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Review

Recycling paths for thin-film chalcogenide photovoltaic waste – Current feasible processes

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ABSTRACT

Thin film chalcogenide photovoltaic technologies (CIGS, CdTe) make use of critical and toxic materials. Therefore a sound recycling of production waste and of end-of-life PV modules is essential to increase the availability of critical materials and to decrease the environmental impact of the products. Several processes to recover metals and semiconductors from thin-film PV modules have been developed, with some being operated at pilot scale. This paper presents an overview of proven recycling concepts. The approaches were divided into three steps: "delamination of the modules", "decoating of the substrate" and "extraction and refining of the metals and semiconductors" to illustrate possible recycling paths. For each step feasible mechanical or chemical processes are presented and their advantages and drawbacks are described. The results show that recycling technologies for chalcogenide photovoltaic modules are sufficiently explored and commercially available. However, the responsibility to set up efficient collection and recycling systems is in the hands of the PV producers and must be supported by appropriate policies.
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1. Introduction

Photovoltaic (PV) energy production is one of the most promising and technological mature technologies for renewable energy production. At the end of 2010 the cumulative photovoltaic capacity around the world reached more than 40 GW [1]. During 2010 alone PV modules with a capacity of 27 GW were produced, of which 1.9 GW were chalcogenide solar cells [2]. Chalcogenide solar cells consist of II–VI and I–III–VI$_2$ semiconductors such as CdS/CuInSe$_2$ (CIS), Cu(In,Ga)Se$_2$ (CIGS) and CdTe. Those compounds are suitable for terrestrial photovoltaic energy generation because of their high efficiency, long-term stable performance and low cost production [3]. Their high optical absorption makes them an ideal material for the conversion of sunlight into electricity [3,4]. Consequently a thin layer ($\leq$2 μm) is sufficient to absorb most of the useful parts of the light spectrum [3,4], therefore those technologies count among thin film photovoltaics.

However, employed materials such as In, Ga and Te are regarded as critical [5–8] and hazardous materials, which, as with Cd, may harm the environment if they are not recovered or disposed of properly. Moreover, several studies show that the availability of In and Te can, under certain conditions, limit the market growth of chalcogenide PV technologies [9–15].

Therefore a proper recycling of photovoltaic waste will become increasingly more important considering the growing number of installations and expansion of production. Several recycling technologies for chalcogenide solar modules have already been developed by PV manufacturers, research institutions and small and medium enterprises – some of them are even in operation (pre-commercially). This paper will present feasible recycling paths for chalcogenide photovoltaic modules. The paths are derived from a review of proven recycling processes analysing available literature and interviewing key experts in this field.

In Section 2 the composition and design of chalcogenide PV modules are described. Possible recycling paths for the recovery of thin-film materials as well as advantages and disadvantages of the processes are presented in Section 3. Environmental impacts, costs, and research and development needs are discussed in Section 4. Concluding remarks are given in Section 5.

2. Composition of chalcogenide photovoltaic modules

A chalcogenide is a chemical compound consisting of at least one chalcogen (oxygen, sulphur, selenium, and tellurium) and at

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least one more electropositive element [16]. Chalcogenide solar cells include CdTe (II–VI semiconductor) and the chalcopyrite family (I–III–VI_2 semiconductors) $CuInSe_2$ and the alloys Cu(In,Ga) Se_2, Cu(In,Ga)S_2 and Cu(In,Ga) (Se,S)$_2$, for simplification following all called CIGS solar cells. A schematic cross-section of the superstrate and substrate layout of CdTe and CIGS solar module is shown in Fig. 1.

For both technologies low-iron float glass is used as cover glass, in some cases fitted with overprints and anti-reflex coatings [17]. The cover glass, or in the superstrate layout the back sheet, is glued by means of a plastic adhesive (ethylene/vinyl-acetate (EVA), polyvinyl butyral (PVB), and others). In most cases EVA is used as encapsulation foil, which mainly consists of a thermoplastic. After the lamination process the thermoplastic becomes an elastomer, which cannot be further melted [18]. For mechanical stability and protection, thin-film PV modules are sometimes framed. Depending on the size and design of the module the frame, often made of aluminium, accounts for 12–15% [17]. The frame, and also junction box and connecting cables, can usually be removed without difficulty. Frameless modules are also very common and are often, but not exclusively, used for structurally integrated roof and facade modules.

CIGS cells are usually manufactured in the substrate configuration, which requires an additional encapsulation layer and/or glass to protect the cell surface. Cells are typically grown on soda-lime glass, but also on metal (steel, titan) or polymer foils. The mostly used back contact is molybdenum. Because of a high absorption coefficient, a very thin CIGS absorption layer (0.1–0.3 μm) is sufficient for absorbing the incident radiation [4], but for compositional uniformity over large areas the absorption layer should be at least 1 μm thick [19]. Commonly CdS is used as buffer layer to form a hetero junction. But also “Cd-free” buffer layers such as ZnS, ZnSe, ZnInSe and In_2S_3 are explored. As front contact a transparent conductive oxide (TCO) such as Indium–Tin-Oxide (ITO) or Al doped ZnO is employed [3,4,17,20].

Flexible CIGS modules, for example produced by the US-American companies *Solo-Power* and *Ascent Solar*, are completely encapsulated by polymers and have a clearly different material composition, where polymers and steel, instead of glass, dominate the types of material. Recycling these modules will be a new challenge because of the use of fluoropolymers [17].

CdTe solar cells can be grown both in substrate and superstrate configuration. However the highest efficiencies are achieved in the superstrate configuration, in which layers of TCO, CdS, CdTe, and the metal back contact are sequentially grown on glass substrates. The glass substrate can be low cost soda-lime glass or for higher processing temperatures alkali-free glass (generally borosilicate). The most commonly used TCO is SnO_x:F, but also ITO, ZnO:Al, and $CdSnO_4$ were applied to grow high efficiency solar cells. The CdS buffer layer forms a hetero junction with the CdTe layer. The thickness of the CdTe layer depends on the deposition methods and is typically in the range of 2–6 μm, whereas just 2 μm are required to absorb most of the useful spectrum of the light. In research thickness is reduced below 1 μm [21,22]. A quasi-ohmic back contact is formed by a combination of a buffer layer and metallization, commonly Cu/Au, Cu/graphite, or graphite pastes doped with Hg and Cu [3,4,20].

Typical indicative material compositions are shown in Fig. 2. The exact composition depends on the module design and the thin-film layer configuration. Therefore, in case the material input in the recycling process in not known, the composition and material content have to be analysed beforehand.

3. Recycling paths

There are different types of photovoltaic waste arising:

- photovoltaic modules turning up as warranty returns, end-of-life-waste or as production rejects (laminates modules or submodules (coated substrates))
- production residues from the deposition processes (chamber scraping, filter load, edge grindings etc.) – normally a mix of thin-film materials at times contaminated with sandblasting abrasives and surface materials of equipment components
- not fully utilized pure raw material input (e.g. sputter targets)

The following chapter describes possible recycling processes to recover thin-film materials from thin-film chalcogenide (sub)modules and production residues. Several companies and research institutes developed recycling processes for thin-film chalcogenide photovoltaics. The US-American CdTe-PV producer *First Solar*, former *Solar Cells*, operates recycling facilities located at their production plants. The facilities mainly treat production waste (submodules, deposition overspray), but can also recycle end-of-life modules or warranty returns. Also the German CdTe-PV producer *ANTEC Solar* has a pilot-scale facility suitable for small samples [23], which is based on a recycling process patented by Campo [24,25]. The Canadian metal refinery *5N Plus* has developed processes to recycle residues, non-laminated and laminated modules of CdTe- and CIGS–PV [26–28]. *Umicore*, a Belgian refinery and precious metal recycler, has developed a hydrometallurgical process to recover Cu, In, Ga and Se from CIGS (production) scrap with a refining capacity of 50t/yr [29]. They also recycle spent indium-tin-oxide targets in their pyrometallurgical recycling line [30]. The German company *Loser Chemie* developed a universal physical-chemical process for the recovery of non-silicon thin-film materials from photovoltaic waste [31,32]. The current capacity of the pilot line is 10 tons per week. The concentrated metals can then be further processed in a metal refinery and the glass can be reused for float glass or container glass production [31–33]. The German company *saperatec* operates a pilot plant for the separation of bondings and coatings. The focus of the operations is currently on the recycling of thin film photovoltaic modules, i.e. delamination and removal of the thin-film materials from the substrate – especially from production

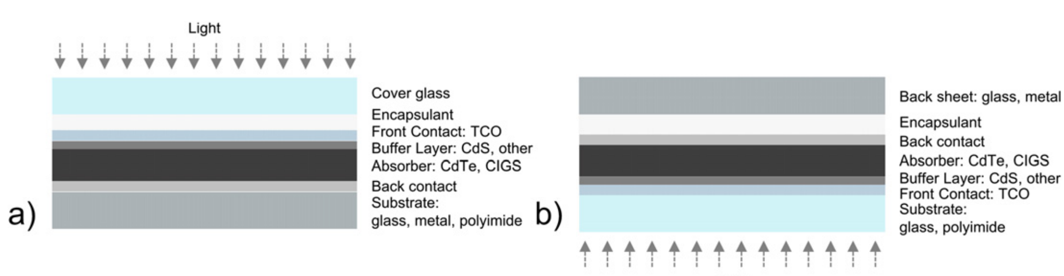

Fig. 1. Schematic cross-section of (a) substrate and (b) superstrate layout for chalcogenide solar modules.

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