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Life Cycle Analysis of Silane Recycling in Amorphous Silicon-Based Solar Photovoltaic Manufacturing

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Abstract

Amorphous silicon (a-Si:H)-based solar cells have the lowest ecological impact of photovoltaic (PV) materials. In order to continue to improve the environmental performance of PV manufacturing using proposed industrial symbiosis techniques, this paper performs a life cycle analysis (LCA) on both conventional 1 GW-scaled a-Si:H-based single junction and a-Si:H/microcrystalline-Si:H tandem cell solar PV manufacturing plants and such plants coupled to silane recycling plants. Both the energy consumed and greenhouse gas emissions are tracked in the LCA, then silane gas is reused in the manufacturing process rather than standard waste combustion. Using a recycling process that results in a silane loss of only 17 percent instead of conventional processing that loses 85 percent silane, results in an energy savings of 81,700 GJ and prevents 4,400 tons of CO₂ from being released into the atmosphere per year for the single junction plant. Due to the increased use of silane for the relatively thick microcrystalline-Si:H layers in the tandem junction plants, the savings are even more substantial - 290,000 GJ of energy savings and 15.6 million kg of CO₂ eq emission reductions per year. This recycling process reduces the cost of raw silane by 68 percent, or approximately \$22.6 million per year for a 1 GW a-Si:H-based PV production facility and over \$79 million per year for tandem manufacturing. The results are discussed and conclusions are drawn about the technical feasibility and environmental benefits of silane recycling in an eco-industrial park centered around a-Si:H-based PV manufacturing plants.

Highlights:

Keywords: amorphous silicon; cryogenic separation; life cycle analysis; photovoltaic manufacturing; silane; recycling

1. Introduction

The world requires inexpensive, reliable, and sustainable energy sources. As world population and affluence increase, it is projected that between 2 to 10 times the energy currently produced will be required over the next 50 years (Lewis and Nocera, 2006; Allen and Shonnard, 2012). In order to provide future generations with sufficient energy, environmental impacts per unit of energy produced will have to be reduced significantly. Solar photovoltaic (PV) technology, which converts sunlight directly into electricity, offers a globally-scalable technically-sustainable solution to the projected enormous future energy demands (Dincer, 2000; Pearce, 2002; Fthenakis, et al., 2009). The potential for PV to be a sustainable technology has been reinforced by numerous life cycle analysis (LCA) studies considering both embodied energy and emissions (Alsema 2000; Alsema and de Wild-Scholten, 2005; Alsema and Nieuwlaar, 2000; Alsema, et al., 2006; Fthenakis and Alsema, 2006; Fthenakis and Kim, 2007; Fthenakis, et al., 2008; Mason, et al., 2006; Palz and Zibetta, 1991; Pearce and Lau, 2002). However, in order to compete economically with fossil fuels in all markets in the current subsidy landscape, further decreases in the levelized cost of electricity from solar are needed (Branker, Pathak & Pearce, 2011). If this were to occur the potential market would be substantial and begin to rival the traditional fossil-fuel-based energy market in size. For example, consider recent work by Keiser that showed that at US\$3 per watt for complete PV systems – and some commercial projects are at this level now – addressable electricity consumption rises to 440 billion kWh, equivalent to over 300GW of capacity in the U.S. alone (Keiser, 2011). To meet the cost goals for solar electricity to compete economically with fossil fuel-fired electricity, work has started exploring the use of industrial symbiosis to obtain economies of scale and increased manufacturing efficiencies for solar PV cells (Andrews and Pearce, 2011; Nosrat et al., 2009; Pearce, 2008a). Of the PV materials, hydrogenated amorphous silicon (a-Si:H)-based PV, a second generation thin film technology, offers the best ecological balance sheet (Alsema, 2000; García-Valverde, Chermi, and Urbina, 2010;

Jungbluth, 2009; Palz and Zibetta, 1991; Pearce and Lau, 2002; Wronski, et al., 2002.). This type of PV has an energy payback time around 1 year and can produce up to 31 times the amount of energy its initial consumption within a 30-year lifetime (Pearce and Lau, 2002). In addition, increased stabilized efficiencies (> 10%) have been demonstrated with dual junction a-Si:H / microcrystalline ($\mu\text{-Si:H}$) tandem solar cells, which further improve ecological performance of Si:H-based PV.

Unfortunately, there is a significant amount of waste in the Si:H material deposition process as only approximately 15 percent of the silane (SiH_4) is used (Briend, 2011). Embodied energy and greenhouse gas emissions of silane are very large, 1,146 MJ / kg silane and 61.3 kg CO_2 equivalents (eq.) / kg silane respectively (Ecoinvent, 2012), suggesting recycling as a promising approach to reduce impacts of a-Si:H manufacturing despite the small quantity of active semiconductor in the entire module. In order to continue to improve the environmental impact of PV manufacturing, this paper investigates this opportunity by performing an LCA on four scenarios: (1) conventional 1GW-scaled a-Si:H-based single junction plant, (2) a conventional 1GW a-Si:H/ $\mu\text{-Si:H}$ tandem cell solar PV manufacturing plant, (3) a 1GW-scaled a-Si:H-based single junction plant coupled to silane recycling plant, and (4) a 1GW a-Si:H/ $\mu\text{-Si:H}$ tandem cell solar PV manufacturing plant coupled to silane recycling plant. Both the energy consumed and carbon dioxide (CO_2) equivalent emissions (all important greenhouse gases included) from the processing of silane are tracked in the LCA and the silane gas is reused in the manufacturing process rather than standard combustion. The results are discussed and conclusions are drawn about the technical feasibility and environmental benefits of silane recycling in an eco-industrial park centered around an a-Si:H-based PV manufacturing plant.

2. Background

2.1 A-Si:H-based PV Manufacturing

The current manufacturing process for Si:H-based PV cells varies between different substrates, methods, and manufacturers, but the actual deposition of a-Si:H and $\mu\text{-Si:H}$ layers are formed by running the substrate through a set of chambers and exposing it to a mixture of hydrogen and silane using plasma-enhanced chemical vapor deposition (PECVD) (Street, 2000; Wronski and Carlson, 2001). The layers of a-Si:H PV are constructed in a p-i-n or n-i-p device structure. The p-layer is usually a boron-doped a-Si:H or a-SiC:H, followed by an i-layer of either undoped a-Si:H or amorphous silicon germanium a-SiGe:H and an n-layer of phosphorous doped a-Si:H (Izu and Ellison, 2003; Street, 2000; Wronski and Carlson, 2001; Wronski et al., 2002).

For the tandem cell structures, $\mu\text{-Si:H}$ has been shown to be able to act as an active layer in p-i-n solar cells (Meier, et al., 1998; Keppner, et al., 1999; Meier, et al., 2006;). Compared to the bandgap of a-Si:H (1.8eV), $\mu\text{-Si:H}$ was found to have a significantly lower energy bandgap of around 1 eV and thus the combination of both materials (two absorbers with different gap energies) leads to a tandem cell structure, referred to as the "micromorph" cell, with superior performance to both single and double junction a-Si:H-based cells because of increased use of the solar spectrum (Meier, et al., 2006).

Silane is the primary source gas for both amorphous and microcrystalline Si:H-based PV manufacturing. When silane is deposited, only approximately 15% of the gas gets used, while the remaining 85% goes unused and is treated as waste (Briend, 2011). The typical method of disposing of unused silane, which is a pyrophoric gas that undergoes spontaneous combustion in air, is to utilize combustion.

2.2 LCA of Si:H PV manufacturing

Hydrogenated amorphous silicon PV has one of the lowest total embodied energy demands for PV modules of 1200 MJ eq. per meter squared (Alsema, 2000) and energy payback times between 1-3 years (García-Valverde, Cherni, and Urbina, 2010; Pearce and Lau, 2002). This is in part due to the minimal amount of silicon needed to produce a-Si:H modules, utilizing only a fraction of what is found in conventional crystalline silicon-based cells and the ability for a-Si:H to utilize roll-to-roll production (Izu and Ellison, 2003), which reduces both production time and transportation costs with the production of these modules (Jungbluth, 2009). The performance of tandem Si:H-based cells is expected to be even better because of the increased cell performance with only an increased energy expense due to the increased thickness of 2.5 microns, of the $\mu\text{-Si:H}$ absorber layer.

Although this ecological performance is excellent particularly when compared to fossil-fuel based energy sources that it replaces, the emissions due to Si:H-based PV manufacturing can cause a carbon emissions / energy cannibalization effect

that limits the carbon neutral growth rate of the technology (Kenny, et al., 2010). Energy cannibalism refers to an effect where rapid growth of an entire energy producing industry creates a need for energy that uses (or cannibalizes) the energy of existing power plants (Pearce, 2008b). Thus, as PV is experiencing rapid growth the industry as a whole could produce no net energy because new energy is used to fuel the embodied energy of future power plants. A similar analysis is made for greenhouse gas (GHG) emissions and thus it is important to reduce the embodied energy and emissions of PV as much as possible to allow for accelerated growth rates of the industry (Kenny, et al., 2010). With global PV production having proved it is capable of >100% growth per annum, reducing the energy payback time is necessary to stay ahead of the carbon neutral growth rate.

3. LCA Methods

3.1 Goal, Scope and Functional Unit

The goal of this study is to determine if in-situ silane recycling is environmentally feasible during PECVD manufacturing of Si:H-based PV (both single junction a-Si:H and tandem a-Si:H/ μ c-Si:H). The scope of this life cycle analysis will be limited to differences in inputs for processing ("cut-off method") of silane for recycling versus not-recycling. The functional unit is 1 kg of silane used in PV production. However, the inventory data associated with these inputs will embody a "cradle-to-gate" system boundary. These results will be compared with previous LCA results from the literature to quantify the variance in embodied energy and greenhouse gas emissions.

3.2 LCA Software, Inventory, and Impact Assessment Methods

SimaPro v. 7.3 (Pré, 2012) was used to complete an energy and emissions LCA of a Si:H based PV manufacturing and silane recycling plants. SimaPro is a life cycle analysis tool that gives a quantitative measure of the impact of a product or service. In this analysis, the Ecoinvent v. 2.2 database of materials was used, which is based directly off of data from industry, evaluated by professionals, and is well known as a reliable database (Jungbluth, Tuchschnid, Wild – Scholten, 2008). Cumulative energy demand was used to analyze the overall energy costs and the model developed by Intergovernmental Panel on Climate Change in 2007 for the global warming potential over a 100 year time period, IPCC 2007 GWP 100a (Pachauri, 2007; Solomon 2007), was used to calculate the CO₂ equivalent emissions for the recycling comparison.

3.3 Silane Recycling

The silane recycling protocol studied here was proposed by Briend (2011) and is outlined in Figure 1. Figure 1 shows the relationship between the 1 GW Si:H-based PV manufacturing plant (4) and the semiconductor recycling plant (5) in an eco-industrial park proposed by Pearce (2008a). This production plant size is evaluated because of the previously discussed benefit of scale (Pearce, 2008a). It should be noted that the use of silane recycling is considered an in-process form of recycling, although the context of the study is in an 8 plant eco-industrial park. The waste silane mixture from the reaction chamber in the PV plant can be recycled by first pumping the waste silane from the reaction chamber at near atmospheric pressure and mixing it with nitrogen to avoid ignition. The diluted silane/nitrogen mixture can be processed using a filter (centrifugal separator, scrubber, etc.). If the mixture is free of corrosive compounds, a compressor can then be used to bring the pressure to 2-35 bars (Briend, 2011). Otherwise, additional filters must be placed before the compressor to reduce corrosion. After obtaining the proper pressure, a cryogenic separator is used to condense/solidify the silane and remove the hydrogen/nitrogen mixture. This point allows another potential recycling opportunity, the nitrogen and hydrogen can also be purified and reused, but will be left for future work. From this point, the silane needs to be distilled to achieve the proper purity level for the reaction chamber and the recycled silane can then be put into storage bottles/tanks or pumped through a system of parallel buffers and run through the deposition process directly again.

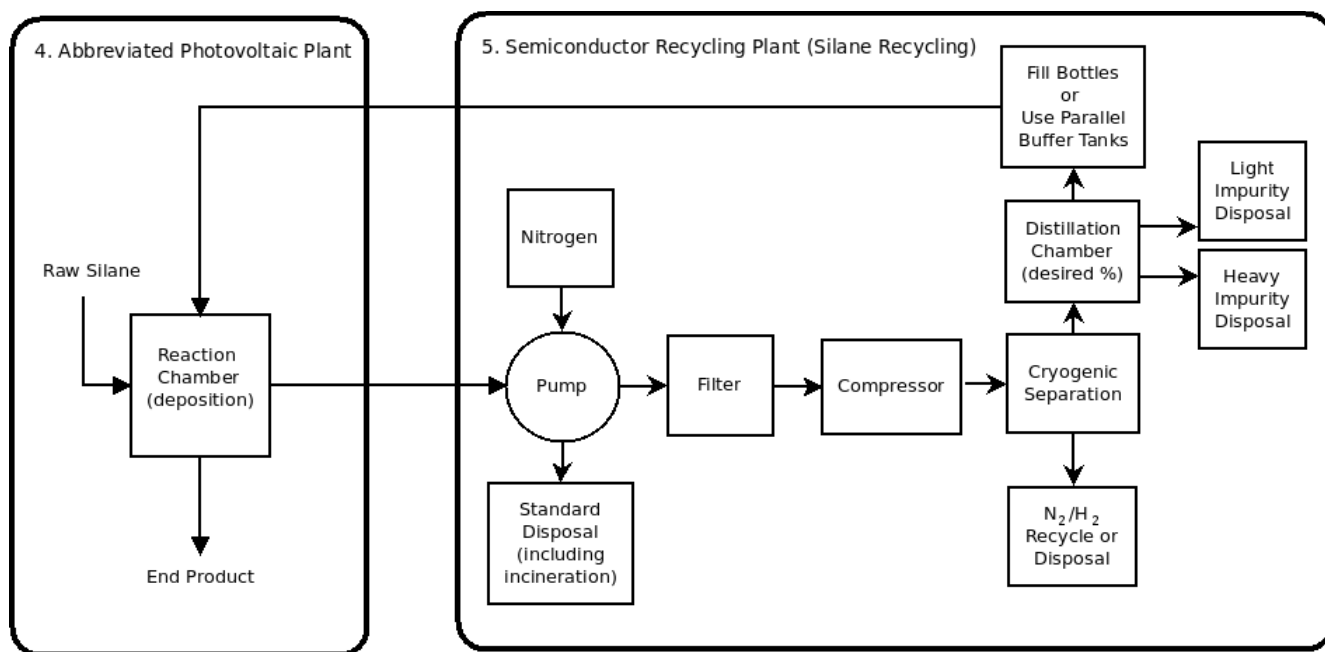


Figure 1. Schematic of LCA of silane recycling in an industrial symbiotic system manufacturing Si:H-based PV.

Currently, only 15% of the silane is used in the standard deposition process (Briend, 2011) and the remaining 85% is wasted. The percentages of potential silane wasted using the process outlined by Briend were calculated using :

$$ar(\%) = 1 - ad(\%) - a_w(\%) \times a_r \quad (1)$$

where a_r is the total percent of waste that can be recycled, a_d is the total percent of silane deposited, a_w is the percent of silane originally wasted, a_r is the fraction due to the efficiency of recoverable silane by use of recycling. From this 85 percent waste, the recycling process outlined by Briend (2011) is able to save up to 80 percent, thus saving up to 68 percent of the original amount injected into the PEVCD. This leaves a waste of 17 percent using silane recycling versus the original 85 percent waste.

The inputs for the energy and nitrogen required to create 1 kg of raw silane and 1 kg recycled silane mixture for both single junction a-Si:H PV and tandem a-Si:H/ μ c-Si:H PV production are shown in Table 1. Inputs for components outlined by Briend (2011) were estimated from manufacturer specifications and calculations for the application of recycling silane (Edwards Limited, 2012; Hijet Engineering Ltd, 2012; Kaeser Compressors Inc, 2012; Praxair Inc, 2003). Electrical use was calculated using the power rating for one hour, divided by the amount of silane recycled for one hour of continuous production from a 1 GW-scaled manufacturer. This results in an overestimate of the amount of electricity used to produce 1 kg of recycled silane. A facility at this scale would present a need for 8.61 kg of usable recycled silane to be produced every hour from an a-Si:H facility and 30.14 kg of usable recycled silane every hour from a tandem a-Si:H/ μ c-Si:H. The inputs differ for a-Si:H and tandem a-Si:H/ μ c-Si:H PV due to a difference in the volume of silane moving through the recycling system, when there is more volume at the same theoretical energy usage, the energy per unit volume is decreased. Inputs for the United States (US) were used when possible, otherwise European (RER) inputs were used. The SimaPro LCA input for 1 part was 1 kg of raw silane (silicon tetrahydride, at plant/RER U) used 100% raw materials. The terms S and U behind their country designation, refers to "system process" and "unit process". The system process includes all processes of the system as a whole, whereas the unit process shows references to previous processes.

After designing an input for the recycled silane component, the LCA can be conducted for the actual deposition using recycled silane by considering Equation 1, which will be referred to as the recycled silane mixture. Using equation 1, it is found that 32% raw silane will be needed for the deposition process in conjunction with 68% recycled silane, due to the

32% loss during the initial deposition process and the recycling process.

Table 1. LCA inputs for 1 kg of Recycled Silane Mix for both a-Si and tandem PV production using 32% of 1 kg input of Raw Silane and 68% of the Recycle Process Silane Inputs for 1 kg output using the process as outlined by Briend (2011) (Figure 1). Ecoinvent ecoprofiles are listed for each input. Acronyms US/US refer to United States (US) made inputs used within the US, and RER refers to European (RER) inputs. The S at the end of the input name means that all processes to create the input are considered as a whole.

Recycle Process Silane Inputs	a-Si:H	Tandem	Unit	Details
Nitrogen, liquid, at plant/RER S	1.875	1.875	kg	Pump: Edwards – iH80 Dry Pump 460V
Electricity, production mix US/US S	0.313	0.090	kWh	
Electricity, production mix US/US S	0.910	0.910	kWh	Filter/Compressor: Centrifugal Separator w/ Kaeser compressor
Electricity, production mix US/US S	0.605	0.605	kWh	Basic Cryogenic Air Separator
Electricity, production mix US/US S	1.742	0.498	kWh	Distillation Column: Vacuum distillation unit

These inputs were determined using a 1GW-scaled PV manufacturing plant, running continuously, with 70 W/m² for the single junction a-Si:H PV and 100W/m² for the tandem a-Si:H/ μ c-Si:H PV produced. It should be pointed out here that the fundamental differences in the inputs between a-Si:H/ μ c-Si:H and single-junction a-Si:H PV is the thickness of the absorber layers. The absorption coefficient of the i-layer of the μ c-Si:H cells are relatively low and necessitate a factor of five increase in thickness. For this analysis, the gas plant was assumed to be located next to the manufacturing plant due to the scale of the PV plant, so transportation of silane and nitrogen to the PV plant was assumed to be negligible to minimize cost and downtime due to transportation time. A sensitivity analysis was done and showed that a transportation distance of 1000 miles had less than 0.1% effect on the results. No other inputs besides those necessary for recycling were included for comparison between the life cycles of raw and recycled silane.

4. Results

The analysis of the cumulative energy demand results in the recycled silane mixture for an a-Si:H PV facility and a tandem a-Si:H/ μ c-Si:H PV facility having an impact of 409 and 397 MJ per kg of usable silane, respectively. For entire production process of raw silane, 1,146 MJ are used to create one kilogram. This means that the amount of energy to recycle silane in an a-Si:H PV plant is only 35.7% and in a tandem a-Si:H/ μ c-Si:H PV plant is even less at 34.6% of what it takes to create raw silane, as shown in Figure 2.

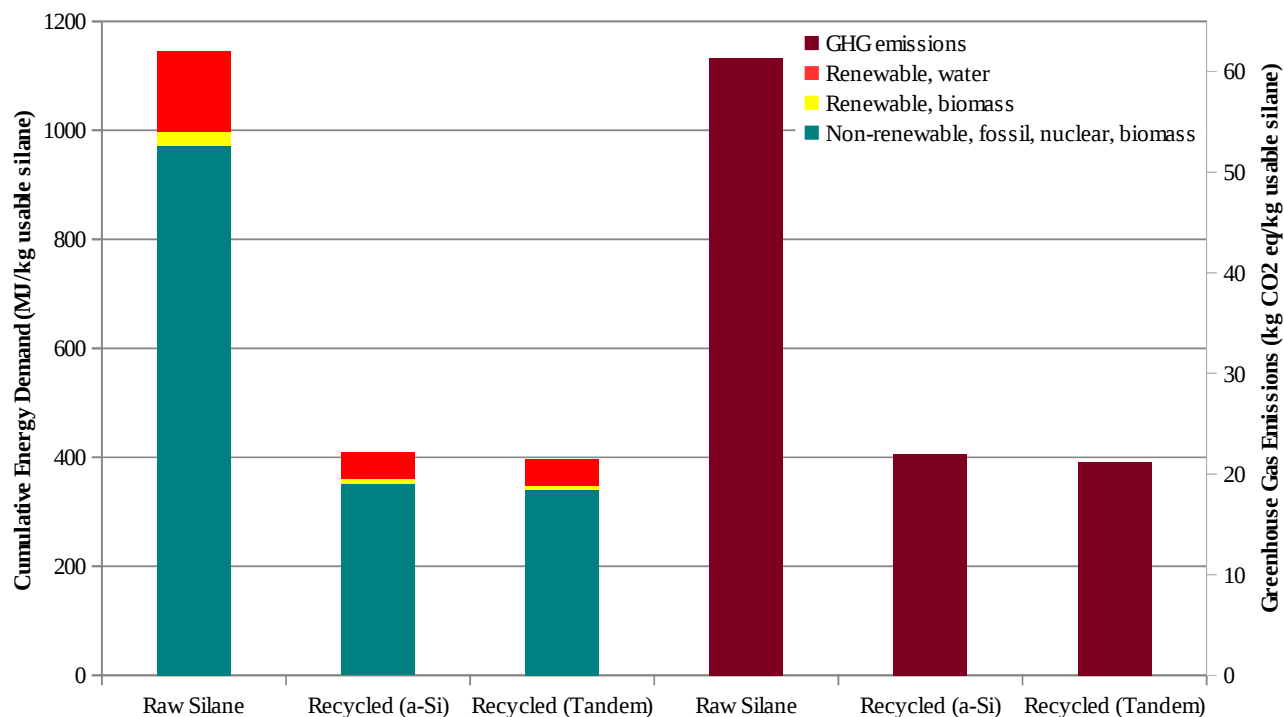


Figure 2. Cumulative energy demand and CO₂ emissions for raw and recycled silane for 1kg of silane, scaled for a 1 GW a-Si:H and tandem a-Si:H/ μ c-Si:H factory.

Using the IPCC 2007 GWP 100a v 1.02 method to determine greenhouse gas emissions in carbon dioxide equivalents for raw and recycled silane, the total emissions are 22 kg CO₂ per kg recycled silane mixture at an a-Si:H plant, 21.2 kg CO₂ per kg recycled silane mixture at a tandem a-Si:H/ μ c-Si:H plant, and 61.3 kg CO₂ per kg raw silane. The amount of greenhouse gas emissions related with the recycled silane mixture is 35.8% for a-Si:H and 34.8% for tandem a-Si:H/ μ c-Si:H PV of that of raw silane as seen in Figure 2.

The amount of CO₂ emitted differs from the amount of energy use by a small amount due to the various inputs required for silane to be produced. Various steps in the production of silane use different forms of energy, and some processes produce a greater amount of emissions for the same amount of energy used.

5. Discussion

As the results in Figure 2 show, recycling silane in Si:H-based PV manufacturing consumes less energy and emits less greenhouse gas emissions compared to silane produced from new resources. This is due to the high level of processing and transportation required in making PV-grade silane from new resources. The difference in total energy demand is 736 MJ per kg between the recycled silane mixture and the raw silane for a-Si:H single junction PV manufacturing. For comparison, 1 kg of silane can produce approximately 129 m² of a-Si:H PV material with 15 percent deposition efficiency. This means that for every meter squared produced, a total savings of 5.7 MJ can be realized. To put this figure into context, for a 1 MW scaled factory, 1 m² of frameless PV material, consumes approximately 1,200 MJ of energy (Alsema, 2000). If it is assumed that the difference in recycling for such a smaller plant will be approximately the same due to the store of the waste gas, reducing the size of the recycling process, and/or limiting the running time for the process, then recycling has the potential to save up to 5.7 MJ of the total manufacturing energy demand for each meter squared created at the MW scale. It should be pointed here that the embodied energy due to the buildings and equipment were not included in the study as it was found to be negligible (<2% of total energy demand in photovoltaic systems –ecoinvent database (SCLCI, 2012)).

If a tandem module of a-Si:H/ μ c-Si:H is compared to a single-junction a-Si:H module, the effect of recycling is increased, as shown in Figure 3. Assuming all inputs and methods are approximately the same except for a total i-layer absorber silicon thickness of 2.5 microns, 1 m² of tandem PV material would have a cumulative energy demand of 1235.6 MJ using raw silane. By recycling silane, this type of PV material has the potential to save 29.1 MJ for every meter squared produced, resulting in a cumulative energy savings of 2.4%.

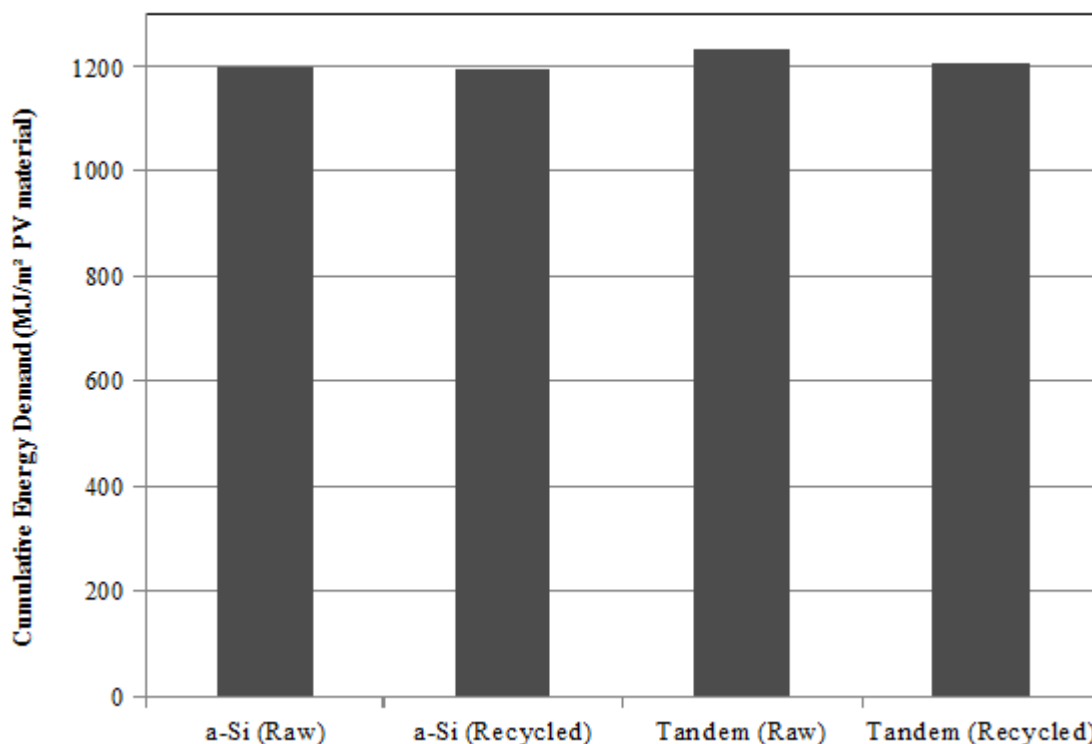


Figure 3. Cumulative energy demand comparison between raw and recycled silane for 1 meter squared of a-Si:H and tandem a-Si:H/ μ c-Si:H photovoltaic material.

A 1 GW single-junction a-Si:H manufacturer running continuously will use 111,000 kg of silane per year of which 94,300 kg of silane will go through the deposition chamber and be wasted. By recycling, there is a potential to save 55,400 kg for reuse in the deposition chamber, with an end result of only 18,900 kg being disposed. When the difference in the amount of energy and CO₂ associated with 1 kg of silane is considered and multiplied by the amount of silane used per year, the end result of recycling silane is approximately 81,700 GJ of energy savings and 4.4 million kg of CO₂ eq per year.

These are substantial energy and emission savings. To put this into perspective, these values can be compared to other methods previously discussed in the literature in relation an industrial symbiosis centered on a 1 GW a-Si:H manufacturing plant. For, example Nosrat et al. investigated the glass used for the front and back substrate of a-Si:H at this scale (2009). The energy that could be saved by maximizing recycling of glass in a-Si:H-based PV in a similarly scaled plant is 220,000 GJ (Nosrat, et al., 2009) . Thus, the embodied energy conserved by recycling silane is about one third that of potential savings from using recycled glass in the back glass layer.

Raw silane has a cost associated with its purchase approximated by Sematech at US\$0.30/g for bulk production (Visokey, et al., 1995), although it should be noted that costs are highly variable. Thus, the process outlined here for recycling reduces this cost by 68 percent, or approximately \$22.6 million/year for a 1 GW a-Si:H-based PV production facility. These cost savings thus help provide a cushion for thin film PV manufacturers from volatile silane cost fluctuations. However, these economic savings also may provide a direct competitive advantage for a-Si:H fabs that recycle silane by potentially reducing overall production costs and presumably market prices. However, it should be noted that the capital and operating costs need to be determined for the recycling process in future work. As previously noted, reduction in the installed cost of PV can have dramatic effects on consumer acceptance and market size (Keiser, 2011).

The impacts of recycling silane in a 1 GW tandem a-Si:H/ μ c-Si:H manufacturing plant under the same recycling assumptions is even more substantial. The tandem fab will use 388,000 kg of silane per year and could save 264,000 kg of silane by recycling. This is equivalent to 290,000 GJ of energy savings and 15.6 million kg of CO₂ eq per year. This represents a larger potential embodied energy savings than integrating recycled glass in the back glass encapsulation layer (Nosrat, et al., 2009). Most strikingly, silane recycling results in a reduction of raw silane purchase costs of approximately \$79.2 million/year for a 1GW-scaled tandem a-Si:H/ μ c-Si:H manufacturing plant. At a thin film PV module cost of \$0.70/Wp a 1GW-scaled plant generates \$700 million in revenue/year, so the potential cost savings from silane recycling are approximately 11% of revenue. The percentage of potential savings from recycling would increase, as cost declines are made possible by improved efficiency and increased market competition reduces margins.

Future work is needed to do a full life-cycle economic analysis of this process that would compare the economic benefit from recycling silane with the increased costs of capital equipment and operating energy costs needed to recycle the silane outlined in Table 1. In addition, Briend et al. (2011) outlined methods to recover both nitrogen and hydrogen, which could also be considered for future work to continue to depress the already small embodied energy, energy payback time, and ecological footprint while increasing the already substantial energy returned on energy invested (EROI) of Si:H-based PV manufacturing. Finally, a full LCA including all impact categories of the environmental analysis should be investigated as energy and silane are not the only contributors in the process to environmental impact.

6. Conclusions

This is the first LCA study of silane recycling in GW-scaled Si:H-based PV manufacturing plants. From the results found in this study, it is clear that not only would silane recycling allow Si:H-based PV manufacturers to become less dependent on silane cost volatility, but that silane recycling reduces energy consumption and emissions in Si:H-based PV production. In the case of a 1 GW a-Si:H plant this amounts to savings of approximately 81,700 GJ and over 4,000 metric tons of CO_{2e} per year. For the more material intensive 1 GW-scaled tandem a-Si:H/ μ c-Si:H PV manufacturing plant the savings are even more substantial: 290,000 GJ of energy savings and 15.6 million kg of CO₂ eq per year. It can be concluded that instead of buying all new silane only to discard 85%, it is more environmentally-responsible for thin film Si:H-based PV manufacturers to begin recycling silane and only supplementing the recycled supply with raw silane. The recycling process outlined here reduces the cost of raw silane by 68 percent, or over \$22 million/year for a 1 GW single-junction a-Si:H-based PV production facility and over \$79 million/year for a tandem a-Si:H/ μ c-Si:H PV manufacturing plant. These savings represent a significant fraction of revenue and thus silane recycling offers the potential to reduce the LCOE of Si:H-based PV, which could in turn make it more competitive in the PV market and increase PV penetration in the overall grid.

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