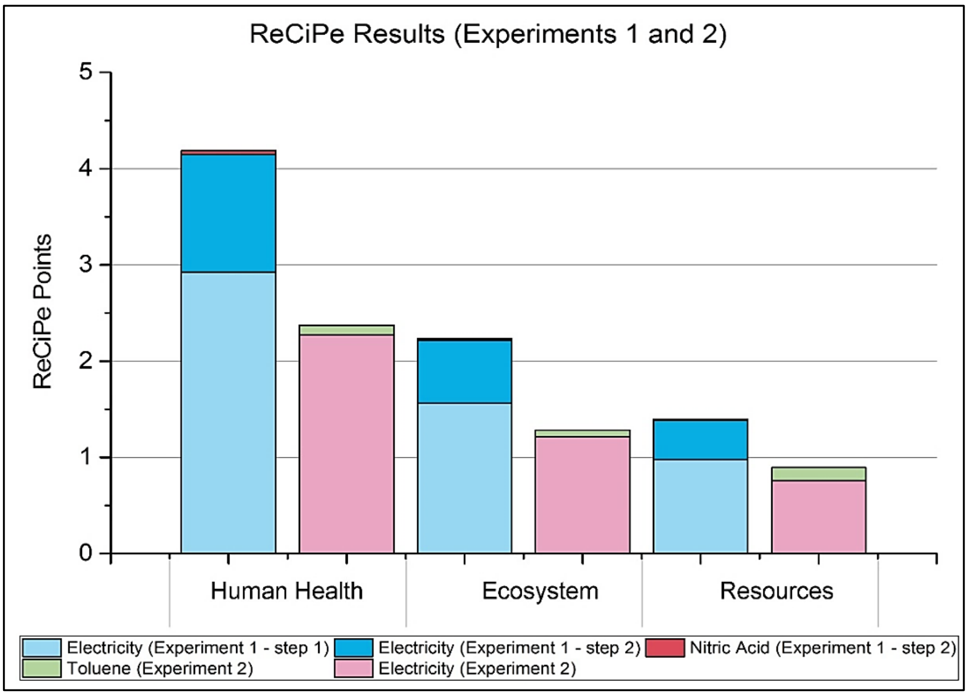

Figure 5: ReCiPe results (Human Health, Ecosystem and Resources) for Experiments 1 and 2.

Figure 5 shows that electricity has the most significant impacts in all three ReCiPe impacts, which incorporate midpoint impacts such as global warming and usage of fossil resources. This is because the electricity mix used in this LCA (from China) is predominantly from fossil fuels, as this is still the most common energy source used worldwide. Besides global warming, the usage of fossil resources is also depletes non-renewable resources such as iron ore, crude oil and coal, which is another significant impact related to the energy consumption. The ultrasonic technology requires the use of electricity, which generates significant environmental impacts, mostly if industrial scale is considered. However, the best results for recovering metals were achieved using this method. In order to overcome this issue, an alternative to increase the ecological benefits, besides recovering materials, is to use electricity from renewable energies. This modification in the inputs of the process would reduce the impacts from electricity significantly, not just for the ultrasonic bath, but for all steps of both processes (Experiments 1 and 2) (Reich, Alsema et al. 2011).

Most of LCA studies about PV cells and modules' production assume coal-fired power plants as the electricity input, as this is the current reality of PV manufactures such as China and the USA (IEA 2018). However, if the LCA assumption for electricity supply is only renewable sources such as hydro- or wind-power or PV, the environmental impacts associated to the PV production are reduced strongly (Reich, Alsema et al. 2011). It was shown in the literature that, assuming low-carbon (including nuclear and renewables) electricity input in the PV module production gives 0.06–1.6 $gCO_{2\text{-eq}}$/kWh whereas using coal electricity generates 30–200 $gCO_{2\text{-eq}}$/kWh for the same process (Reich, Alsema et al. 2011).

The results for Experiment 2 show lower impacts compared with Experiment 1, mainly because this recycling process uses less energy. The results from the electricity usage demonstrate the importance of developing recycling processes that require low energy during all their steps, or, as discussed, the use of renewable sources of electricity. Future studies must focus on the optimisation of this process to reduce the toluene loss and the time needed to separate the components.

The use of HNO_3 also has small impacts in all categories analysed. The most significant result is for human health ReCiPe impacts, due to its high corrosivity, particularly to the eyes, skin, and mucous membranes in humans (for example, an oral dose of 10 mL can be lethal for humans (Information)). As well as the results for HNO_3, the use of C_7H_8 has a much lower impact when compared with the electricity effects on all ReCiPe categories. This substance can affect humans' nervous system (brain and nerves) causing headaches, dizziness, unconsciousness, memory loss, nausea, incoordination, cognitive impairment, and vision and hearing loss may become permanent with repeated exposure, as well as some other damages to the human brain. Additionally, human health effects of potential concern may include immune, kidney, liver, and reproductive effects. The effects of C_7H_8 on animals are similar to those seen in humans ((IRIS) 2005).

6. Conclusion

An LCA of two different recycling processes for Si-based solar modules was conducted, analysing endpoint ReCiPe impacts in human health, ecosystems and resources depletion. Both processes started with the manual removal of the junction box and Al frame. Experiment 1 consisted in two additional steps which are using a thermal method to delaminate the PV module, followed by a chemical process using HNO_3 to remove metals from the recovered solar cells. Experiment 2 follows the initial step of separating the junction box and Al frame with a chemical separation (C_7H_8 + heating plate) and a final manual separation of the remaining materials.

The results demonstrate that the use of electricity in both experiments is the main source of environmental impacts for all ReCiPe categories. The impacts are mainly due to the global warming potential from the fossil fuels, as well as the depletion of coal for the generation of electricity. One of the possible paths that can be taken to reduce the environmental impacts associated with energy consumption is the use of different electricity supplies, which has been demonstrated in the literature (Reich, Alsema et al. 2011).

The impacts from the chemicals used, HNO_3 and C_7H_8, are minor compared with the impacts from the electricity use, however considerable. The use of HNO_3 has small impacts in all ReCiPe categories analysed, but the most significant result is for human health, due to its high corrosivity particularly to the eyes, skin, and mucous membranes in humans. C_7H_8, in turn, can affect humans' nervous system (brain and nerves) causing, particularly, damages to the human brain. Lower impacts might be achieved by increasing the efficiency of these processes.

Comparing Experiments 1 and 2, it can be concluded that the overall impacts from Experiment 2 are approximately 50% lower than Experiment 1, mainly due to the impacts from the electricity consumption. However, the use of HNO_3 (Experiment 1) has lower impacts when compared with the use of C_7H_8 considering all three ReCiPe categories analysed. Recovering valuable metals such as Si, Ag, Al, Cu, and Ni has become essential when PV modules reach their end-of-life as they will need to be recycled in the near future to meet legislative requirements in most countries (Weckend, Wade et al. 2016). Unfortunately, the economic viability is still a challenge, as it has been shown that landfill is the cheapest option yet (Deng, Chang et al. 2019). It has been shown that the costs of some valuable raw materials are essential in the reduction of the overall production cost of Si-based PV modules, and it is expected that 70–75% of the metal value from PV wastes could be recovered with the technologies that currently exist (Yi, Kim et al. 2014), which encourages the recovery of clean and reusable materials from solar cells and modules.

In summary, the key finding of this study is that the use of electricity from non-renewable sources causes the main ecological impacts from both experiments conducted. The use of chemicals should be taken into consideration, even with smaller impacts, due to its harms to humans and the environment.

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