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RECYCLABILITY CHALLENGES IN “ABUNDANT” MATERIAL-BASED TECHNOLOGIES

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ABSTRACT: Much current research in photovoltaic technology is directed towards using “abundant” base metals like copper and zinc (e.g., CZTS or more recently CZTSSe) to overcome the challenges of material scarcity posed by the use of tellurium, indium, germanium, and gallium in current generation products (e.g., CdTe, CIGS, a-Si/thin-film Si). The supply of these materials is limited because they are minor byproducts of the production of copper, zinc, lead, and aluminum, so that their economic production inherently is linked to that of the base metals. But, although the base metals currently are abundant, their reserves are not inexhaustible. In addition to concerns on resource availability, the main sustainability metrics for large-scale PV growth are low cost and minimum environmental impact. As the numbers of photovoltaic installations grow, greatly displacing traditional power-generation infrastructures, recycling will play an increasingly important role in managing their end-of-life fate, so relieving pressure on the prices of critical materials. Identifying the potential issues in current technologies can help implement a take-back- or recycling-program ahead of time. Our work explores the potential for material recycling of various established commercial photovoltaic technologies, along with those under development. It sheds light on a dimension of sustainability that has not been investigated before. Based on entropy analyses, documented by the experience of recycling electronic products, we show that recycling some types of PV modules based on “abundant” materials could be burdened by complexity and lack of value, thereby creating concerns about end-of-life environmental impacts, and resource availability.

Keywords: photovoltaics, recycling, sustainability, cost reduction

1 INTRODUCTION

Assessing the sustainability of a continued rapid growth of photovoltaic markets requires investigating three measurable aspects: Cost, resource availability, and environmental impact [1]. A few times in the past, the availability of resources also have been impacted by geopolitical constraints [2].

The question of cost concerns the affordability of solar electricity compared to other energy sources throughout the world. The effects of environmental impacts include local-, regional-, and global-ones, as well as land use that must be considered over a long time-horizon. Finally, the availability of material resources matters to current- and future-generations under the constraint of affordability. More concisely, photovoltaics (PV) must meet the requirement for generating abundant electricity at competitive prices, while conserving resources for future generations; they must have environmental impacts lower than those of current modes of power generation, and preferably also less than those of alternative future energy-options.

Different photovoltaic technologies pose different challenges. For example, first-generation crystalline-silicon photovoltaics rely on abundant silicon, but its costs are relatively high due to the energy-intensive process required to produce the semiconductor materials. By comparison, although second-generation technologies are cheaper, they are less efficient. Cadmium telluride thin-film modules, for example, have the lowest production costs, but there are concerns about the availability of tellurium and the toxicity of cadmium used as precursors to CdS and CdTe. Similarly, there are similar concerns about the availability of materials for copper indium gallium (di)selenide (CIGS), i.e., gallium, indium, and some high-performance silicon technologies use potent greenhouse gases (e.g., NF₃) that has a Greenhouse Gas Potential (GWP) 17,000 higher than that of CO₂ [3].

The availability of materials to support a very large growth of photovoltaics is of concern. A recent European Commission report [4] lists four elements, viz., germanium, gallium, indium, and tellurium as being critical in terms of risks to supplies and of economic importance to the European Union markets. A U.S. DOE report focusing on U.S.- and global-markets also deems the last three as critical, but omits germanium [5]. This report deems indium as having the highest short-term criticality. Most sources agree that using gallium, indium, and tellurium in photovoltaics will increase, due to the high growth rate of the PV industry. Other applications, such as integrated circuits, optoelectronics, and display technologies, require the same elements, in particular gallium and indium. The availability of these materials is limited because they are minor byproducts of the supply of aluminum, zinc, copper, and lead; accordingly, their generation inherently is linked to that of the base metals, and thus, the rate of production of the latter must be examined. The production of copper is expected to peak within 50 years, that for Zn and Pb in about 20 years, whereas the amount of Al from known reserves could keep increasing through to the end of the century [6–8]. Considering that currently installed solar panels have an expected lifetime of 30 years, given the proper technologies for recycling, they could become a significant source of materials to ensure the development of future photovoltaics. On the other hand, the low end-of-life value of photovoltaics technologies based on abundant materials could create a new problem as there will be no economic incentive for recycling. We discuss both aspects in this work.

2 THERMODYNAMIC INDICATORS OF SUSTAINABILITY

As discussed, the state of sustainability herein is defined by characterizing cost, availability, and

environmental impact. The last two indicators can be estimated using mass-, energy-, and emission-flows, while cost/economic performance can be described by the laws of thermodynamics based on analogies. For example, the generation of entropy can impart information on the reversibility of a system change. Using entropy generation as a sustainability indicator, we then can assess technologies for producing or recycling a product in light of the physical limits of the resources used for a given task, and the impacts of these limits on the cost of accomplishing it. This assessment would reflect the minimum energy interaction (available work) to accomplish the production, or recycling separations. The minimum work is the exergy expenditure for the process. We can determine this minimum exergy expenditure by implementing the Stodola theorem i.e., $W_{used} = T_0 S_{gen}$, where S_{gen} represents the rate of entropy generation associated with the process of separation. Entropy is an additive property; thus the entropy of a system consisting of two or more compounds equals their combined entropies; this holds for all combinations of states of the compounds (or subsystems) [9].

The entropy of mixing describes a fundamental thermodynamics aspect of purification. Because of the increased number of possible configurations, mixing two separated substances results in a quantified increase in entropy [10].

Earlier studies examined the recycling potential of photovoltaics using the value of scarce materials [11,12], but the complexity of the device has not been considered so far. Other investigators used the Shannon entropy index to relate the complexity of a variety of common products to their current recycling rate [13]. However this approach fails to explain empirical recycling rates in the recovery of valuable materials at low concentrations. We found that the Shannon entropy method does not apply to photovoltaics because most of the module's mass is glass. Our work presents an alternative diversity metric applicable to a wider range of products. Renyi entropy represents a more generalized form of diversity measure than does the Shannon entropy; it is sensitive to both common- and rare-elements. The value q in equation 3 below is called the "order" of the diversity, and when $q=0$, entropy generation is insensitive to species frequencies; it is known as species richness. When q is lower than unity, the entropy-based evaluation of rare materials is a more sensitive one, while values greater than unity enable the evaluation of commoner materials [14]. In this work, we propose an alternative approach using a two-step analysis to first identify the materials that will be recycled, based on both material dilution and value. We calculated a complexity index (using Shannon- or Renyi-entropy) and a product's end-of-life value, and we map the values on a Sherwood plot which correlates a material's price to its dilution and was shown to be applicable for electronic recycling; recycled elements lie above the Sherwood line [9]. This line is useful for identifying materials to be recycled in solar panels.

3 METHODOLOGY

We compared recyclability as a function of Shannon entropy (H) and Renyi (R) entropy for 39 products, including electronics products described elsewhere [31]. Previous study found that Shannon entropy was

applicable for most electronic products, solar panels are dominated by their glass content, whereas printed circuit-boards, computers, and telephones are dominated by metals that have a higher monetary value.

Table I: Product used to compare Shannon- and Renyi-entropy based on published LCI information and recycling rate [15–32]

Product	Recycling Rate (%)
Lead acid battery	98
Car	95
Car tire	95
Washing machine	90
Fridge	90
Air conditioning	90
Soup can	62
Aluminum can	58
Laptop	40
Laser printer	34
Desktop computer	34
Color laser printer	33
2L Diet Coke bottle	31
Milk 1 gallon	28
Large laser toner	25
T5 Lamp	24
PET? bottle	22
Glass container	19
Jam container	17
Non-LCD television	17
LCD 42in television	17
Juice bottle	17
Wine glass bottle	15
Ink-jet printer	15
Alkaline battery	15
Chair	15
Coffee container (plastic)	13
Tide bottle	13
Cell phone	11
Laser toner	10
Cereal box	10
Aspirin container	9
CFL	7
Tetra Pak	5
PCB1	5
PCB2	5
PCB3	5
Office chair	2
AC-DC adapter	0

The value at end-of-life is calculated using the following equation wherein m_i is the mass of material i (kg), and k_i is the value of the material i (\$ per kg). The value of material is estimated to be 20% of new material for plastic, and 60% for metals. This established ratio is based on the average price differences between new- and scrap-material [33–37]; we used it to calculate the value of recycled materials since the prices of new material are more widely available than the values of recycled material. Sensitivity analyses were performed for the ranges of parameters shown in Table II.

$$\text{Product recycled value} = \sum_{i=1}^M m_i k_i \quad (\text{equation 1})$$

Table II: Economic values of recycled materials used for our base-case scenario and for sensitivity analyses.

	Min	Base	Max
Metal (% compared to new)	50	60	70
Plastic (% compared to new)	10	20	30
Glass (\$/kg)	0.04	0.06	0.08

The Shannon (H) and Renyi (R) entropies are defined by equations 2 and 3. Both H and R are expressed in terms of ‘bits’ denoting the number of binary separation steps needed to obtain any material from the mixture. c_i corresponds to the mass concentration of each element being recycled in the product.

$$\text{Shannon Entropy (H)} = -\sum_{i=1}^S c_i \ln c_i \quad (\text{equation 2})$$

$$\text{Renyi Entropy (R)} = \frac{(-\ln \sum_{i=1}^S c_i^q)}{q-1} \quad (\text{equation 3})$$

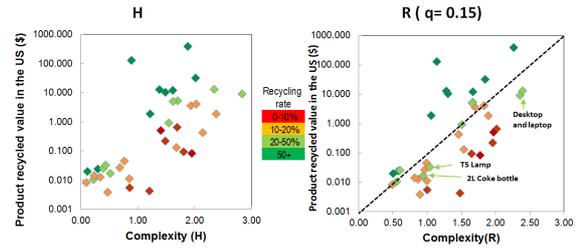
The Shannon entropy is a fundamental measure in information theory. It provides a measure of uncertainty regarding a discrete random variable; in its general form, c_i would denote the probability assigned to the value of element i to be correct, thus in our case to be recycled. The Renyi entropy is a generalized form of the Shannon entropy, which maps an entropy measure called the Renyi entropy of order q to every real number in a range from 0 to max c . We considered values of q covering the mass fractions range of 0 to 1 to find a relationship accurately predicting which products will be recycled. We established this relationship based on the data shown earlier from common products, and applied it to 10 types of photovoltaics panels representing various kinds of technologies.

4 RESULTS

4.1 Shannon versus Renyi Entropy for various products

The first step was to determine a relationship that best relates the material’s complexity to the product’s recycled value. Figure 1 illustrates the relationship between Shannon Entropy (H) and Renyi Entropy (R) when $q = 0.15$, i.e., the best value for minimizing the number of “false positives” from the apparent recycling boundary. False positives correspond to data points situated in the wrong side of the apparent recycling boundary. For example, in Figure 1, there are 4 products with a recycling rate in the 20-50% range, which are below the apparent recycling boundary. Some products are recycled even if they are below market value because of policies (such as bottle bills) and regulations (electronic bans); therefore, products such as desktop computers, coke bottles, and T5 lamps are below the apparent recycling line, their actual recycling rate actually is higher.

As Figure 1 shows, Renyi entropy generates a better estimate of the likelihood for recycling compared to Shannon entropy, in particular for products with diluted concentrations since H tends toward zero for such products.


Figure 1: Comparison between Shannon- and Renyi-entropy for the products described in Table 1.

4.2 Material inventory of photovoltaics panels

Table III shows the material inventory for photovoltaic panels based on the published structures of devices. The materials’ usages are calculated in term of elements to be recycled and are normalized for 1 m² laminate; therefore, other elements such as the frame and connectors are not included. When values for contacts are not explicitly specified in the design, we assumed a surface coverage of the contact of 5%, and thickness of 500 nm. The values of glass and ethyl vinyl acetate (EVA) were kept constant.

Photovoltaics panels contain numerous materials and only those present in sufficient concentrations to be economically extracted will be recycled. Calculating a complexity index based on all the materials given in Table 3 would not be representative of the actual elements that are likely to be recycled. Therefore, using the material inventory, the concentration and value of each material is evaluated and plotted on a previously calculated Sherwood Plot for which the recycled materials in a product were demonstrated to lie lay above the Sherwood line [50]. Figure 2 illustrates the method for the “research” CIGS panels described in Table 3. We note that glass and molybdenum were considered for recycling in this case since they are sufficiently close to the Sherwood line to warrant this.

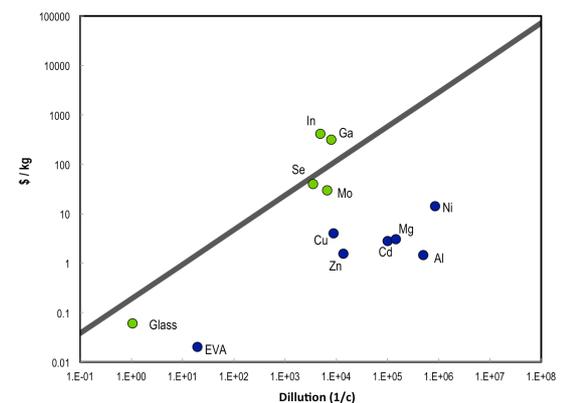

Figure 2: Concentration and value of various materials in a CIGS panel compared to Sherwood line from [50]. The green-labeled materials are expected to be recycled whereas the blue are not.

Table IV gives the inventory of recycled material that we used to compute the complexity and value of the photovoltaics panel at the end-of-life. We note that the alternative kesterite photovoltaics have a slightly lower amount of recyclable material (99.32-99.35 %) compared

to other photovoltaic panels on glass substrates (99.40 - 99.45 %).

4.3 End-of life options for photovoltaics

The recyclability of the solar panels is calculated based on the updated material inventory from Table IV; the results are shown in Figure 3 with details in Table V.

Table V: Value of photovoltaics, Shannon entropy and Renyi entropy for the base-case material value.

	kg/m ² (%)	mK (\$/m ²)	H (bits)	R (q=0.15) (bits)
CIGS – research	18.8	3.69	0.30	1.18
CIGS- commercial	18.8	2.69	0.30	1.16
CdTe	18.8	2.14	0.31	0.97
CZTSS	18.8	1.61	0.30	1.03
CZTSSe	18.8	1.45	0.30	1.02
m-Si	10.5	0.89	0.69	1.06
c-Si	10.4	0.68	0.64	0.93
HIT Si	10.54	1.05	0.72	1.08
OPV	0.40	0.44	0.58	0.87
GaAs thin film	0.41	39.95	0.32	1.31

We note from Table V that the Shannon entropy (H) values are in a narrow range (0.3 to 0.31) for all thin-film solar panels with double-glass substrates; therefore, the Shannon entropy is not a sufficiently sensitive indicator of the complexity of the PV modules.

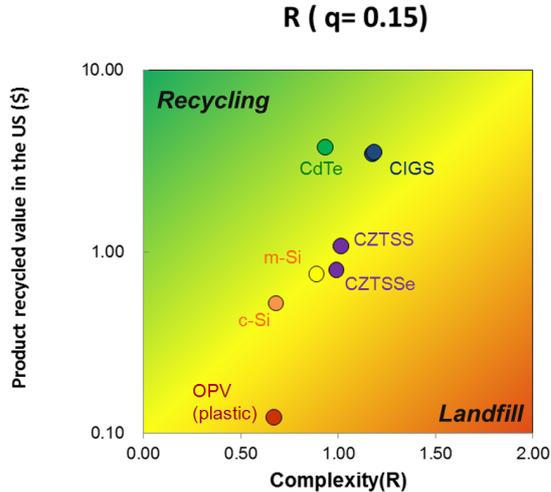


Figure 3: End-of-life options for photovoltaics panels as a function of the product’s recycled value and complexity (R) using the Renyi entropy for $q=0.15$.

At this stage, it seems that large amounts of waste may be produced at the end of life since there will be little motivation for recycling low value, highly mixed materials. This particularly is true for new type of devices, such as polymer photovoltaics, which are based on organic materials with low residual value compared to inorganic materials, so reducing the incentive to recycle them at their end of life. To be profitable, recycling organic photovoltaics should cost less than 0.44 \$/m². The value of organic photovoltaics considered in this work is likely to be higher than the actual commercial

devices because there is a strong ongoing initiative for replacing indium by other alternative transparent oxides [51].

Since the main objective of PV manufacturers is to reduce the cost of the technology, there is a decreasing concentration of valuable material, but, in many cases this is accompanied with an increase in the number of materials used in the device. In particular, while evaluating devices based on those “earth-abundant” materials proposed as substitutes for CdTe- and CIGS-technologies, we observed their higher complexity than CdTe panels and much lower end-of-life values, both of which reduce the likelihood of recycling at end-of-life. Considering an end-of-life value of 1.45 \$/m², for CZTSSe panels, the cost for recycling would need to be below 77 \$/tonne in today’s cost structures. Based on previous estimates for recycling CdTe panels where the cost was calculated to be 126 \$/tonne (not including transportation) [12], it is unlikely that panels with earth-abundant materials that are more complex would be recycled without any policy incentives, such as a mandatory manufacturer take-back.

5 DISCUSSION

This paper establishes a new metric for assessing the likelihood for landfilling or recycling of end-of-life PV modules, based on both the devices’ complexity and end-of-life value. Our investigation shows that technologies based on “abundant” and inexpensive materials could create a long-term waste problem since the manufacturer will have little incentive to take back and recycle end-of-life modules when these materials have little recovery value. Adding to this challenge is the complexity introduced by some multi-element designs currently being pursued. We determined that recycling is necessary to ensure sustainable growth of PV, regardless of the types of materials used. Previous studies of the recyclability of common products including electronics, show that policy incentives and regulation can raise the recycling rate of specific products. To ensure the long-term success of PV while evaluating alternative options based on common materials, we suggest that the cost of PV should include end-of-life management options; also policy options should be considered, such as the manufacturer’s liability for the fate of end-of-life modules.

Table III: Solar Technology considered, and the associated material inventory.

Device	mc-Si	HIT mc-Si/a-Si	c-Si	CIGS	CIGS	CdTe	CZTSS	CZTSSe	GaAs	OPV
Description	Research	Commercial	Layer transfer process	Research	Commercial	Commercial	Kesterite Cu ₂ ZnSnSe ₄ Research	Kesterite Cu ₂ ZnSn(Se,S) ₄ Research	Thin film Alta technology Research	P3HT:C ₆₀ PCBM Plastic substrate Research
Ref.	[38]	[39]	[40]	[41]	[42]	[43], [44]	[45]	[46]	[47], [48]	[49]
η (%)	25	19.0	19.1	20.3	11.15	11.2	9.7	10.7		5
Materials usage (g/m ²)	Glass (8900) EVA (960) Al (1.35) Si (183.36) Ti (0.14) Pd (0.003) Ag (0.21) Mg (0.07) PET (349) Other (103)	Glass (8900) EVA (960) Ag (0.53) Zn (0.90) a-Si (0.09) Si (228) PET (349) Other (103)	Glass (8900) EVA (960) Al (1.84) Si (90.43) PET (349) Other (103)	Glass (17800) EVA (960) Mo (2.81) Cu (2.12) In (3.84) Ga (2.34) Se (5.28) Cd (0.19) Zn (1.37) Ni (0.02) Al (0.04) Mg (0.13) Other (1.99)	Glass (17800) EVA (960) Mo (4.11) Cu (1.19) In (2.35) Ga (1.31) Se (2.96) Zn (0.45) Al (1.35) Other (0.15)	Glass (17800) EVA (960) Sn (0.75) Cd (10.54) Te (11.54) Ni (0.02) Al (0.03) Mg (0.13) Other (0.64)	Glass (17800) EVA (960) Mo (3.29) Se (8.14) Cu (2.76) Zn (1.41) Sn (2.64) Cd (0.22) In (0.30) Ni (0.05) Al (0.05) Mg (0.14) Other (2.01)	Glass (17800) EVA (960) Mo (2.50) Se (4.66) Cu (2.76) Zn (1.60) Sn (2.95) Cd (0.22) In (0.30) Ni (0.05) Al (0.05) Mg (0.14) Other (3.65)	PET (346) EVA (50) Ga (3.38) As (3.18) Al (0.32) Pt (0.43) Ti (0.18) Au (1.62) Pd (0.003) Ge (0.009)	PET (346) EVA(50) In (0.68) Sn (0.23) Al (1.15) Ti (0.05) Other (1.8)

Table IV: Recycled material inventory considered to compute the value and recycling complexity of photovoltaics at the end-of-life

Device	mc-Si	HIT	c-Si	CIGS Research	CIGS Commercial	CdTe	CZTSS	CZTSSe	GaAs	OPV
Recycled material (g/m ²)	Glass (8900) Si (183.36) Ag (0.21)	Glass (8900) Ag (0.53) Si (228)	Glass (8900) Si (90.43)	Glass (17800) Mo (2.81) In (3.843) Ga (2.34) Se (5.28)	Glass (17800) Mo (4.11) In (2.35) Ga (1.31) Se (2.96)	Glass (17800) Cd (10.54) Te (11.54)	Glass (17800) Mo (3.29) Se (8.14) In (0.30)	Glass (17800) Mo (2.50) Se (4.66) In (0.38)	PET (346) Ga (3.38) Pt (0.43) Au (1.62) Pd (0.003) Ge (0.009) Other (53.67)	PET (346) In (0.68)
Recyclable material (% mass)	86.5	86.6	86.4	94.9	94.9	94.9	94.8	94.8	86.8	86.80

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