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Enabling Circularity in the Photovoltaic Supply Chain Through Improved Upcycling and Recovery of Critical Materials from End-of-Life Modules

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ABSTRACT

The Department of Energy's Solar Energy Technology Office recently set goals to reduce the cost of recycling photovoltaic (PV) modules to rival that of landfilling by 2030. However, current recycling costs are 3-9x more expensive than landfilling costs, which results in 90% of panels being landfilled. There is significant opportunity posed by the large volumes of crystalline silicon modules set to come offline in the near future, which are estimated to contain $15B worth of recoverable materials over the next 25 years. One key opportunity is that many recyclers struggle to achieve metallurgical grade purity of critical and valuable materials such as silicon, silver, copper, and other trace metals. The full value contained in end-of-life panels is therefore not being recovered and increasing the purity of these materials offers the potential to improve recycling profitability. Here we present a combined method for the upcycling of silicon and the extraction and purification of valuable trace metals by innovative melt purification and deep eutectic separation methods that avoid the use of strong acids. The process was developed on samples received from a variety of scrap and PV-specific recycler partners in the US. Further, we identified high value, specialty material applications that can utilize upcycled silicon at higher than metallurgical grade purities but that do not require the high purities associated with the solar or semiconductor industries (e.g. > 9N). The process presented here is benchmarked against state-of-the-art techniques for silicon purification and recovery of trace metals to determine economic viability, environmental impact, and ease of implementation for recyclers. The aforementioned upcycling and extraction process will not only benefit recyclers and further a circular economy for the PV industry, but will enable onshoring of domestic supply chains for energy and semiconductor relevant critical materials.

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Introduction & Motivation

Solar panels represent an increasingly important component of our energy landscape, with current estimates at more than 5 million residential photovoltaic (PV) systems installed nationwide [1]. Domestic solar panel capacity is currently growing at 21% annually, and 9.8 million metric tons of panels are predicted to reach EoL before 2060 [2,3]. Unfortunately, 90% of these devices end up in landfills at EoL [4]. This sustainability issue can be primarily attributed to the prohibitively high costs of recycling solar panels. When compared to landfilling costs ($1-5/panel), the cost of recycling panels ($15-45/panel) is not currently economically viable for widespread adoption [5].

Despite these challenges, there are incredible opportunities to harness the aging supply of solar panels as not only a key source for the recovery of critical and valuable materials, but as a pathway to significantly decrease global carbon emissions. The value of recoverable raw materials from EoL solar panels is expected to exceed $15 billion by 2050 [6]. Recovery and recycling of these materials would simultaneously circumvent the landfilling of waste and establish more resilient domestic supply chains for these resources to meet our Nation's energy and material needs. Additionally, the manufacturing of solar panels is projected to account for up to 14% of the global carbon budget [7]. A reduction in carbon emissions from these processes would have significant implications for decreasing our carbon footprint.

With this in mind, it is increasingly important to establish a circular economy for the solar industry. We currently lack the infrastructure necessary for effective decision making on panel decommission, and end-to-end logistics for circular solar supply chains. Additionally, the development of scalable and economically viable methods for extracting and recovering critical materials from EoL panels remains one of the largest barriers to establishing sustainability within the solar industry. Closing these gaps through the development of infrastructure and technology to enable a solar circular economy is essential for achieving our nation's sustainability goals.

While materials such as glass, Al, Cu, and Ag have seen some progress towards recyclability from EoL solar panels, Si has been identified as a material for which these developments are currently lacking. Extraction and purification of Si from EoL panels remains a challenge, as the resulting materials are often not recycled at a purity required for widespread reuse. Identification of the impurities that are present in recycled Si using current solar recycling methods, understanding how these impurities impact performance in Si-based technologies, and development of novel purification methods to achieve desirable performance metrics for these technologies is of utmost importance for achieving circularity with solar materials.

Review of Related Work and Current State of the Technology Industry Uses
Scrap/e-waste Recycling of Solar Modules
Recycling of PV modules by scrap/e-waste recyclers is often conducted through mechanical methods. However, characterization of these samples indicates that they are of low purity, containing mixtures of metal as well as glass. Inductively coupled plasma mass spectrometry (ICP-MS) data ranges of Si-rich samples obtained from solar panel recyclers is shown in Table 1. While the majority of the composition of these samples was Si, there were significant concentrations of Al found in all samples. Na, Ca, and Mg were also observed in appreciable quantities, indicating high levels of glass contamination. In addition to these major contaminants, other minor ones such as Fe, Ag, Cu, and Pb were observed in all samples. These high levels of impurities, especially with regards to glass contamination, are detrimental to Si upcycling efforts. These high levels of impurities, especially with regards to glass contamination, are detrimental to Si upcycling efforts.

Table 1: ICP-MS Composition Ranges for Si Samples Obtained from Solar Panel Recyclers. Trace Elements Include the Sum of wt.% of All Other Tested Elements Not Listed (Ni, Li, B, Mn, Co, Zn, Sr, Ba, Tl)

Element	Composition range (wt.%)	Element	Composition range (wt.%)
Si	40.6-48.0	Fe	0.2-1.6
Al	2.0-2.6	Cu	0.41-0.55
Na	3.9-6.2	Pb	0.33-0.60
Mg	0.80-0.96	Ti	ND-0.23
Ca	2.5-4.5	Sn	ND-0.36
Ag	0.01-0.10	Trace elements	0.04-0.07

In response to this, various comprehensive methods that have not yet achieved commercial viability are reported for the complete recycling of Si solar panels to extract Si and other critical materials in high purity and yield. Of the existing reported methods, the European Full Recovery End of Life Photovoltaic (FRELP) process and the Arizona State University (ASU) process have received much attention and are summarized in Table 2. Recent reports indicate high costs compared to revenues for the ASU and FRELP methods [8]. For the recycling of full solar panels to be economically viable, alternative solutions must be developed that allow for the production of high-quality materials at low cost.

Table 2: Comparison of FRELP and ASU Methods for Total Solar Panel Recycling [8].

FRELP	Materials recovered	Glass, Al, Cu, Ag
	Process summary	Glass is removed from polymer and cells by knife cutting after heating in an IR belt furnace. Remaining material is incinerated, and ash is treated by sieving, acid leaching, and filtration. Leachate is electrolyzed to refine the metals.
	Advantages	Demonstrated at pilot scale (1300 modules daily)
	Disadvantages	Does not recover Pb and Sn
ASU	Materials recovered	Recovers glass Ag, Pb, Sn, Cu, Si.
	Process summary	Thermal decomposition of organic compounds, followed by leaching by nitric acid and electrowinning to recover critical materials.
	Advantages	Designed to achieve high-purity end materials
	Disadvantages	Currently a laboratory-scale separation technique, comparatively high cost

Technology Approach
SOLAR Program Overview
The Securing critical materials supply chains by enabling phOtovoltaic circuLARity (SOLAR) program, lead by Battelle and funded by the NSF Convergence Accelerator (Track I, Phases 1 & 2), seeks to solve the above issues through the development of a consortium and technology toolkits that address key challenges to enable solar industry stakeholders to close the gaps in circularity for PV. SOLAR brings together leaders in industry and academia, including the National Renewable Energy Laboratory (NREL), the Electric Power Research Institute (EPRI), Texas A&M's Energy Institute, and Equitable Solar Solutions (ESS) in order to leverage their expertise in this field. To achieve circular PV supply chains, To achieve circular PV supply chains, three main focus areas are being pursued [9]:

Focus Area 1: Sorting Toolkit (EPRI, ESS, and NREL): Development of condition assessment and sortation technologies to identify module state of health and navigate EoL decision making. These advances are expected to increase reuse and repair rates, and will involve training of technicians to support panel assessment in the field.

Focus Area 2: Upcycling Toolkit (Battelle): Development of Si upcycling and metal recovery technology and figures of merit for new application markets to support the emerging PV recycling industry. These advances will result in more economically viable processes to reduce the landfilling of solar panels.

Focus Area 3: Logistics Toolkit (NREL and TAMU): Development of integrated EoL PV module logistics models to address regulatory challenges around EoL PV transportation, storage, and recycling. These tools will account for multi-agent decision making to enable a viable path to a circular economy.

For the purposes of this article, the following sections will describe the technology development with regards to Focus Area 2 for the development of an upcycling toolkit for the PV recycling industry.

Silicon Upcycling Toolkit

These surrogates are being used to develop the following Si purification method, which is to serve as an alternative to current industry practices which typically lead to Si purities less than metallurgical grade (e.g., 99%).To accomplish this, we are engaging with solar and e-waste/scrap recyclers to develop methods by which higher purity Si can be recovered from EoL solar panels. Methods that eliminate the presence of glass powder and lower the concentration of metal impurities will purify lower-purity Si, and enable use of higher purity Si in alternative products outside the PV industry (e.g., open loop recycling). In tandem, we are utilizing surrogate samples with relative metal impurities modeled after Si samples provided by solar panel recyclers. These surrogates are being used to develop the following Si purification method, which is to serve as a replacement the ASU and FRELP methods of recycling. Here, we describe a combined method using directional solidification via modified Czochralski (Cz) growth and subsequent deep eutectic extraction to isolate Si and other critical materials (Figure 1).

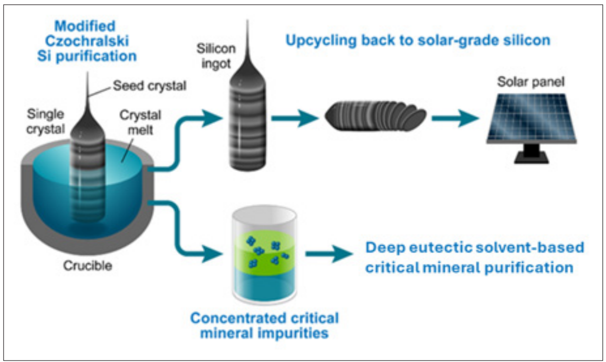

Figure 1: Process Flow Diagram for the Modified Cz Si Purification of Solar Si Followed by Deep Eutectic Solvent (DES) Extraction of Critical Materials from the Residual Material from this Process.

Directional Solidification of Silicon

Using the modified Cz growth method, Si obtained from solar panel recyclers is heated to produce a molten Si phase. A rod-mounted Si single crystal is then dripped into the molten phase and slowly removed and rotated. This method allows for the growth of a high purity single crystal rod of Si. Metal impurities have significantly higher solubility in the melt phase, and will therefore have a high degree of separation from the grown Si ingot [10-12]. We anticipate this method will yield a highly pure single crystal of Si appropriate for a number of applications, including Si-anode batteries, high-temperature thermoelectric materials, and SiC growth.

Deep Eutectic Solvent Extraction of Metals

The resulting concentrated metal impurities from this modified Cz growth will be subsequently extracted using targeted DES methods to isolate these critical materials. DES methods present significant advantages over acid purification methods that are used in processes such as the FRELP and ASU methods, in that they are safer and have a lower environmental impact [13]. Additionally, these compounds can be specially made to target the solvation of specific metals, allowing for these materials to be easily isolated [13,14]. These methods may allow for the recovery of metals such as Al, Fe, Cu, Ag, Pb, Zn, and Ni that are present in solar cells. These isolated materials may then be sold, offsetting the recycling cost to make this an economically viable method for solar material recovery.

To test this theory, preliminary experiments have been conducted using two different DESs to dissolve a Si rich component of a solar panel recycler's processing streams. The solar fraction was highly heterogeneous, containing impurity metals such as Ag, Ni, Al, Ca, Mg, Na, Cu, Fe, and Mg, and had no prior chemical treatments. The DESs were synthesized in-house and two deep eutectic solvent systems were utilized for this experiment. The solar panel samples were then added to the DES and mixed for two hours while heating. The first solvent system appears to be selective for the extraction of a number of transition- and post-transition metals over other alkali and alkali earth metals. The second solvent was selective for Ag and Cu at higher than 25% but less than 75% recovery, whereas other metals were not recovered effectively (e.g., Ni, Al, Ca, Mg, Na). With additional process optimization, both through different DES systems and ratios, the DESs could present a viable path forward to tune selectivity to successfully extract specific elements from the Cz method impurities stream while decreasing economic hurdles.

Table 3: Summary from DES for the Dissolution of Impurity Metals from a Silicon-rich Component from a Solar Panel Recycler's Processing Streams. Dissolved Metals are Shown for two DES systems.

Solvent dissolution	Solvent System #1	Solvent System #2
High (75% or greater)	Ag, Ni, Al, Cu, Fe	--
Low (25% or lower)	Mg, Na	Ni, Al, Ca, Mg, Na

Silicon Upcycling to Value Added Products

To develop avenues by which economic viability of recycling of EoL solar panels may be achieved, specialty material applications that can tolerate and potentially even benefit from the impurities found in recycled Si are being investigated. Our team has identified Si-anode Li-ion batteries, thermoelectric materials, and specialty ceramics as three possible applications for providing added value to recovered Si feedstocks. For Si-anode Li-ion batteries, dopants and defects can be utilized for nanostructuring and modification of anode expansion coefficients, which dramatically improves battery cycling performance [15]. Additionally, alloying and doping have been shown to increase conversion efficiencies in thermoelectric materials at high temperatures [16,17]. The synthesis of specialty ceramics can also benefit from these impurities, as they can act as sintering aids [18,19]. Identification of the both the positive and negative effects of specific impurities on the performance of these specialty materials will enable selective purification strategies to be employed to yield targeted materials that leverage dopants and defects for enhanced performance for these specialty applications.

Discussion

The development of recycling infrastructure and processes for the recovery of critical materials from EoL panels for sale and/or upcycling are of utmost importance on SOLAR. As mentioned earlier, the ASU and FRELP methods of recycling are not currently commercially viable for large scale recycling of solar panels due to cost exceeding revenue for both of these processes. Further details regarding the cost and revenue for these processes are detailed in Figure 2 [8]. While current estimates for potential revenue for both of these processes are around \$0.4/kg, increasing the purity of Si recovered to solar grade enables a maximum theoretical revenue greater than \$1.4/kg. Even if solar grade purity Si cannot be achieved, extraction of Si feedstocks of intermediate purity (between metallurgical-grade and solar-grade) enable revenues far exceeding what is currently being achieved. Additionally, evaluation of technologies that make effective use of these products, specifically for Si, will provide market pathways by which these extracted materials can be reincorporated into the global supply chain, rather than being landfilled. The scaling of the aforementioned SOLAR recycling process, optimization of the purity of the resulting recycled silicon, and evaluation of its feasibility for upcycling and incorporation into Si-based technologies will ultimately dictate the potential for this technology to be economically viable. With this recycling technology coupled with our developing tools for assessing the state of health of commissioned solar panels and logistics for transporting, storing, and recycling EoL panels, we envision the toolkits created from SOLAR will be transformative for enabling circularity within the solar industry.

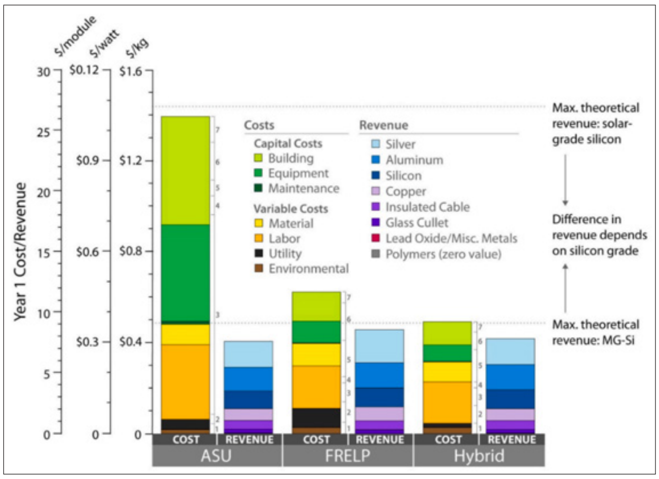

Figure 2: Technoeconomic Assessment Costs from Three PV Recycling Processes [8].

Conclusions & Recommendations

The NSF Convergence Accelerator funded project, SOLAR, is using convergent research approaches to bridge the main gaps in achieving material circularity for photovoltaics. Specifically, the Upcycling toolkit focuses on 1) purifying Si using a modified, industrially acceptable method, 2) recovering critical and valuable metals using DESs, and 3) identifying new products and markets for upcycled Si to create new, open loop supply chains for photovoltaic materials. Ideally, the technology developed here can be directly used or modified for use in existing recycling facilities or by material end users to increase the revenue associated with photovoltaic recycling processes, thereby enabling recycling to be cost competitive with landfilling.

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