Techno-economic analysis of the use of atomic layer deposited transition metal oxides in silicon heterojunction solar cells

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Abstract
The industry for producing silicon solar cells and modules has grown remarkably over the past decades, with more than a 100-fold reduction in price over the past 45 years. The main solar cell fabrication technology has shifted over that time and is currently dominated by the passivated emitter and rear cell (PERC). Other technologies are expected to increase in market share, including tunnel-oxide passivated contact (TOPCon) and heterojunction technology (HJT). In this paper, we examine the cost potential for using atomic layer deposition (ALD) to form transition metal oxide (TMO) layers (MoO$_x$, TiO$_x$, and aluminium-doped zinc oxide [AZO]) to use as lower cost alternatives of the p-doped, n-doped and indium tin oxide (ITO) layers, respectively, the layers normally used in HJT solar cells. Using a bottom-up cost and uncertainty model with equipment cost data and process experience in the lab, we find that the production cost of these variations will likely be lower per wafer than standard HJT, with the main cost drivers being the cost of the ALD precursors at high-volume production. We then considered what efficiency is required for these sequences to be cost effective in $/W and discuss whether these targets are technically feasible. This work motivates further work in developing these ALD TMO processes to increase their efficiency towards their theoretical limits to take advantage of the processing cost advantage.

KEYWORDS
atomic layer deposition, passivating contact solar cell, techno-economic analysis, transition metal oxide

1 | INTRODUCTION

The production and market for photovoltaics (PV) modules have increased in scale and reduced in cost significantly over the past decades, as shown in industry reports such as the International Technology Roadmap for Photovoltaics (ITRPV), which estimated cumulative shipments of 789 GW at the end of 2020, and a more than 100-fold reduction in market price (in $/W) over a 45-year period. Demand for PV modules is expected to remain strong.

The technology used to produce the solar cells in these modules continues to change over time. Figure 1 shows the current and projected market share of different silicon cell technologies of two industry
roadmaps—the ITRPV\(^2\) produced in Europe and CPIA\(^3\) produced in China. The once-dominant aluminium back surface field (Al-BSF) technology was a small fraction of production in 2020, and the market has been taken over by the passivated emitter, rear-contact (PERC) technology. These roadmaps predict an increasing market share for passivated contact technologies such as tunnel-oxide passivated contact (TOPCon) and heterojunction technology (HJT, sometimes abbreviated SHJ). The reason for interest in these passivating contact technologies is their higher efficiency potential because of improved full area surface passivation and the avoidance of a silicon-metal contact, leading to reduced surface recombination and higher voltages.\(^4\) Many research groups and manufacturers are developing these technologies, with high efficiencies reported at both lab and industrial scale. A time series of high lab efficiencies are summarized in Figure 2, with references in the caption. Industrial-scale records at time of writing are 25.2% for TOPCon\(^5\) and 26.3%\(^6\) for SHJ solar cells.

In this work, we focus on a related group of as yet uncommercialized technologies—those utilizing transition metal oxides (TMOs) as part of passivating electron and hole contacts.

For research groups developing passivated contact technologies, it is important to consider economic factors, as the industry is not only focused on efficiency but also considers the cost (\$/cell or \$/W). It is helpful in this case to carry out cost analysis of processes that are under development to understand the commercial potential as well as barriers to adoption. The insights from such analysis can help focus research on the key factors that must be improved in order that the new technology can become competitive.

In this work, we review solar cell manufacturing bottom-up cost data from the National Renewable Energy Labs (NREL), including a recent 2021 update. We build on this to estimate the cost of possible implementations of novel TMO layers in passivating electron and hole contacts and as transparent conductive oxide (TCO) layers. We first outline the technical advantages and disadvantages of these structures, including the potential solar cell efficiency. We then complete a cost analysis of a range of proposed structures to identify process cost improvement and efficiency improvement targets for these technologies.

2 | NREL BOTTOM-UP COST DATA FOR ALTERNATIVE SOLAR CELL TECHNOLOGIES

The NREL group has published process step cost data for many PV production sequences using a bottom-up cost of ownership model. For example, in 2019, Woodhouse et al\(^25\) analyised the cost of manufacture in 2018 for polysilicon, wafer, solar cell and module fabrication. For solar cell fabrication, two industrial sequences were included, Al-BSF and PERC, and three emerging sequences, n-PERT, HJT, and interdigitated back contact (IBC). The NREL methodology is to seek detailed data from equipment suppliers and PV manufacturers to give the most accurate cost result possible, but for reasons of confidentiality, the exact bottom-up input parameters are not revealed.

In this paper, we provide recent (2021) data from the NREL cost models for PERC, n-PERT and HJT sequences as well as new analysis of the TOPCon structure. These data have been provided by NREL in summary form based on the most up-to-date data from the NREL model as at December 2021. The detailed step-by-step cost data in graphical and tabular form are shown in the supporting information, Section S1. In Figure 3, we show a summary of the 2018 and 2021 NREL cost data as well as cost data obtained from PV Infolink.\(^26\)

For each of the technologies shown, there has been an improvement in solar cell efficiency and a reduction in cost over time. However, in 2018 and 2021, advanced solar cell technologies such as n-PERT, HJT and TOPCon have higher manufacturing cost per watt than the market-leading PERC technology. In particular, HJT is higher cost because of the need for large amounts of expensive low-temperature silver paste and a higher depreciation cost from the expensive PECVD equipment. We note that in the market, the manufacturing cost in \$/W is not the only determining metric for...
competitiveness. For example, (i) solar cells with higher efficiency will result in higher efficiency modules which require comparatively less space and system mounting hardware and so a lower balance of systems cost, and (ii) modules that degrade more slowly or have lower failure rates will have increased electricity production, reducing the levelized cost of electricity (LCOE).

Examining the NREL cost data between 2018 and 2021: (i) There have been significant reductions in depreciation costs for PERC, n-PERT and HJT. This is due to decreases in equipment cost, in particular because of an increasing number of lower cost Chinese based equipment suppliers, as well as improvements in equipment throughput. (ii) Metallization paste costs reduced significantly for PERC due to reductions in usage (g/wafer), less so for n-PERT. There was an increase for HJT due to a cost update for the low-temperature paste cost being significantly higher than the cost of the input silver. (iii) Other materials costs are fairly similar between technologies and have not changed significantly and (iv) for labour cost, the headcount per production capacity decreased due to increases in equipment throughput, but this was counteracted by an increase in the assumption for labour rates in China.

PV Infolink has 2021 estimated manufacturing cost data for PERC, TOPCon and HJT, but the data are not broken down into individual cost categories. The NREL cost results are broadly in line with this, with the advantage of having a detailed step-by-step resolution as well as categorization into cost components. We start with this NREL data as a strong basis to build additional cost estimates applied to novel TMO solar cell architectures.

3 | SOLAR CELL ARCHITECTURES EMPLOYING TMOs

The data from industry roadmaps indicate increasing manufacturing volumes for TOPCon and HJT. Within research institutes, there is interest in exploring possibilities to increase the efficiency or reduce the cost of these types of solar cells. Recently, TMOs have emerged as an appealing alternative for doped silicon films in passivating contacts due to their relatively high bandgap resulting in significantly lower parasitic optical absorption compared to heavily doped a-Si layers on the front and rear sides of HJT solar cells. To compensate for the low lateral conductivity of a-Si, indium tin oxide (ITO) is usually used as the TCO on these structures, but alternative TMO layers are of interest if they can reduce reliance on the high-cost indium.

3.1 | TMOs—benefits for silicon solar cells

Although HJT structures have achieved high efficiency by employing doped a-Si layers that provide excellent surface passivation, a high parasitic optical absorption of a-Si layers limits the photo-generated current and thus limits the performance of the solar cells. TMOs usually have a wider bandgap than doped a-Si, resulting in less parasitic absorption. TMOs can be used in both passivating electron and hole contacts due to their wide range of work functions and band alignment offsets. In Figure 4A, we show various TMOs and their relative band alignment to silicon, and in Figure 4, we show a band diagram of these layers in a full solar cell structure. When applying MoOx on silicon, the significant difference in workfunction results in band bending in silicon, thereby increasing the hole concentration at the interface resulting in a high hole conductivity and low electron conductivity. In combination with an a-Si interface passivation layer, this results in a very effective passivating hole contact with a record efficiency of 23.5% to date. In the case of TiOx, which has been reported to be an effective electron transport layer on silicon with excellent passivation effect, we can see that there is a close band offset with the conduction band of silicon, while there is a large discrepancy in the valence band offset, providing high conductivity to electrons transporting from TiOx into the silicon bulk, while holes are effectively repelled. It is also verified that the passivation effect on the silicon surface can be preserved by the TiOx/i-a-Si:H stack after an annealing process of up to 300°C, while providing a low contact resistivity of around 30 mΩ cm². TCOs are often used as part of the passivating contact structure to compensate for the relatively low lateral conductivity of the thin TMO or doped a-Si layers used. In addition, the TCO layer can serve as an antireflection coating, as in the case of a HJT solar cell.

Because of these potential advantages, there has been interest in demonstrating the use of these layers in silicon solar cells. Titanium oxide (TiOx), niobium oxide (NbOx), and tantalum oxide (TaOx) have been used in passivating electron contacts, while molybdenum oxide (MoOx), tungsten oxide (WOx), vanadium oxide (VOx) and chromium oxide (CrOx) have been used in passivating hole contacts.
in silicon solar cells to replace the highly doped a-Si layers in HJT structures. ITO, doped indium oxide [In$_2$O$_3$:X (X are dopants such as H, Sn, and Zn)], and doped zinc oxide (ZnO: Al, Be, B, and Ga) have been considered as TCOs for solar cell structures.

Figure 2 details high efficiencies achieved using MoO$_x$ and TiO$_x$ in the laboratory. A full area atomic layer deposited (ALD) TiO$_x$ was successfully applied in an n-type silicon solar cell device as an electron selective contact and achieved an efficiency of 22.1%. More recently, a 23.5% conversion efficiency was reported for a HJT structure with MoO$_x$ instead of boron-doped a-Si. A higher short-circuit current density was achieved in the solar cell with MoO$_x$ than the standard HJT solar cell, due to the lower parasitic absorption in the MoO$_x$, with efficiency equivalency between the standard HJT and MoO$_x$ structures. The fundamental limit imposed by TMO passivating electron and hole contacts is above 27%, indicating that there is still ample room for improvement.

Various TMOs have been explored as an indium free TCO, but AZO has the closest electro-optical properties to ITO. Tests have shown that sputtered AZO has the same efficiency potential as ITO. Other reports show that ALD-deposited AZO can give equivalent efficiency to ITO for HJT solar cells when used on either the front, rear, or both sides of the solar cell, with efficiency values of 23% and 22.5%. Efficiency parity has also been shown for large area solar cells, with efficiency values over 21.1%.

Some groups have applied passivating contacts with TMOs for both the electron and hole contacts. These structures are referred to as dopant-free asymmetric heterocontacts (DASH) solar cells, A fundamental and theoretical investigation suggest that the band structure is suitable for next-generation high-efficiency solar cells.

Recently, Bullock et al. demonstrated a solar cell with an electron and hole selective TMO contact—TCO/MoO$_x$ (thermally evaporated)/i-a-Si:H/c-Si/i-a-Si:H/TiO$_x$ (ALD)/LiF/Al that resulted in an efficiency of 20.7%. Although the TCO used was ITO, AZO is expected to have efficiency parity to ITO when implemented in DASH structures.

When considering the cost and efficiency potential of these structures, it is important to identify a benchmark efficiency for standard HJT solar cells as a comparison. We have selected the 26.3% efficiency HJT solar cell, which is the highest lab efficiency we have identified for non-IBC HJT solar cells.

### 3.2 TMO fabrication methods

There are a variety of techniques to deposit thin TMO layers, with a comparison of methods summarized in Table 1. Sol-gel spin coating is widely used in depositing TMOs such as ZnO and TiO$_x$ as transport layers in organic solar cells, perovskite solar cells or dye-sensitized solar cells. However, it is usually challenging to achieve a uniform thin-layer such as required in passivating contact layers for silicon solar cells with only a few nanometres. Thermal evaporation and plasma-enhanced chemical vapour deposition (PECVD) are also popular methods to deposit thin TMO layers in solar cells. Nonetheless, its fast surface reaction rate and shadowing effects limits the conformity of the films.

**TABLE 1** TMO deposition methods and their advantages and disadvantages

<table>
<thead>
<tr>
<th>Method</th>
<th>ALD (batch)</th>
<th>ALD (spatial)</th>
<th>PECVD</th>
<th>Sputtering</th>
<th>Thermal evaporation</th>
</tr>
</thead>
<tbody>
<tr>
<td>Uniformity</td>
<td>Superior</td>
<td>Superior</td>
<td>Good</td>
<td>Very good</td>
<td>Poor</td>
</tr>
<tr>
<td>Substrate temperature (°C)</td>
<td>50–100</td>
<td>20–200</td>
<td>200–300</td>
<td>20–200</td>
<td>50–100</td>
</tr>
<tr>
<td>Deposition rate (Å/s)</td>
<td>0.1–1</td>
<td>1–10</td>
<td>10–100</td>
<td>1–10</td>
<td>1–20</td>
</tr>
<tr>
<td>Impurity level</td>
<td>Very low</td>
<td>Very low</td>
<td>Very low</td>
<td>Low</td>
<td>high</td>
</tr>
</tbody>
</table>

Note: Exact values for uniformity and impurity levels will change according to equipment design, but commercially equipment uniformity specifications of >98% for ALD and >95% for both PECVD and sputtering have been found.

Source: Data obtained and adapted from French et al.
successfully been introduced in PERC solar cell production with superior performance, as well as lower processing cost, which is crucial for industrialisation.\(^\text{80}\) Within the PV industry, batch ALD is favoured over spatial ALD, and this trend is expected to continue.\(^\text{1}\)

Compared to the TMO layers in passivating contacts, optimized TCO layers are significantly thicker (\(~75\) nm) to ensure high bulk conductivity while avoiding excessive parasitic absorption and having good antireflective properties. The industrial standard deposition technique for ITO layers is magnetron sputtering.\(^\text{81}\) Sputtering results in the desired layer quality while being economical for thicker layers, but it also induces near-surface defects.\(^\text{82}\) On the other hand, ALD causes negligible process-induced interface defects.\(^\text{83}\) Offers high control of the film thickness and composition. Nonetheless, its slow growth rate is still a major economic bottleneck.

ALD growth of indium oxides has additional challenges of high precursor cost, low growth rate and long nucleation time.\(^\text{84,87–91}\) On the other hand, ALD of ZnO is a mature process, and the commonly used Zn precursor is a low-cost chemical that is readily available.\(^\text{82,92}\) Moreover, low-temperature growth of TCO on temperature-sensitive a-Si can be achieved by ALD grown ZnO at temperatures as low as 100\(^\circ\)C.\(^\text{93}\)

With the performance potential of TMOs and a high quality of deposition using ALD, these materials hold promise for industrial applications in HJT devices. Hence, we examine the feasibility of applying ALD TMOs into industry from a perspective of techno-economic analysis. Four solar cell architectures employing TMO layers are proposed based on a standard HJT solar cell structure, as shown in Figure 5. Seq A is a standard HJT, Seq B, C and D replace the p-doped, n-doped and ITO layers with MoO\(_x\), TiO\(_x\), and AZO, respectively. Seq E combines all three ALD TMO layers into one structure. The process sequence assumed for each of these is shown in Table S5.


### 4 | COST ANALYSIS

#### 4.1 | Methods

In order to estimate the cost of implementing the ALD TMO layers in high-volume production, a cost model using a Monte Carlo uncertainty approach was used as described in previous work.\(^\text{95}\) In brief, the method is a bottom-up cost of ownership model that allows for Low, Med and High values for each input instead of assuming a single number. The model then uses a Monte Carlo approach to sample each input variable using a two-half log-normal distribution based on the uncertainty values (Low = 10th percentile, Med = median, High = 90th percentile). The two-half distribution allows the Med value to be the median of the distribution, and the log-normal distribution avoids the generation of negative numbers. Repeating this process many times, the output of the analysis is a distribution of calculated costs together with an understanding of the key factors that lead to cost uncertainty. The Monte Carlo model was run using a python script on a Windows notebook computer. This method is particularly useful when processes are being developed in the lab, where exact data are difficult to obtain, such as (i) material cost and usage at production volumes, (ii) equipment cost and throughput, and (iii) production solar cell efficiencies.

#### 4.2 | Assumptions for ALD TMO deposition

The cost of growing ALD layers is comprised of two main components—(i) the cost to purchase, maintain and operate the equipment (including depreciation of the purchase price, and electricity, labour, and maintenance expenses); and (ii) the cost of the materials, including the ALD precursors.

The cost of equipment is based on data from an equipment manufacturer—Leadmicro’s list price data for the KF Series (KF20000S) industrial batch type ALD system. This fully automated system with auto-guided vehicle and manufacturing execution systems is optimized to produce high-quality ultra-thin aluminium oxide (Al\(_2\)O\(_3\)) for the PERC solar cell rear surface passivation.\(^\text{82}\) This system can be operated between 150\(^\circ\)C and 300\(^\circ\)C for the growth of Al\(_2\)O\(_3\) and other oxides (SiO\(_2\), TiO\(_x\), ZnO) and has a 20,000 wafer/h throughput (for 4-nm Al\(_2\)O\(_3\)) with specified non-uniformity <3%, uptime of 98% and a breakage rate of <0.02%. Although most ALD systems have an inherent drawback of wrap-around deposition, the KF-series have been modified to allow for single-side deposition.\(^\text{82}\) This system has a low tri-methyl-aluminium (TMA) consumption (~1 mg/wafer) and operational costs of RMB 0.03 per wafer. This six-tube batch
system can have maintenance on each tube independently to better suit high-volume manufacturing.

A key factor for equipment running cost per wafer produced is the throughput of the equipment. Since the TMO layers we are evaluating are different from Al₂O₃, it is necessary to adjust the throughput to match the new process. The KF200005 is a six-tube system with a capacity of 1000 wafers per tube. This implies that the 20,000 wafer/h for Al₂O₃ is achieved using a batch time of 18 min. For the new TMO layers analysed here, the thicknesses, cycle times and the number of cycles may not be the same as for Al₂O₃, so we estimate the processing time based on the number of ALD cycles and the cycle time (second per cycle) observed during process development on a laboratory ALD system (Cambridge Nanotech’s Fiji G2). The assumed thickness of the MoO₃ layer was 4 nm based on the record efficiency for this type of structure. For TiOₓ, a 3-nm layer was the optimized thickness. For TiOₓ, it may be possible to use a thinner layer, which would further reduce cost, as 1.5 nm has been used successfully. For AZO, a thickness of 75 nm was selected based on optimal anti-reflective properties. These assumptions and calculated throughput are shown in Table 2, where the throughput for MoO₃ and TiOₓ are similar to that of Al₂O₃. The throughput calculations for AZO were very different for two reasons: (i) the film is significantly thicker, requiring many more cycles, and (ii) the need for deposition on both sides of the wafer. Regarding the latter, since TCOs are needed on both sides of the HJT, we can choose to complete two single-sided depositions or one double-sided deposition. A double-side deposition of AZO is possible in this ALD tool by modifying the wafer loading—the batch size is reduced by one third based on advice from the equipment manufacturer, ensuring sufficient space between the wafers and uniform precursor coverage. Despite the smaller batch size, this is the lowest-cost approach, since both the front and rear sides are deposited simultaneously. These throughput estimates are subject to uncertainty, for example, it may be that a lower or higher purge time is needed per cycle once each process is optimized for industrial scale and throughput. On the negative side, there is a risk that scaling up the lab process to an industrial scale will require longer process times to achieve good uniformity or properly purge the chamber. On the positive side, there may be a way to more densely load the chamber. Overall, we anticipate that the likelihood of faster throughput is lower than the likelihood of slower throughput, and so we use an uncertainty range in the wafer throughput (wafer/h) of +10%/-20% in the cost model.

Other input parameters used were based on the data sheets with uncertainty ranges applied. These are shown in Table 3. For those parameters where information was not readily available, our own estimate with wide uncertainty range was applied. This was done in line with the methodology—if wide uncertainty ranges on certain variables do not introduce significant impact on the resulting cost analysis, then there is no need to further investigate and refine these variables until other uncertainties are reduced. In the supporting information, other factory assumptions are described in Table S6.

To calculate materials costs, we must determine the most appropriate precursors to use and then estimate the usage of the precursor in each ALD process. For MoO₃, we selected the most common Mo precursor bis(tert-butylimido)-bis(dimethylamido)molybdenum because of its capacity to be deposited under low processing temperatures and the flexibility of using oxygen or ozone plasma as co-reactants. For TiOₓ, the most widely used Ti precursors are TiCl₄ and tetrakis(dimethylamino)tin (TDMAT), while water and ozone are used as the oxidant. In our experiments and for the cost modelling, we assume the use of TDMAT, which produces non-corrosive and non-toxic by-products, which is environmentally friendly and safe for industrial use. For AZO, we selected DEZ and TMA as they are the most common Zn and Al precursors, respectively. Regarding precursor usage, while the ALD process can in theory be carried out with nearly 100% precursor utilization, if exactly the correct amount of precursor is released into the chamber, practical considerations such as over-filling the chamber to decrease processing time, less reactive precursors, lower sticking probability, slow nucleation rate and deposition on chamber walls would result in lower utilization.

**TABLE 2** Throughput assumptions for Monte Carlo analysis of ALD deposition costs

<table>
<thead>
<tr>
<th></th>
<th>MoO₃</th>
<th>TiOₓ</th>
<th>AZO</th>
<th>Comment</th>
</tr>
</thead>
<tbody>
<tr>
<td>Batch size (wafers)</td>
<td>6000</td>
<td>6000</td>
<td>4000</td>
<td>For AZO, the double-sided deposition means fewer wafers are loaded each batch.</td>
</tr>
<tr>
<td>Deposition thickness (nm)</td>
<td>4</td>
<td>3</td>
<td>75</td>
<td></td>
</tr>
<tr>
<td>Deposition cycles</td>
<td>36</td>
<td>66</td>
<td>480</td>
<td>Number of cycles based on experimental experience, includes some nucleation cycles.</td>
</tr>
<tr>
<td>Average thickness deposited per cycle (nm)</td>
<td>0.111</td>
<td>0.045</td>
<td>0.156</td>
<td>Note: after nucleation, growth rate is higher than this.</td>
</tr>
<tr>
<td>Cycle time (s)</td>
<td>30.1</td>
<td>30.1</td>
<td>20.1</td>
<td>Includes pulse 1st + purge + pulse 2nd + purge</td>
</tr>
<tr>
<td>Total process time per batch (minutes)</td>
<td>18.1</td>
<td>33.1</td>
<td>161</td>
<td></td>
</tr>
<tr>
<td>Throughput (wafer/h)</td>
<td>19,934</td>
<td>10,873</td>
<td>1493</td>
<td></td>
</tr>
<tr>
<td>Low throughput</td>
<td>15947</td>
<td>8698</td>
<td>1194</td>
<td>20% lower throughput is possible if process cannot be optimized for speed</td>
</tr>
<tr>
<td>High throughput</td>
<td>21,927</td>
<td>11,960</td>
<td>1642</td>
<td>10% higher throughput is possible if the wafer load size could be improved, or cycle time reduced</td>
</tr>
</tbody>
</table>
In a lab environment, utilization rates close to 50% have been reported, but for industrial ALD use, very high utilization of precursors have been claimed for Al2O3 (1 mg/wafer of TMA for 4 nm is close to 100% utilization). The actual utilization rate of precursors is a significant unknown that is best determined by demonstrating an optimized process in industrial-sized equipment. To account for this uncertainty, we base our range of precursor usage on utilization between 70% and 90%. A wafer area of 252 cm² (G1) was assumed to match with the NREL 2021 HJT cost analysis. The assumptions and calculations for precursor usage are shown in Table 4.

The cost of ALD precursors in a research setting is very high since a relatively small amount of precursor (typically 25 g) is purchased at one time. One of the ALD precursors (TMA) is currently used in industrial quantities. For lab quantities (25 g), research institutes pay around US$ 100 /g. In 2016, one report estimated TMA costs at EUR 0.75/g, and industry contacts of the authors suggested that at manufacturing volumes, it is currently possible to purchase this for $0.2/g. This is an extraordinarily large cost reduction. Part of the reason for this low cost is that lower cost ‘solar grade TMA’ can be used instead of semiconductor grade, a cost reduction of 10 times. Other factors are presumably the advantage of economies of scale in manufacturing, handling and transporting the pyrophoric precursor.

For the other precursors, firm large volume pricing information was not available. Making use of the Monte Carlo uncertainty approach, we estimated a range of possible costs for each precursor to understand our sensitivity to this factor. Two approaches were considered for estimating the possible range of the cost of precursors in large volumes. The first option was to assume the cost reduction table.

### Table 3: Other equipment assumptions

<table>
<thead>
<tr>
<th>Tool cost (US$ million)</th>
<th>1.8</th>
<th>1.44</th>
<th>1.98</th>
<th>List price was RMB 12 million. Converted using 0.15 USD per RMB. +10%/−20%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Facilities capital cost (% of tool cost)</td>
<td>20</td>
<td>10</td>
<td>25</td>
<td></td>
</tr>
<tr>
<td>Maintenance (%)</td>
<td>1.5</td>
<td>1</td>
<td>2</td>
<td>Specification of uptime &gt;98%</td>
</tr>
<tr>
<td>Equipment floorspace (m²)</td>
<td>80</td>
<td>70</td>
<td>90</td>
<td></td>
</tr>
<tr>
<td>Electrical power (kW)</td>
<td>100</td>
<td>90</td>
<td>110</td>
<td>Specification is 100 kW</td>
</tr>
<tr>
<td>Maintenance spare parts (% of capex per year)</td>
<td>3.5</td>
<td>2</td>
<td>5</td>
<td></td>
</tr>
<tr>
<td>Operators per tool</td>
<td>0.3</td>
<td>0.1</td>
<td>0.5</td>
<td>System is fully automated, but some operators for loading and monitoring assumed</td>
</tr>
<tr>
<td>Maintenance personnel per tool</td>
<td>3</td>
<td>2</td>
<td>4</td>
<td></td>
</tr>
<tr>
<td>Oxygen flow (standard lpm)</td>
<td>20</td>
<td>10</td>
<td>50</td>
<td>For ozone generation—per tool</td>
</tr>
<tr>
<td>Nitrogen flow (standard lpm)—pump purge</td>
<td>240</td>
<td>240</td>
<td>240</td>
<td>Per tool</td>
</tr>
<tr>
<td>Nitrogen usage (standard litres)—load lock vent</td>
<td>3500</td>
<td>3500</td>
<td>3500</td>
<td>Per tube vent</td>
</tr>
</tbody>
</table>

Note: Author estimates from data sheets and personal communication, with uncertainty ranges added to account for possible variations.

| TABLE 4 Data on precursor usage |
|----------------------------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
|                                | MoOx (1:2.9)    | TiOx            | ZnO (bifacial AZO) | Al2O3 (bifacial AZO) | Comment/Source |
| Thickness (nm)                 | 4               | 3               | 142.5            | 7.5              | See text       |
| Wafer area (cm²/wafer)         | 252             | 252             | 252              | 252              | M3 wafer       |
| Volume of layer (cm³/wafer)    | 0.00010         | 0.00008         | 0.00359          | 0.00019          | Calculated     |
| Density of layer (g/cm³)       | 4.0             | 4.23            | 5.6              | 3.987            | 104.106.107    |
| Mass of layer (g/wafer)        | 0.00040         | 0.00032         | 0.0201           | 0.00075          | Calculated     |
| Molar mass of layer (g/mol)    | 142.8           | 79.9            | 81.4             | 101.9            | Data sheets    |
| Moles of layer (mol/wafer)     | 2.82E-06        | 4.00E-06        | 0.000247         | 7.39E-06         | Calculated     |
| Precursor name                  | (tBuN)2(NMe2)2Mo | TDMAT           | DEZ              | TMA              |                |
| Molar mass of precursor (g/mol)| 326.33          | 224.00          | 123.00           | 144.20           | Calculated     |
| Ratio of moles of precursor to moles of layer | 1 | 1 | 1 | 2 |                |
| Mass of precursor (100% utilization) (g/wafer) | 0.00092 | 0.00090 | 0.0304 | 0.00213 | Calculated     |
| Mid usage (g/wafer)—80% material utilization | 0.00115 | 0.00112 | 0.0380 | 0.00266 | Calculated     |
| Low usage (g/wafer)—90% material utilization | 0.00102 | 0.00100 | 0.0338 | 0.00237 | Calculated     |
| High usage (g/wafer)—70% material utilization | 0.00132 | 0.00128 | 0.0434 | 0.00304 | Calculated     |
between lab and industry pricing for TMA would be repeated for other precursors—a roughly 500 times cost reduction. This is an optimistic approach, and it assumes that a lower purity ‘solar grade’ precursor could similarly be used, and that the same drivers for economies of scale are available. Instead, we use a more conservative approach, described in other work, and use an economies of scale model to estimate high-volume pricing based on low volume pricing.

As in previous work, an uncertainty range of the ‘doubling factor’ (the cost multiplier for every doubling of purchase volume) can be used to identify a range of high-volume prices. In this work, a production volume of 210 million solar cells per year (∼1 GW/yr) is assumed, and the extrapolated price range using this method is also shown in Table 5. The range of possible doubling factors was varied between a low of 0.75 and a high of 0.95. This resulted in a cost reduction range between 3 and 30 times. This range was cross checked with indicative pricing (100 kg/year, ex works) from a supplier—TDMAT at $5/g was close to the mid value, and DEZ at $0.8/g was between the mid and high value. No volume cost information was available for the Mo precursor used in our lab. Because there are still many unknowns to these costs, such as required purity, shipping and packaging, we kept the large uncertainty range within the Monte Carlo model to check sensitivity to this pricing, and this is discussed further in the sections below.

The other materials used in ALD are nitrogen (for pump purging and chamber venting) and oxygen (to generate ozone as the oxygen source). In experimental work, DI water (negligible cost) was used as the oxygen source; however, advice from industry was that ozone-based processes are preferred in production due to reduced purge times and improved uniformity. Nitrogen and oxygen usage per solar cell are calculated based on estimated flow (Table 3) and equipment throughput.

### Table 5 Data on precursor costs at manufacturing scale

<table>
<thead>
<tr>
<th>Precursor Name</th>
<th>Mo</th>
<th>TDMAT</th>
<th>DEZ</th>
<th>TMA</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lab purchase amount (g)</td>
<td>25</td>
<td>25</td>
<td>2000</td>
<td></td>
</tr>
<tr>
<td>Lab purchase cost (US$/g)</td>
<td>76.4</td>
<td>33.5</td>
<td>2.2</td>
<td></td>
</tr>
<tr>
<td>Annual usage (wafers/year)</td>
<td>210,000,000</td>
<td>210,000,000</td>
<td>210,000,000</td>
<td></td>
</tr>
<tr>
<td>Mid estimate usage (g/wafer)</td>
<td>0.00115</td>
<td>0.00112</td>
<td>0.0380</td>
<td></td>
</tr>
<tr>
<td>Annual precursor usage (g/year)</td>
<td>241,868</td>
<td>235,338</td>
<td>7,976,520</td>
<td></td>
</tr>
<tr>
<td>Number of doublings of purchase quantity</td>
<td>13.2</td>
<td>13.2</td>
<td>12.0</td>
<td></td>
</tr>
<tr>
<td>Mid projected cost (0.85 doubling factor) US$/g</td>
<td>8.9</td>
<td>3.9</td>
<td>0.31</td>
<td>0.20</td>
</tr>
<tr>
<td>Low projected cost (0.75 doubling factor) US$/g</td>
<td>1.7</td>
<td>0.75</td>
<td>0.07</td>
<td>0.15</td>
</tr>
<tr>
<td>High projected cost (0.95 doubling factor) US$/g</td>
<td>39</td>
<td>17</td>
<td>1.2</td>
<td>0.3</td>
</tr>
</tbody>
</table>

Note: For TMA, industrial scale cost data are available, so the economies of scale model is not used.

### Figure 6 Cost breakdown for ALD TMO layer alternatives

Panel (A) shows the cost of the MoO₃ layer compared to the front PECVD a-Si:H layers, (B) shows the cost of the TiO₂ layer compared to the rear PECVD a-Si:H layers and (C) shows the double-sided ALD AZO layer cost compared to the front and rear Sputter ITO layers it replaces. For each cost component, the height of the bar is the median value, and the error bars show the 10th and 90th percentile values of that cost component to indicate the uncertainty. The cost components are offset to allow easier viewing of the error bars.

#### 4.3 Results for ALD TMO deposition

Once these input data were used in the Monte Carlo cost model, the cost distributions shown in Figure S5 were obtained. A cost breakdown is shown in Figure 6, where each process is compared to the process(es) that they are partially or completely replacing. For the MoO₃ and TiO₂ layers, they are replacing half of the corresponding PECVD layers (the doped a-Si layers). For the AZO double-sided layers, they are replacing the entire front and rear sputtered ITO layers.

An analysis of variance was completed for each of the layers to determine the input parameters to the cost model that contributed most to this uncertainty, with the results shown in Figure 7. For the hole (MoO₃) layer (Figure 6A), the cost is dominated by materials, with a very large uncertainty. The median of the cost estimate is 1.6 c/cell, 10th percentile 0.7 c/cell and 90th percentile 5 c/cell.
cell (Figure S5a). The cost of the doped a-Si:H layer this replaces is 2.1 c/W, so there is no guarantee that the ALD MoO, layer will reduce the cost in $/cell. The large uncertainty is primarily due to not knowing the cost of purchasing very large quantities of the precursor and is also impacted by uncertainty in the precursor utilization rate. The impact of these two parameters on the layer cost can be seen graphically in Figure S5d, where every iteration of the Monte Carlo analysis is plotted against these two parameters, and the total cost of the layer is shown in the colour scale. A fit to these data is shown in Figure 7A. From these figures, we can see that precursor costs of around $20/g (compared to $75/g in lab quantities) could lead to layer total costs between 2.4 and 3.2 c/cell, depending on the precursor usage per wafer.

For the electron (TiO) layer (Figure 6B), the cost and uncertainty are much lower than for MoO, with the 10th, 50th and 90th percentile cost 1.0, 1.4 and 2.9 c/cell, respectively (Figure S5b). The materials cost is similar to the equipment depreciation; however, there is more uncertainty contributed from materials costs. In this case, the precursor cost uncertainty is the main source of variance, followed by the throughput uncertainty of the ALD equipment. The relationship between these two variables and the layer cost are shown in Figure 7B (with the Monte Carlo points shown in Figure S5e).

For both the MoO, and TiO, layers, the depreciation and maintenance costs are remarkably low compared to the PECVD process they are partially replacing. The reason for this is the relative capital cost and throughput of the respective equipment. For MoO, for example, the equipment cost of around US$ 1.8 M, and a throughput of nearly 20k wafer/h means such a tool can process more than 800 MW/year, a capital cost of approx. US$0.0023 per W of annual capacity (US$/W). In comparison, the PECVD data in the NREL database have a toolset capital cost of approximately US$0.040/Wcap. The depreciation value per produced watt can be calculated by dividing over the depreciation period (5 years). Maintenance labour costs are relatively low due to the high up-time (98%), and maintenance spare parts costs are very low based on advice from the manufacturer.

For the AZO (Figure 6C), the median total cost of 7.6 c/cell is attractive compared to the cost of the front and rear ITO layers (total cost 10.4 c/cell). However, there is significant uncertainty in both the materials and depreciation costs (Figure S5c), and the 90th percentile value of the cost estimate (11.1 c/cell) is more expensive than the ITO layers. In comparison to the hole and electron layers, AZO layer is much more expensive. This is because of the double-sided deposition of a much thicker layer (75 nm). This requires many more ALD cycles, which reduces the equipment throughput and therefore increases the equipment depreciation cost. The usage of precursors is also much higher because of the thicker layer, but this is somewhat mitigated by the comparatively low cost of the zinc precursor that makes up most of the layer. It is uncertainty in the cost of the zinc precursor that is most critical, followed by the uncertainty of the equipment throughput. The relationship between these variables and the layer cost are shown in Figure 7C (Monte Carlo iterations shown in Figure S5f).

### 4.4 | Cost comparisons of solar cell sequences

We now consider the impact of incorporating these TMO layers into different solar cell fabrication sequences. We define sequences as shown in Table S5, and images of each are shown in Figure 5.

The cost of each of these sequences is shown as a step-by-step breakdown in Figure S6. The cost of standard HJT processes were taken from the NREL data but converted from $/W to $/cell to allow the calculation of cost independent of efficiency. One important factor in this analysis is the cost of the a-Si deposition and how much this will reduce if the doped a-Si layer is not required. The NREL cost data combine the cost of the intrinsic and doped a-Si double layer into one process, since these layers are deposited sequentially within the same tool without breaking vacuum. However, for the analysis of the ALD TMO processes, we need to deposit the intrinsic a-Si layer only. Since the thickness of the intrinsic and doped a-Si layers are similar, and have similar deposition rates, avoiding the doped layer deposition will roughly halve the processing cost. However, since wafer loading and unloading times will not change, we assume that only 40% of the cost is saved.

#### 4.4.1 | Sequence B—HJT with ALD molybdenum oxide

As seen comparing Figure S6b to Figure S6a, the cost of the MoO, layer could be lower than the cost of the p-type a-Si layer, thus providing a cost benefit in $/cell. This potential cost advantage needs to be balanced against any possible solar cell performance difference, which can be done by examining the Monte Carlo uncertainty model data, combined with an exploration of the impact of an efficiency

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**FIGURE 7** Key cost drivers and their impact on total layer deposition cost for ALD deposition of (A) MoO, (B) TiO, and (C) AZO. These are fits to the Monte Carlo data shown in Figure S5d-f.
deficit or improvement. Figure 8 shows the conditions where the MoO$_x$ sequence has improved (blue) or worse (red) cost performance in solar cell $$/W. If the projected cost of the MoO$_x$ layer is at the upper end of our estimate (5 c/cell), and if there was no efficiency difference to HJT, then the MoO$_x$ sequence would be approximately 0.5 c/W more expensive. However, if the cost of MoO$_x$ were lower, at approximately 1 c/cell, then at equal efficiency, the cost would be around 0.2 c/W cheaper. The zero-contour line indicates the break-even region of cost and efficiency and can be used to identify a target efficiency difference between this structure and standard HJT. For example, even if the layer can be deposited at the lower cost of 1 c/cell, the efficiency of this structure can be at worst 0.2%$_{\text{abs}}$ lower than standard HJT.

As shown in Figure 2, the best lab demonstration of Seq B has an efficiency of 23.5%, which in that experiment was a similar value to that obtained with the standard HJT control solar cells. Although very promising, when compared to the best lab non-IBC HJT efficiency of 26.3%, this is still 2.8%$_{\text{abs}}$ lower, a deficit that is too large for this sequence to be cost-competitive. Further work is needed to improve the best lab efficiency beyond 26%, which is not impossible since the fundamental efficiency limit imposed by MoO$_x$ is 26.9%, and since it also offers the advantage of significantly lower parasitic absorption when applied to the front of the solar cell.

Even if the efficiency of this MoO$_x$ structure can match standard HJT cells, it is necessary for the cost of processing the MoO$_x$ layer to be less than 2 c/cell in order to obtain an advantage in $$/W. This presents an important area of research that should be carried out in parallel to efficiency improvements. From Figure 7A, we see that 2 c/cell requires a Mo precursor cost of between 10 and 15 $/g, depending on the precursor usage per wafer. Researchers should therefore seek to optimize and demonstrate high utilization of precursors. It is also important to investigate the cost of this MoO$_x$ precursor at manufacturing volumes. If there are indications that this precursor will be too expensive, alternative Mo precursors will need to be identified and tested for their performance.

In review, the cost analysis indicates that further research is needed to improve efficiency towards its full potential in order to realise the cost benefits of MoO$_x$.

4.4.2 | Sequence C—HJT with ALD titanium dioxide

In a similar way, comparing Figure S6c to Figure S6a, the median cost of the TiO$_x$ layer is lower than the cost of the n-type a-Si layer. The trade-off in cost and efficiency is shown in Figure 9. If we consider the best case from a cost perspective—if the TiO$_x$ process is around $0.01/cell—then the efficiency of this sequence needs to be at worst 0.2%$_{\text{abs}}$ lower than that of standard HJT to be cost competitive.

Currently this sequence, a TiO$_x$/i-a-Si:H stack in combination with ITO as electron contact, is yet to be demonstrated experimentally, but a similar structure using SiO$_2$ as the passivation layer together with TiO$_x$ as the electron selective contact has achieved 22.1%. This is 4.2%$_{\text{abs}}$ lower than the best non-IBC HJT solar cell, so for this technology to have a chance of being competitive, it is necessary to match and then exceed this efficiency significantly. The fundamental efficiency limit imposed by a passivating contact with TiO$_x$ is well over 27%, indicating that efficiency parity with standard HJT solar cells is feasible with additional development resources.

If this efficiency gap can be bridged in the lab, then the cost uncertainty and its impact on economic competitiveness is relevant. Cost related research should focus on precursor costs, and also ensure high throughput by keeping the ALD cycle time short. Another pathway to keeping the cost of this layer low is to demonstrate high efficiency with the use of a thinner TiO$_x$ layer—such as the 1.5 nm used by Bullock et al instead of the assumed 3 nm here.
4.4.3 | Seq D—HJT with ALD aluminium-doped zinc oxide

Similarly, comparing Figure S6d to Figure S6a, the cost of the AZO layers is expected to be comparable to the cost of depositing one of the ITO layers. Since the AZO cost includes depositing both sides simultaneously, the total cost of both front and back ITO is compared with AZO. The trade-off in cost and efficiency is seen in Figure 10. Depending on the eventual cost of the AZO process, the new process could be competitive even if it results in lower performance. For example, if the AZO layer costs 8 c/cell (near the median of the cost estimate), the structure could be 0.4% lower efficiency and still achieve cost parity. If the AZO layer is 6 c/cell (the lower end of the estimate), and the efficiency could be the same as standard HJT, this would save approx 0.7 c/W.

Considering the potential efficiency of this sequence, as outlined in Section 3.1, replacing ITO with ALD AZO can result in efficiency parity to standard HJT solar cells. At efficiency parity, the AZO replacement of ITO is cost effective as long as the AZO cost is less than 10 c/cell.

Another motivation for AZO as a replacement for ITO is that indium is an expensive metal with limited supply that has had increasing demand from products such as touch screens and flat panel displays. If solar cell technologies such as HJT (that use ITO layers) increase in popularity, this will increase demand pressures. It is quite possible that supply and demand dynamics lead to a significant increase in indium prices in the future. Based on the cost data from NREL (Figure S4a,b), 50% of the ITO process cost is related to the materials, mostly comprising the ITO sputter target. As an indication of possible cost fluctuations, if ITO target costs were exactly proportional to the indium market price, then a 20% indium price increase would result in a 10% increase in the cost of the ITO process.

To explore the sensitivity of this result to ITO cost, we adapted the model to include a variation of ITO layer costs, as shown in Figure 11. If the total ITO layer cost was to increase 25% (ITO cost ratio 1.25, right), the cost benefit of AZO becomes even stronger. Conversely, if ITO deposition costs were to reduce by 25% (the left figure), AZO would still be cost competitive if it can have efficiency parity and the layer cost is less than 7.7 c/cell.

From this analysis, we see that ALD AZO has great potential as an ITO replacement, as it is likely lower cost than the ITO alternative and does not face the same cost pressure from indium shortages. This motivates further work to demonstrate it can achieve efficiency parity to ITO in an industrial setting.

4.4.4 | Seq E—Three ALD layers combined

Finally, we consider the possibility of combining all three of the proposed TMO layers into one structure. By using MoO\textsubscript{x}, TiO\textsubscript{x} and AZO together, there is the potential to reduce costs overall if a high efficiency can be achieved, as well as reduce sensitivity to indium prices. This comparison can be seen in Figure 12, which indicates that if the three ALD TMO processes are at the median value of their cost range, the sequence could be cost-competitive even with a 0.5%\textsubscript{abs} efficiency deficit compared to standard HJT.

A solar cell that incorporates all three ALD TMO layers has not yet been experimentally demonstrated; however, as described in Section 3.1, the best laboratory efficiency that incorporates MoO\textsubscript{x} (thermally evaporated, not ALD) and TiO\textsubscript{x} (ALD) is 20.7%,\textsuperscript{17} 5.6%\textsubscript{abs} lower than the best non-IBC HJT solar cell. The analysis of Schmidt et al\textsuperscript{4} calculates an efficiency limit of 26%, and any improvements developed for MoO\textsubscript{x}-only or TiO\textsubscript{x}-only sequences could potentially be combined into this more complex structure. The 20.7% result used...
ITO, not AZO, however as described in Section 3.1, many experiments are showing that ALD AZO can give efficiency parity to ITO, so a combination of all three ALD TMO layers has the potential to achieve similar efficiencies to standard HJT.

5 | CONCLUSIONS

We have built a cost model to help understand the cost drivers of TMO layers formed by ALD and assessed the cost and efficiency targets required for three alternative use cases in passivated contact solar cells. We found that these ALD deposited TMO layers have the potential to be lower cost (per cell) than the doped a-Si and ITO currently used in HJT solar cells. The key factors that determine the cost are slightly different for each ALD layer, but in all cases, the costs of the precursors at industrial volumes are the most important.

While lower production costs per cell may be achievable, this will not result in lower production costs in $/W unless the resultant solar cell efficiency is high enough. The combined impact of solar cell efficiency and TMO layer costs on the relative cost in $/W between the TMO sequences and standard HJT is shown for each of the alternatives considered. In exploring these relationships, we identify efficiency targets that must be met, even if the ALD TMO layers are at the lower end of our cost estimates. Efficiency limit analysis indicates that with further development, the proposed solar cell architectures using ALD TMO layers could reach efficiency parity with standard HJT. If this was achieved, then the architectures using AZO would be competitive in $/W cost, even if the TMO layer costs are on the higher side of expectations. For the MoO	extsubscript{3} and TiO	extsubscript{2} architectures, achieving efficiency parity alone may not be sufficient, since high Mo and Ti precursor costs would lead to a more expensive process. This motivated investigations into alternate or lower purity precursors and firmer volume pricing. The use of AZO as a replacement TCO for ITO has particular merit if indium costs were to increase due to increased demand from the solar and other industries. In this work, we provide motivation and targets for continued work to demonstrate high solar cell efficiency using TMO layers in parallel to clarifying industrial throughput and high-volume precursor costs.

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CONFLICTS OF INTEREST

There are no conflicts to declare.

DATA AVAILABILITY STATEMENT

The data that supports the findings of this study are available in the supplementary material of this article.

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