ABSTRACT

The authors present a manufacturing cost analysis for producing thin-film indium phosphide modules by combining a novel thin-film vapor–liquid–solid (TF-VLS) growth process with a standard monolithic module platform. The example cell structure is ITO/n-TiO2/p-InP/Mo. For a benchmark scenario of 12% efficient modules, the module cost is estimated to be $0.66/W(DC) and the module cost is calculated to be around $0.36/W(DC) at a long-term potential efficiency of 24%. The manufacturing cost for the TF-VLS growth portion is estimated to be ~$23/m2, a significant reduction compared with traditional metalorganic chemical vapor deposition. The analysis here suggests the TF-VLS growth mode could enable lower-cost, high-efficiency III-V photovoltaics compared with manufacturing methods used today and open up possibilities for other optoelectronic applications as well. Copyright © 2016 John Wiley & Sons, Ltd.

KEYWORDS
cost; III-Vs; thin-films; InP; modules; manufacturing

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1. INTRODUCTION

Historically, III-V material systems grown by the vapor–solid method, such as metalorganic chemical vapor deposition (MOCVD), have been used in the highest performance solar cells [2], especially in demanding extraterrestrial scenarios [3]. However, the cost of these cells in modules has been significantly higher than conventional electricity generators [1]. These costs are driven by three main factors: epitaxial single crystal growth substrates, inefficient use of organometallic precursors, and capital depreciation due to low throughput [1,4]. There is significant opportunity to reduce costs and increase performance if new process technologies can be developed to address these factors. Recently, we have created a new process called thin-film vapor–liquid–solid (TF-VLS) growth, which drastically mitigates the cost components mentioned previously. The technique has been shown to yield high optoelectronic quality III-V thin films on non-epitaxial substrates, thus providing an attractive route for producing large scale III-V solar panels at high efficiency and low cost [5]. However, no detailed cost analysis has yet been performed for this approach. Here, we present one such analysis using TF-VLS-grown indium phosphide (InP) configured in a standard thin-film module platform. The results show the potential of this new growth technique for lowering the cost of III-V photovoltaics.

We want to point out that the process flow and the associated cost structures described in this paper are applicable to other III-V materials with only minor adaptations. Additionally, although the focus in this paper is on photovoltaics, other applications that use III-Vs, such as power devices or solid-state lighting, may also benefit from this low-cost growth process.
1.1. Thin-film vapor–liquid–solid (TF-VLS) growth of indium phosphide and resulting optoelectronic quality

Thin-film vapor–liquid–solid (TF-VLS) growth has so far been demonstrated with InP as a model material system [5]. InP has a direct band gap of 1.34 eV, with a Shockley–Queisser efficiency limit of ~33% under AM1.5G illumination. Additionally, the high absorption coefficient allows a thin layer, ~1–2 μm, to efficiently absorb incoming light. With these properties in mind, TF-VLS-grown InP is an ideal material for the absorber layer in a single junction solar cell. The TF-VLS process is shown in Figure 1.

First, the non-epitaxial substrate is prepared, where we have used 1 μm of sputtered molybdenum (Mo) on a glass substrate. Then ~1 μm thick indium (In) is deposited by a physical vapor deposition process along with a ~50 nm silicon oxide (SiOx) cap. The entire stack is heated to a temperature of 450–750 °C. In this temperature range, the In is liquid and the SiOx cap confines it and prevents it from dewetting. Phosphine (PH3) gas is then introduced. The phosphorus (P) containing vapor diffuses through the SiOx cap and into the In liquid. Upon supersaturating the In liquid, InP solid precipitates out of the solution. Each InP nuclei then grows rapidly in the lateral direction until the entire In film is converted to InP. Crucially, the InP nuclei are spaced tens to hundreds of microns apart, which determines the lateral grain size. The large nuclei spacing enabled in the TF-VLS process arises from the creation of large P depletion zones around each nuclei [6].

Once an InP nucleus is formed whose size exceeds the critical threshold (~4 atoms) [5], it rapidly consumes the phosphorous atoms and sub-critical nuclei in its spatial proximity because of the rapid diffusion of P atoms in liquid In (as compared with the flux of incoming P atoms through the solid-state SiOx cap). This P depletion zone prevents further nucleation events and growth of other precipitates to beyond the critical threshold size in the surrounding area and thus results in large grain sizes. This process results in homogenous polycrystalline thin-films with ~2 μm thickness (determined by the thickness of initial In film) and lateral grain sizes of 50–1000 μm. Further, patterning a nucleation promoter can selectively control where the InP nucleates [6]. As-grown InP is n-type due to native defects that stabilize the Fermi level closer to the conduction band. Alternatively, it can also be doped p-type in situ or ex situ. Conceptually, in an in situ process, a p dopant such as Zn, Cd, or Mg can be introduced in the vapor phase along with the PH3. Ex situ doping can be accomplished similarly using a gas-phase diffusion process. Because of the fast diffusion of P inside liquid In, the rate-limiting step is diffusion of P through the SiO2 cap layer. So, for a given cap thickness, the growth rate is controlled by the partial pressure of P vapor. Growth times are <5 min at 10 Torr PH3 partial pressure and 750 °C, and this can be shortened at industrial scale with higher PH3 partial pressures. From this process overview, TF-VLS growth clearly addresses the three main cost components of traditional III-V manufacturing: sputtered Mo replaces the epitaxial wafer substrate, physical vapor deposition can be used in efficiently, and the growth throughput can be much higher.

The optoelectronic quality of TF-VLS-grown InP approaches that of single crystal [5]. Figure 2 shows the 1-sun implied open-circuit voltage (VOC) and external luminescence efficiency of n-type and p-type VLS InP compared with single crystal wafers of comparable doping. The VOC implied by the measured photoluminescence efficiency is the maximum open-circuit voltage attainable given perfect contacts [7]. For as-grown TF-VLS n-InP, the measured implied VOC is ~930 mV, which is only 40 mV below the value obtained for a single crystal InP reference of similar doping concentration. For ex situ Zn doped TF-VLS p-InP, the implied VOC currently reaches ~870 mV (Figure 2). With a short-circuit current density (JSC) of 32 mA/cm² and fill factor of 81%, which are close to what we have achieved on reference solar cells made from single crystal p-InP [8], the projected maximum efficiencies for n-type and p-type TF-VLS InP are 24.1% and 22.6%, respectively.

1.2. Current cell structure and benchmark cell performance

A cell based on TF-VLS InP can be configured as an n-body or p-body device, with the n-body configuration...
more desirable due to the higher implied $V_{OC}$ from luminescence efficiency measurements. However, a suitable window layer deposited by a non-epitaxial process has not yet been demonstrated for an $n$-body cell. For a $p$-body cell, amorphous titanium oxide ($n$-TiO$_2$) is an effective window layer, and our previously published $n$-TiO$_2$/p-InP single crystal wafer-based devices have reached an efficiency of 19.2% [8]. We have adapted this cell structure for our TF-VLS-grown InP, as shown in Figure 3(a). A 60 nm indium tin oxide (ITO) layer serves as our transparent conducting front electrode and anti-reflection coating. Below that is a 10 nm layer of $n$-TiO$_2$, which is deposited by atomic layer deposition. TiO$_2$ has an optical band gap of 3.4 eV, making it a high-transparency window layer. Furthermore, the conduction bands of TiO$_2$ and InP are well aligned, which enables this heterojunction to freely extract minority electrons from the InP and block majority holes [8]. The TiO$_2$ role is similar to that of the cadmium sulfide window layer in copper indium gallium selenide (CIGS) or cadmium telluride (CdTe) cells. Next is our 2 $\mu$m TF-VLS $p$-InP absorber layer, which was ex situ doped using surface diffusion of Zn vapor at 425 °C. Finally, 1 $\mu$m Mo serves as the back contact.

As seen in Figure 3(b), preliminary $p$-body cells have reached AM1.5G power conversion efficiency of 12.3%, open-circuit voltage of 675 mV, short-circuit current density of 29.9 mA/cm$^2$, and fill factor of 61.0% with no finger grid over an area of 1 x 1 mm$^2$ defined by the ITO front electrode. For comparison, our reference cells fabricated on single crystal $p$-InP have reached power conversion efficiency of 19.2%, open-circuit voltage of 785 mV, short-circuit current density of 30.5 mA/cm$^2$, and fill factor of 80.1% with finger grid over an area of 5 x 5 mm$^2$ [8]. For both the TF-VLS and single crystal cell, the $J_{SC}$ is short of the ~35 mA/cm$^2$ limit for InP under AM1.5G. Although the open-circuit voltage of the TF-VLS cells is respectable for a first iteration, it is below the optically implied $V_{OC}$; thus, further improvements in the contacts are both possible and necessary. Because of the current growth scheme [5], Mo is the back contact to $p$-InP, but this is not ideal and may be a source of $V_{OC}$ loss. The optimal contact technology and heterojunction structures for TF-VLS-grown material are still under development, but with maturation, could allow cells to approach the implied $V_{OC}$.

1.3. Proposed module architecture and manufacturing process flow

To perform a full module cost analysis, we are proposing an adaptation of a standard process for producing monolithically integrated CdTe or CIGS modules. These have been demonstrated already in high-volume production [9,10] and are proven low-cost technologies on a $/m^2$ basis [11–13]. Because of similar equipment requirements as for CIGS or CdTe growth, TF-VLS growth should be able to use comparable tools. Additionally, the cells can be adapted to use existing module encapsulation and downstream processes. This minimizes process up-scaling risk and allows TF-VLS cells to benefit from advancements in module technology as well.

A common misconception is that large-scale use of III-Vs is infeasible because the group III components are simply too expensive to be economical. However, with a move to thin-films and high materials utilization efficiency, this is not necessarily the case. The proposed cell structure used for cost analysis is similar to the one presented in the previous section with adjustments to allow for monolithic integration. The complete module and proposed process steps are shown in Figure 4.

To begin, a 1 $\mu$m thick layer of Mo is sputtered onto a soda lime glass (SLG) substrate followed by the P1 laser scribe to isolate the back contacts for each cell. This thickness was chosen based on the thickness of the Mo back contact in CIGS modules today, which typically ranges from 240 nm to 500 nm. Next, a 1 $\mu$m thick layer of In is sputtered, along with a 50 nm SiO$_x$ cap on top. Then, the TF-VLS growth is performed and afterwards the SiO$_x$ cap is removed by a hydrofluoric acid (HF) rinse. The
10 nm TiO₂ layer is sputtered on, followed by the P2 scribe to the Mo substrate. ITO sputtering and the P3 scribe complete the monolithic cells. The module is then finished with standard busbar connections, ethylene vinyl acetate and front glass encapsulation, and edge seals. The cost breakdown for each step is presented in the next section.

2. MANUFACTURING COST ANALYSIS

In order to help illuminate the cost drivers of this technology and understand its potential to compete with existing systems, the authors performed a manufacturing cost analysis of the InP module architecture described in Section 1. In order to compute these costs, the authors map out a potential process flow for manufacturing these modules at scale, shown in Figure 4(b), based on conversations with manufacturers and experts in both industry and academia. Then, the cost of ownership for each step, which includes the materials, labor, depreciation, utilities, and maintenance costs are computed using a bottom-up cost model developed at the National Renewable Energy Laboratory. These input data are gathered from material suppliers, equipment vendors, and industry, and then aggregated and anonymized because the data is often business sensitive. Step-by-step costs are then combined to obtain the total module costs. All calculations are performed in Excel. More detailed information on and examples of this cost modeling approach can be found in [1].
2.1. Module cost components

As seen in Figure 5, for the benchmark 12% efficient module, the projected cost per watt peak is estimated at $0.66 assuming 500 MW(DC) annual production and US manufacturing. $0.19/W(DC) of the total, or 29%, is due to the TF-VLS growth process. The assumed global effective yield is 95%, meaning 5% of total modules produced are discarded. In reality, the yield losses are measured at several points in the manufacturing process flow and the defective modules removed at earlier points in the process, so that the cost of the full yield loss is not incurred at all steps. The cost of materials and equipment for each step is based on a survey of material suppliers, equipment vendors, and industry members. Labor counts were similarly based on discussion with industry and equipment vendors and are closely matched to industry norms. It is important to note that these cost calculations assume US manufacturing and the purchase of new equipment for each step; lower costs can oftentimes be realized through the purchase of used or refurbished equipment. We assume an unskilled US labor rate of $11.55/h and a skilled labor rate of $21.81/h with 33% benefits on wage and salary, although the actual wage rates will vary by location within the USA. On an area basis, the projected cost is ~$79/m² for the entire module and ~$23/m² for the TF-VLS steps alone. The modules in our analysis are assumed to be monolithic modules with a size of 1.2 × 0.6 m².

2.2. Cost breakdown of indium phosphide thin-film vapor–liquid–solid growth

In this section, we describe assumptions made in analyzing each step, the cost advantages of the TF-VLS growth process versus MOCVD, as well as examine the similarities to CdTe and CIGS deposition. In traditional MOCVD, the three major components that substantially impact costs are the epitaxial wafer substrate, low materials utilization efficiency, and low throughput resulting in high capital depreciation costs [3,4]. On the other hand, thin-film technologies such as CdTe and CIGS avoid all three issues, and thus are able to achieve a lower cost per square meter [11–13]. As the TF-VLS growth process for III-Vs parallels that of CdTe and CIGS deposition, it gains many of the same advantages. The entire TF-VLS growth process can be split into five main steps as shown in Figure 6 and detailed in the National Renewable Energy Laboratory bottom-up cost model.

First, a simple sputtered Mo on glass replaces the epitaxial wafer as the growth substrate. This represents a substantial manufacturing process gain as there is no longer the upfront cost of the wafer, re-surfacing, or breakage costs. The cost of 3.2 mm tempered SLG is estimated as $6.26/m², and the sputtered Mo is 250 nm thick. We assume the throughput of sputtering the Mo is 120 modules per hour and that the Mo target price is $150/kg. Next, the In layer is sputtered onto the SLG substrate. Here, the starting assumption is 1 μm of elemental In (5 N purity) is sputtered from rotatable source targets priced at $910/kg. While our preliminary lab cells used 6 N purity In, commercial CIGS cells are using 5 N In. Pricing will of course vary somewhat by supplier and region of the world, and the price of In is typically quite volatile. For the rotary sputtering process, our model assumes a target utilization of 85% and a substrate collection efficiency of 85%. Based on our interviews, this utilization is typical for thin film rotary sputtering, although this varies depending on the module size, sputtering tool, and configuration of the modules in the tool during deposition. High-efficiency planar targets may also be used for deposition, but this is not explored in this work. We also assume that the In remaining on the target and collected from the chamber walls is recycled with a net value of 75% of the original material value, based upon advice provided by a relevant manufacturer. Even as the highest cost step in the process, the materials cost is still

![Figure 5. Indium phosphide (InP) module cost breakdown. Total areal cost is $79/m², with $23/m² coming from non-thin-film vapor–liquid–solid (TF-VLS) steps.](image-url)
$0.07/W(DC) or $8.40/m², which is quite low compared with the use of metalorganic precursors such as trimethylindium. The price of In is sensitive to supply chain dynamics from competing uses such as ITO and CIGS [14]. It is possible to move to electrodeposited In [15] in the future with a goal of realizing higher materials utilization efficiency and increased ease of recovery. This would help to further decrease the sensitivity of module cost to In price increases. Subsequently, the 50 nm SiOₓ cap layer is sputtered on top.

The second major step is phosphorization. As the process requirements for this step are very similar to those of the selenization/sulfurization steps in CIGS manufacturing, we assume similar tools can be used. Namely, the temperatures, pressures, and types of precursor are very similar. Assuming a selenization and sulfurization furnace with a 2-min reaction time, 50-min heating time, 75-min cooling time, and 40-module batch size, the estimated cost for phosphorization is $5.38/m², or $0.045/W (DC) at 12% AM 1.5G power conversion efficiency. Additionally, a utilization efficiency of 70% for the phosphine was assumed, with a cost of $350/kg. Finally, an HF rinse removes the SiOₓ growth cap. Here, we assume a 10:1 solution of H₂O:HF with a materials cost of $0.80/liter, and equipment costs and HF usage sourced from equipment vendors. The time to etch the 50 nm thick SiOₓ layer was assumed to be 2.1 min with a 0.2-min set-up time and 40 modules per batch.


The growth process time depends on the temperature and PH₃ partial pressure (or equivalently, concentration) and is independent of the total area. The TF-VLS process is also relatively insensitive to the flow patterns in the chamber, dramatically simplifying chamber design compared with MOCVD. Additionally, by tuning the SiOₓ cap thickness and porosity, P diffusion through the cap can be maximized. This diffusion rate is also controlled by the difference in chemical potential between the P in the vapor phase and the P dissolved in the In liquid. This means that the growth rate, to first order, is linearly dependent on the P partial pressure. For reference, a lab scale process with identical SiOₓ cap and In thicknesses (50 nm/1 µm), yet a low PH₃ partial pressure of 10 Torr, already results in growth time of <5 min. For industrial scale processes of ~100 Torr PH₃ partial pressure, growth times of <2 min are reasonable. In the future, further gains in throughput may be found by combining load lock systems to minimize the time spent ramping the temperature.

2.3. Module cost per watt projections

For the benchmark 12% efficient module case, the total module manufacturing costs are estimated to be $0.66/W(DC). With modest improvements in device performance within the mid-term, this would decrease to $0.57/W(DC) for 15% efficient modules. In the long-term, assuming suitable hole-selective window layer and n-type InP absorber layer with 930 mV open-circuit potential (the optically implied Voc) and 24% efficient modules, $0.36/W(DC) could be attainable. For comparison, while the future costs of photovoltaics is uncertain, it is generally expected that module manufacturing costs will decline to $0.40–$0.50/W in the next several years, with multiple companies targeting costs near or below $0.40/W before or by 2018 [16–18]. The estimated minimum sustainable prices (MSP) for these modules were also computed and are shown in Figure 7. All MSPs assumed sales, general, and administrative; and research and development costs of 7.6% and
4.3% of sales price, respectively. A 28% corporate tax rate, and 7-year, straight-line depreciation for equipment is also assumed. Note that sales, general, and administrative and research and development costs can vary dramatically by company and according to the stage of a company’s developments; our assumptions are based on the FY14 year-end financial statement of First Solar, the largest thin-film photovoltaics manufacturer. The nominal weighted average cost of capital was assumed to be 15% for the benchmark case, 13% for the short-term case, and 9% for the long-term case, reflecting a potential for decrease in cost of capital as the technology matures and the perceived risk decreases. However, the weighted average cost of capital and its evolution in time also contain significant uncertainty, and thus the bulk of this analysis is more focused on module cost rather than MSP.

3. CONCLUSIONS

In this paper, we have presented a cost analysis of InP solar cell modules manufactured with the TF-VLS growth process on a standard monolithic thin-film module platform. The initial cell architecture is a simple stack of ITO/TiO2/p-InP/Mo. At an annual production capacity of 500 MW(DC), the short-term benchmark case of 12% efficient modules is expected to reach a manufacturing cost of $0.66/W(DC), while the long-term potential case of 24% efficient modules is expected to reach a cost of $0.36/W(DC). We also demonstrate that the TF-VLS growth process is ideally suited to make more economical use of the group III metal In by both using it in elemental form and with higher utilization efficiency. TF-VLS growth addresses the three main cost components associated with traditional MOCVD growth: the epitaxial wafer substrate, low utilization efficiency of expensive metalorganic precursors, and high capital depreciation costs due to low throughput. Avoiding these issues enables a lower manufacturing cost of ~$23/m² for the example of InP (TF-VLS steps only). In the future, it is also possible to use the TF-VLS growth process to produce low-cost epitaxial growth substrates for subsequent MOCVD growth of III-V heterojunctions to produce more complex multijunction devices. The TF-VLS process also has broader applicability and may be used as the base for other industries such as integrated circuits, solid-state lasers, solid-state lighting, and power devices.

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REFERENCES


