



Decoupling Thin Film CdTe Growth from Packaging: Toward Record Specific Power in Low Cost Polycrystalline PV

Preprint

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*Presented at the 2017 IEEE Photovoltaic Specialists Conference (PSVC)
Washington, D.C.
June 25-30, 2017*

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Contract No. DE-AC36-08GO28308

Conference Paper
NREL/CP-5K00-68786
September 2018



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Suggested Citation

Clayton-Warwick, Deborah, Michael Kempe, Matthew Dabney, Teresa Barnes, Colin Wolden, and Matthew Reese. 2018. *Report Title: Decoupling Thin Film CdTe Growth from Packaging: Toward Record Specific Power in Low Cost Polycrystalline PV: Preprint*. Golden, CO: National Renewable Energy Laboratory. NREL/CP-5K00-68786. <https://www.nrel.gov/docs/fy18osti/68786.pdf>.

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303-275-3000 • www.nrel.gov

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This work was authored in part by the National Renewable Energy Laboratory, operated by Alliance for Sustainable Energy, LLC, for the U.S. Department of Energy (DOE) under Contract No. DE-AC36-08GO28308. Funding provided by the U.S. Department of Energy Office of Energy Efficiency and Renewable Energy Solar Energy Technologies Office. The views expressed herein do not necessarily represent the views of the DOE or the U.S. Government. The U.S. Government retains and the publisher, by accepting the article for publication, acknowledges that the U.S. Government retains a nonexclusive, paid-up, irrevocable, worldwide license to publish or reproduce the published form of this work, or allow others to do so, for U.S. Government purposes.

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Decoupling Thin Film CdTe Growth from Packaging: Toward Record Specific Power in Low Cost Polycrystalline PV

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Abstract — There are critical material and scientific barriers to producing high efficiency, flexible, lightweight solar cells that maximize specific power in a cost-effective manner. III-V solar cells have produced the highest specific power of any photovoltaic technology due to their extremely high efficiency, however, they are also among the most expensive to produce. This work explores a novel lift-off approach with thin-film solar cells, particularly CdTe, to achieve high specific power at low costs. Thin-film devices can be delaminated from their heavy, glass growth substrates post-growth by exploiting a mismatch in coefficient of thermal expansion between the two. This allows thin-film PV to be decoupled from high temperature growth requirements and repackaged using lightweight materials. In this work we evaluate different materials and approaches for achieving large area delamination of CdTe films from their glass superstrates. It is found that epoxy bonded to a rigid handle provides robust and reproducible delamination at the TCO/CdS interface upon submersion in liquid nitrogen. Following delamination, a transparent front contact is re-deposited and selectively etched to expose the back contact to produce a functioning device. Effects of delamination and contributions to device functionality from CdCl₂ and CdS layers are discussed. Reproducible delamination of high efficiency, thin-film solar cells over large areas will enable work to further improve device efficiency through passivation and reconstruction of the previously buried interface between the transparent contact and CdS.

I. INTRODUCTION

The specific power, or power-to-weight ratio, of a solar cell is determined by the efficiency of the photovoltaic (PV) device and the weight of the final package (substrate, encapsulation, etc.). Currently, GaAs solar cells produce the highest specific power of any PV technology due to their high efficiency and ability to reduce weight via substrate removal. Additionally, removing and reusing expensive single crystal substrates lowers production costs for III-V based PV. However, they are still extremely expensive compared with other PV technologies [1]. In particular, the production cost per area of thin films, such as CdTe and CuIn_xGa_{1-x}Se₂ (CIGS), are two orders of magnitude less than GaAs and other III-V's [2, 3]. At current thin-film PV production module efficiencies of 14-17% [4, 5, 6], specific powers approaching

500 W/kg hypothetically could be achieved by reducing thin-film package weight from 16.7 kg/m² to 350 g/m². Package weights as low as this have been reported for commercial III-V products [7]. The bulk of a conventional module's weight is due to the double glass module construction. Thus, post-growth delamination and repackaging using lightweight materials can theoretically make high specific powers achievable with the low production costs from thin-film PV growth. Other attempts have been made to decrease the weight of thin-film devices by growing directly on lightweight polymeric films or ultra-thin metal foils. However, because of high-temperatures (500-600°C) used in conventional processing steps, this approach can be complicated and requires extensive development to decrease growth temperatures, to manage the dimensional stability of the substrate, and limit impurity diffusion.

In this paper, we explore the thermo-mechanical delamination/lift-off of completed CdTe devices by exploiting a mismatch in coefficient of thermal expansion (CTE) between the thin film and growth substrate. This produces separation at or near the front contact/CdS interface (Figure 1a-b), which is particularly beneficial because of its proximity to the p-n heterojunction. Recombination near this junction is thought to be a major source of voltage loss in CdTe devices, which can be detrimental to device performance [8, 9]. By gaining access to this interface, post-growth reconstruction and passivation can be performed to reduce recombination and improve device efficiency. However, before this can be addressed, delamination of high-efficiency thin films must be made reproducible over large areas. This requires a careful understanding and control of adhesion at the interface at which lift-off occurs. While delamination of epitaxial materials may be well understood [10, 11], the polycrystalline nature of thin films introduces new challenges. This paper aims to initiate the study of delamination in thin films, which will enable reductions in package weight while maintaining efficiency. Additionally, by altering the newly exposed interface, we can probe the material system in new ways. This type of knowledge will lay the groundwork in improving device efficiency through interface modifications.

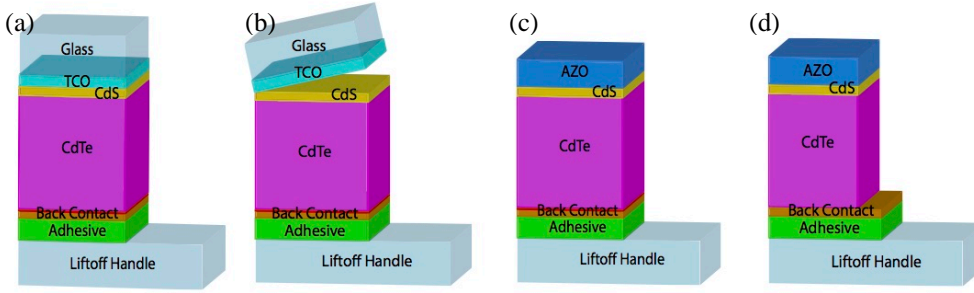


Figure 1: (a) CdTe device structure prior to lift-off; (b) Delamination occurs at the front contact (TCO)/CdS interface; (c) New front contact material (AZO) is regrown; (d) Selective etch performed to partially expose back contact.

II. MATERIALS AND METHODS

All CdTe devices were grown in the superstrate configuration using Corning® 7059 glass coated with a fluorinated tin oxide (FTO) using chemical vapor deposition. 100 nm of oxygenated cadmium sulfide (CdS:O) was deposited via rf magnetron sputtering in an oxygen/argon ambient. CdTe (~4 μm) was then deposited using close-spaced sublimation (CSS) with the substrate held at 600 °C in an oxygen/helium ambient. Vapor-phase CdCl₂ annealing was also performed in a CSS configuration with the substrate temperature fixed at 400 °C in an oxygen/helium ambient. Finally, a Cu/Au back contact was evaporated and annealed post-deposition to promote Cu diffusion [12].

Figure 1 displays the current process of delaminating and re-contacting devices. First, a glass handle was adhered to the Cu/Au back contact using several types of adhesives, including silver paint, pressure sensitive adhesives, epoxy (Hysol® 1C™), thermoplastic polyurethane (TPU), and ethylene-vinyl acetate (EVA). The structure was then submerged in liquid nitrogen to induce delamination. To first order, it is believed that the primary cause for delamination is a mismatch in CTE, which causes each layer to contract by varying amounts. This strains the interfaces and leads to fracture along either the weakest interface or the region with the highest strain.

After lift-off, the front contact was regrown (Figure 1c) using aluminum-doped zinc oxide (AZO), a transparent conducting oxide (TCO) that can be deposited at low temperatures (~60°C) via sputtering. Because the Cu/Au back contact was made inaccessible by the adhesive and handle, it must be subsequently exposed. A selective etch was performed to remove all device layers down to the back contact (Figure 1d) using Kapton® stickers (4 mm diameter) as an etch mask and ferric chloride as the etchant (Figure 2).

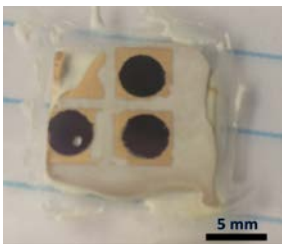


Figure 2: CdTe is in the substrate configuration after lift-off. Selective etch performed to access Cu/Au back contact (gold squares) for performance measurements.

III. RESULTS AND DISCUSSION

Key factors affecting uniformity of lift-off were determined by testing various types of adhesive. Silver paint, which has a grainy surface texture made up of silver particles after solvent evaporation, resulted in a rough, blistered surface of CdTe. This transfer of texture from the adhesive to film was also seen with various tapes (carbon, 3M™ XYZ-axis electrically conductive, Kapton®). In addition, tapes provided inconsistent and partial lift-off that was dependent on pressure and method of application.

Of the adhesives tested, epoxy provided the most consistent lift-off, with clean delamination of more than 1 cm² areas. Using epoxy, a functioning CdTe device has been delaminated and re-contacted to make a working device. Before lift-off, this device had an efficiency of 12.3% and its J-V curve (black) can be seen in Figure 3. After delamination and regrowth of the front contact, the efficiency was ~7.5% (red curve in Figure 3). Solar cell efficiency is defined as $\eta = V_{oc}J_{sc}FF/P_{in}$ where V_{oc} is the open-circuit voltage (x-intercept in Figure 3), J_{sc} is the short-circuit current density (y-intercept in Figure 3), FF is the fill factor, and P_{in} is the

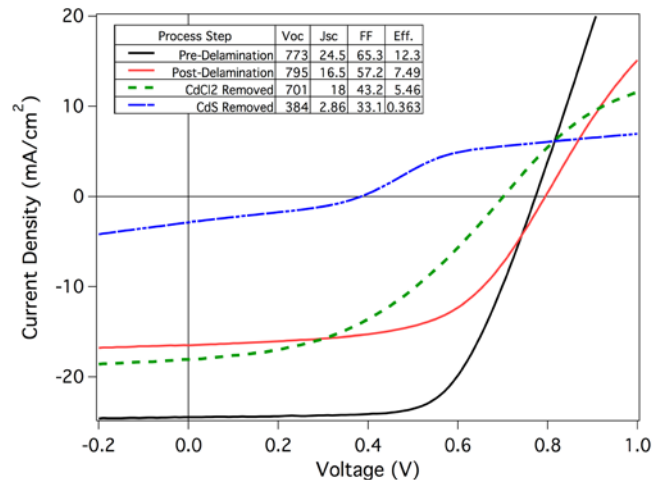


Figure 3: J-V curves for a CdTe device before lift-off (black curve), after lift-off (red), after rinsing with DI water to remove CdCl₂ (dashed green), and after rinsing with HCl acid to remove CdS (dashed blue).

incident power. By comparing the black and red J-V curves, it can be seen that the decrease in efficiency following delamination is largely due to a considerable decrease in J_{SC} as well as an increase in series resistance. The increased series resistance is largely due to a poorly optimized front contact ($\sim 80 \Omega/\text{sq}$). The decrease in current density is likely caused by large cracks that formed during AZO deposition (Figure 4). Because epoxy has a much higher CTE than CdTe, it expands to a greater degree when heated during sputtering. This, in turn, applies enough tensile stress on the CdTe film to cause cracking. A minor problem that results is the creation of shunt pathways through AZO deposition within the cracks. As seen in Figure 3, this results in a slight decrease in shunt resistance.

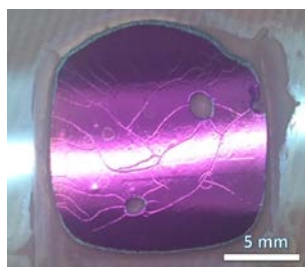


Figure 4: Large cracks seen in films after AZO deposition and before selective etching.

More importantly, these cracks can drastically reduce the contacted cell's effective area, making the area used for J_{SC} calculations (that of the 4 mm Kapton® sticker) too large. The efficiency measurements reflect these limitations and current work is focused on addressing the source of such cracking. The measurement of interest then, is V_{OC} because it does not depend on area. Of particular interest is the increase in V_{OC} after lift-off. Immediately after the growth of the superstrate device, higher efficiency/voltage was seen. Over the course of months it dropped from ~ 850 mV to the pre-delamination voltage of 773 mV. This is likely due to Cu migration, which can be a source of metastability that can be reflected in a voltage loss [13]. It is commonly observed that this effect can be modestly reversed with elevated temperatures, such as that seen during regrowth of the front contact ($\sim 60^\circ\text{C}$).

To roughly determine the importance of the layers near the TCO/CdS interface, some further processing was done. Because CdCl_2 is a water-soluble salt, this layer was removed by rinsing with DI water after delamination and before AZO deposition (the accumulation of a monolayer of CdCl_2 at the CdS/TCO interface is supported in detail by a comprehensive photoemission study reported in a separate paper [14]). The removal of this layer resulted in an apparent decrease in V_{OC} of roughly 100 mV (dashed green curve in Figure 3). This seems to suggest that CdCl_2 plays an important role in maintaining voltage in CdTe devices. On a separate sample, the CdS layer was removed by swabbing the newly exposed interface with 10% HCl acid. By removing this layer, the heterocouple between CdTe (p-type) and CdS (n-type) breaks down and the V_{OC} decreases by about a factor of two (dashed blue curve in Figure 3).

V_{OC} measurements provide an assessment of the quality of the heterojunction, and it is clear that altering the interface also alters the performance of the solar cell. Thus, by improving the previously buried TCO/CdS interface, such as through surface reconstruction and passivation, it is possible to improve the efficiency of CdTe devices using this lift-off technique. Additionally, it is possible that parameters of a chosen adhesive may be tuned to access other buried interfaces, allowing for a much more robust understanding of CdTe devices.

While epoxy may serve as a good adhesive to demonstrate the reliability of thermo-mechanical lift-off, it is not practical for long-term studies. This is due not only to the aforementioned cracking, but also its rigidity. To move toward flexible and lightweight devices, a different adhesive must be used. TPU has been shown to result in clean delamination (Figure 5a), but this adhesive has its own complications. Namely, its adhesion to glass is weak such that

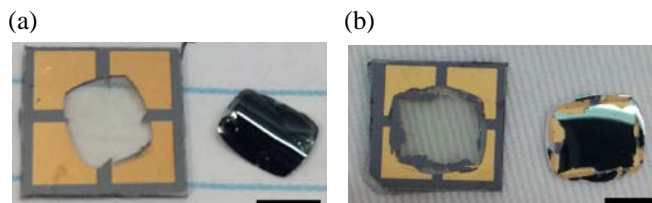


Figure 5: Delaminated CdTe film using (a) TPU (which has detached from its glass handle and curled up) and (b) EVA with a small plastic handle (5 mm scale bars).

when the CdTe/TPU/glass handle structure is submerged in liquid nitrogen, the TPU detaches from the glass handle while simultaneously lifting off the CdTe. Because the CTE of TPU is much higher than CdTe, the isolated CdTe/TPU multilayer curls up, leaving the CdTe film susceptible to cracking and tearing. Possible solutions to this issue are being explored in current research. Finally, EVA has shown some promising results (Figure 5b), yet delamination is inconsistent and not reproducible. Future work will focus on understanding why this is the case and how to correct it.

IV. SUMMARY AND CONCLUSIONS

In this paper, we examined the effects of various types of adhesives (silver paint, pressure sensitive adhesives, epoxy, TPU, EVA) on delamination of CdTe thin-film solar cells. Qualitative results are summarized in Table 1. We found that epoxy results in the most consistently clean lift-off over large areas (larger than 1 cm^2). Using epoxy as the adhesive, we delaminated CdTe films, regrew the front contact (by sputtering AZO), and selectively etched to make complete devices. Because of cracks in the film, efficiency was reduced relative to the starting value. The partial recovery of V_{OC} showed us that delamination might be a tool to modify the interface and junction in delaminated cells. A thin layer of CdCl_2 at the interface appears to help reduce voltage loss.

TABLE I
SUMMARY OF ADHESIVES USED FOR DELAMINATION

Adhesive	Level of Delamination	Appearance
Silver paint	Complete	Blistered surface
Pressure sensitive adhesives	Partial	Textured surface
Epoxy	Complete	Rigid, cracks after AZO deposition
Thermoplastic polyurethane (TPU)	Varied	Curled, easily torn/cracked
Ethylene-vinyl acetate (EVA)	Varied	Smooth, some partial lift-off

While epoxy provides consistent and clean lift-off that has been useful in initial studies, its rigidity precludes it from use in flexible delaminated devices. Therefore, future research will be done to identify, understand, and optimize other promising adhesives and handles. Once reliable delamination is achieved, improvements to the interface can be investigated. Examples include regrowth of a CdS layer after its removal, passivation of the exposed interface prior to depositing the front contact, and methods to eliminate cracking in delaminated films.

ACKNOWLEDGEMENTS

This work was supported by the U.S. Department of Energy under Contract No. DE-AC36-08GO28308 with Alliance for Sustainable Energy, LLC, the Manager and Operator of the National Renewable Energy Laboratory. Funding provided by Office of Naval Research. The U.S. Government retains and the publisher, by accepting the article for publication, acknowledges that the U.S. Government retains a nonexclusive, paid-up, irrevocable, worldwide license to publish or reproduce the published form of this work, or allow others to do so, for U.S. Government purposes.

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