

ARENA
FINAL
REPORT

**Advanced High-Efficiency Silicon Solar Cells Employing
Innovative Atomic Scale Engineered Surface
and Contact Passivation Layers**

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Solar Industrial Research Facility (SIRF) located at the University of New South Wales.

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Executive Summary

The objective of this project was to develop atomic-scale thin films that can be used as part of a passivating contact or surface passivation film. We developed innovative materials for both passivating electron and hole contacts enabled by doping of promising binary transition metal oxides. Aluminium doped titanium shows a significantly higher level of surface passivation for minority carriers as well as a considerably lower contact resistance for majority carriers. Zinc doping of nickel oxide can alleviate its poor contact performance on crystalline silicon and in combination with an amorphous silicon interface passivation layer, we demonstrated one of the most promising passivating hole contacts to date. In both cases, we demonstrated that doping improves the thermal stability of the contact. We also showed that the contact resistance between screen printed silver and a phosphorous diffused emitter is significantly reduced by capping the silicon nitride antireflection coating with a thin aluminium oxide film. This process is not only demonstrated on the lab scale but is successfully transferred to high-volume manufacturing in collaboration with our industry partner Leadmicro.

As part of the project, we also designed and installed a pilot-scale atomic layer deposition (ALD) reactor at the Solar Industrial Research Facility (SIRF) at UNSW's Kensington campus. We transferred several processes from the lab onto the pilot-scale reactor and this will enable us to conduct higher technological readiness level research in this field in the future. We also did a detailed techno-economic analysis of various ALD thin films and demonstrated that they can be significantly cheaper than their counterparts and will result in a lower levelized cost of electricity as long as a similar device performance can be obtained.

As in any research project, we also had some unexpected positive findings in this project which will be further investigated in future projects. This includes technology to mitigate potential induced degradation at the solar cell level, an enhanced carrier injection from interface passivation layers coated with high-bandgap transition metal oxides, and the first demonstration of the direct growth of graphene passivating contact.

Overall, the project has met all its milestones, was able to transfer one process to high volume production and has provided promising unexpected results that warrant further investigation.

1 Project Overview

1.1 Project summary

Silicon solar cells are dominating the photovoltaic market, and this dominance is expected to continue in the next decade(s). The most efficient way to achieve further cost reduction for silicon solar cells is to increase the solar cell efficiency. This project brought together the academic Australian leaders in the field of atomic layer deposition (ALD) and novel passivated contacts with a leading equipment manufacturer ensuring fast transfer of project results to the industry. The first part of this project focussed on exploiting the benefits of bifacial ALD on improved majority carrier transport, which simultaneously results in a significant cost reduction and increase in solar cell efficiency. The second part of this project focuses on exploiting the intrinsic merits of ALD for developing novel tunnel oxide layers and extreme workfunction materials that will allow the realisation of innovative dopant free passivated contacts. The final part of the project focused on developing bifacial transparent conductive oxide deposition using ALD, again offering significant cost benefits.

1.2 Project scope

Solar cells provide an effective source of clean renewable electricity generation. To be commercially viable, it is necessary to develop new solutions to lower the \$/W of solar cells and thus create a viable source of clean electricity for the future. The technology roadmap for photovoltaics written by a consortium of academic and industry experts forecasts the complex technological challenges the photovoltaic research community faces in the mid and long term [1]. The cost of solar cells has significantly been reduced in recent years and now the majority of the costs are associated with factors that scale with the area of a solar system, such as the PV module, mounting system, installation, permits, etc. For this reason, the focus is shifting from cost reduction to increasing solar cell efficiency, even if this increases the manufacturing cost of these solar cells.

It is clear that to improve the efficiency further, it is necessary to reduce the electrical losses at the surfaces and specifically the contact between the semiconductor and the metallisation. Current technology has pushed this to its limit and new disruptive solutions are required. As a result carrier selective or passivating contacts have recently attracted significant interest in the photovoltaic scientific community [2]. These passivating or carrier-selective contacts *by design* exhibit extremely low recombination currents for minority carriers (*i.e.* passivation) while having a low resistivity for majority carriers (*i.e.*, carrier selectivity). An elegant way to look at this was recently proposed by Brendel *et al.* [3] based on the theoretical framework proposed by Wuerfel *et al.* [4] which attributes carrier selectivity to the asymmetry in the electron and hole conduction. Brendel *et al.* defined the electron conductance ρ_n and hole conductance ρ_p as follows (in this case for a hole-selective contact):

$$\rho_n = \left(\frac{dJ_n}{dV}\right)^{-1} = \frac{v_{th}}{f_c j_c}, \quad (1)$$

$$\rho_p = \left(\frac{dJ_p}{dV}\right)^{-1} = \frac{\rho_c}{f_c}. \quad (2)$$

Where $J_{n,p}$ is the electron/hole current, V is the voltage, v_{th} is the thermal voltage, f_c is the contact fraction, and j_c is the minority carrier combination current. When dividing the electron and hole conductance we can define the selectivity S_{10} :

$$S_{10} = \log_{10} \left(\frac{\rho_n}{\rho_p}\right) = \log_{10} \left(\frac{v_{th} j_c}{j_c \rho_c}\right), \quad (3)$$

where \log_{10} is the logarithm with base 10. This selectivity is a very useful parameter to consider as it is an excellent figure of merit to estimate the efficiency potential of a certain contact. This is shown schematically in Figure 1 where we plot the upper limit efficiency as a function of the j_c and ρ_c of a contact. You can move along the iso-selectivity lines (dotted lines in Figure 1) by changing the contact fraction. This is, for example, already exploited in the PERC solar cell where the optimal rear contact fraction (with a selectivity of 13 [3]) is around 1% thereby limiting the solar cell efficiency to 27.2%. Unfortunately, the selectivity of the electron contact in a PERC cell is ~ 12 [3]. This limits the PERC solar cell to roughly 25%, in very good agreement with champion results achieved by UNSW in 1999 [5].

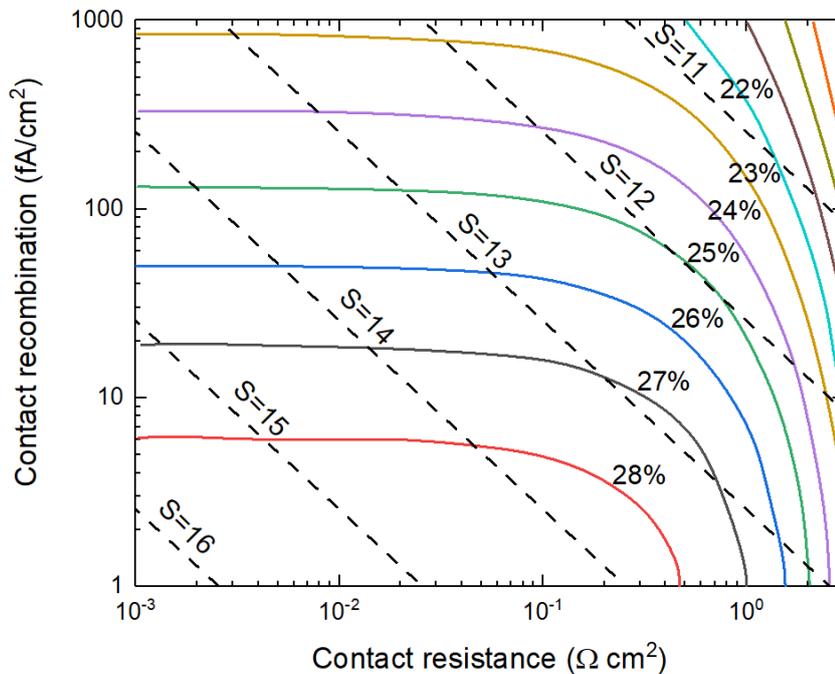


Figure 1: (Left) Upper limit efficiency of a silicon solar cell as a function of the minority carrier recombination current j_c and majority carrier contact resistance ρ_c . The dashed lines indicate iso-selectivity lines. Please note that according to the definitions in Eq. 1-3, we can move along the iso-selectivity line by changing the contact fraction f_c . Hence, this allows for optimising the efficiency ceiling for a certain contact system when the j_c and ρ_c values are known. This calculation was adapted from Brendel et al. and assumes a short-circuit density of 43.6 mA/cm^2 and assumes that the solar cell efficiency is limited by only one contact.

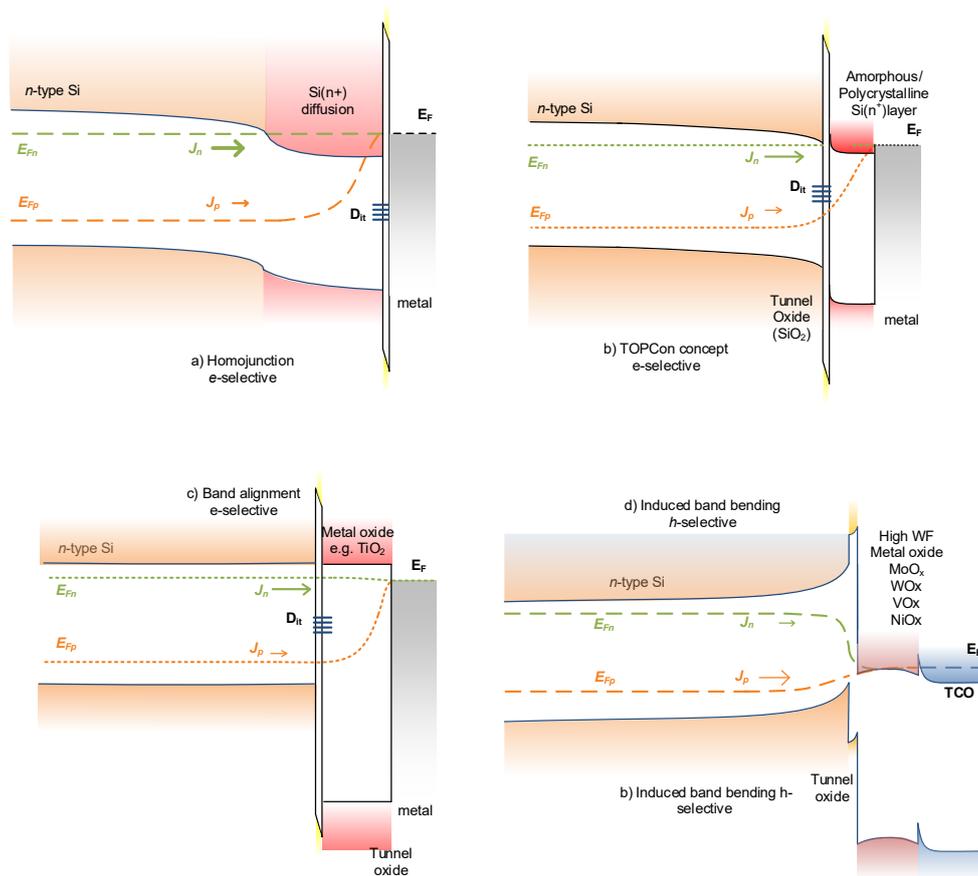


Figure 2 Types of carrier selective contacts. Graph is adapted from Macco et al. [6]

In Figure 2, we show various ways to fabricate a carrier-selective contact. The first type shown in Fig. 2(a) is the well-known diffused electron collector (typically referred to as emitter). This system is currently dominating the photovoltaic industry and has a reasonable selectivity of 12 for electrons and 13 for holes [3]. The types shown in Figure 2(b) – (d) are typically referred to as passivating contact and these contact systems are the main focus of this project. It can be seen that these contact systems typically consist of a multilayer structure of a layer with a high level of surface passivation such as silicon oxide and amorphous silicon in combination with a material that results in the selectivity of the contact. This passivation layer predominantly aims to reduce j_c of the contact as this is typically unacceptably high due to a high density of interface states at the c-Si surface. The second layer in the structure yields the carrier selectivity, and this can be grouped in three different categories. As shown in Figure 2(b), the first category of materials has a high asymmetry in electron/hole conduction. Most notable examples are doped amorphous silicon and doped poly-silicon, which are, e.g., applied in heterojunction and TOPCon solar cells, respectively. The second category shown in Figure 2(c) relies on band alignment with either the valence band (for a hole contact) or with the conduction band (for an electron contact). The band offset for the other band should be as high as possible to ensure minimal conductance for the other charge carrier. The third and last category shown in Figure 2(d), relies on extreme work function materials which result in a band-bending in the underlying silicon. This band bending results in a shift of the quasi-Fermi levels towards the valence band (for hole contacts) or conduction band (for electron contacts) and this gives the contact a high selectivity. In Figure 3, we show the material which will be investigated in this project.

The efficiency potential of passivating contacts have already been demonstrated at the solar cell device level and they have resulted in a string of world record silicon wafer solar cell efficiencies recently such as the 26.7% efficient all-back-contact heterojunction with intrinsic thin film solar cell from Panasonic [7] and the 25.7% efficient TOPCon (tunnel oxide passivated contact) solar cell from Fraunhofer ISE [8]. Both have now overtaken the landmark 25% efficiency result set by UNSW in 2008 after recalibrating a measurement result from 1999 after the change in the AM1.5G spectrum [5].

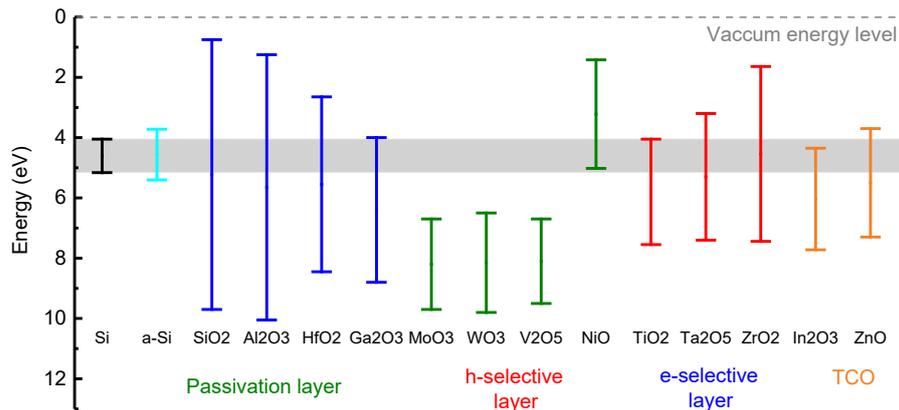


Figure 3: Schematic illustration of the band alignment with Si of various metal oxides that will be investigated in this project.

Most research groups use thermal evaporation or low-pressure chemical vapour deposition [LPCVD] for the synthesis of carrier selective or passivated contacts. These synthesis methods do not allow for control of the material properties at the atomic scale, a level of control that is desirable to achieve the maximum potential of these new contacts. Often insufficient control inhibits the take up of these novel contacts by industry. In this project, we exploited the unique capabilities of atomic layer deposition (ALD) to synthesise innovative electron and hole contacts that exhibit a very high carrier selectivity. ALD is a chemical vapour deposition (CVD) technique in which the deposition process involves self-limiting (half) reactions. It allows for conformal deposition of films over large areas with “digital” thickness control. The resulting films are pinhole free, have low defect density & low stress, and have excellent adhesion to the underlying substrate due to strong chemical bonding. ALD offers unprecedented control of the material composition as the growth precursors that are used during the self-limiting reactions can be easily changed. This allows the straightforward synthesis of multilayer structures that can be tailored at the atomic scale towards desired material properties such as work function, phase, density and type of defects, etc. It is, for example, very straightforward to grow a layer stack that consists of a layer with a high level of surface passivation at the interface such as aluminium oxide (Al₂O₃) [9] capped by a layer such as molybdenum oxide (MoO_x) that has already shown great potential as hole-selective contact for silicon solar cells [10]. This will enable functionalities that were not feasible previously, e.g., in terms of workfunction, fixed charge density, and carrier conductivity.

1.3 Outcomes

The project has met all the milestones set at the beginning of the project with some additional unexpected positive results that will be explored further in future projects. The project made significant progress in the development of passivation contacts.

We demonstrated that the most promising electron selective contact titanium oxide could significantly be improved by doping the layer with aluminium. Aluminium doping improved both the contact properties as well as the level of surface passivation, thus resulting in an improved selectivity according to Equation 3. In addition, the layer had an improved thermal stability which is desirable from an industrial point of view. We also made significant progress with passivating hole contacts where we found that we could improve the performance of nickel oxide by doping the film with zinc. The addition of zinc significantly improved the contact between nickel oxide and silicon. In addition, the nickel oxide layer was also more thermally stable. After adding a hydrogenated amorphous silicon interface passivation layer, we were able to achieve a contact with a selectivity of 15, which is among the most promising passivating hole contacts developed to date.

We also conducted a techno-economic analysis of the processes developed in this work and demonstrated that these processes typically have significantly lower costs than their peers and will be appealing options as long as they reach efficiency parity.

In terms of unexpected outcomes, we identified that extreme workfunction materials do not only offer benefits due to their low absorption but that they can also enhance the performance of the underlying interface passivation layer. We showed a significantly higher carrier injection from hydrogenated amorphous silicon capped by a molybdenum oxide layer compared to boron-doped hydrogenated amorphous silicon. This further increases the efficiency ceiling of this contact and its appeal to the photovoltaic industry. We also showed that ALD layers can effectively protect solar cells against potential induced degradation. We were also able to synthesise graphene directly on ALD nickel oxide, solving a critical technology barrier for the application of graphene films by using a functional layer as the catalyst for the graphene growth.

1.4 Transferability

The project was intrinsically focussed on the rapid transfer of results from the lab to the factory. For this purpose, our industry partner Leadmicro donated a pilot-scale ALD reactor to UNSW, that was installed at the Solar Industrial Research Facility at UNSW's campus in Kensington as shown in Figure 4. This pilot-scale ALD system can process 200 samples simultaneously and has room for 4 different precursors. The process chamber is identical to the full production tool apart from its length, ensuring that processes developed at UNSW can quickly be transferred to the industry. During the project we have developed processes for thermal and ozone assisted aluminum oxide, intrinsic titanium oxide, and aluminum doped titanium oxide. Figure 5 shows that often quite some work is required to transfer a new process onto a pilot-scale tool, in this case taking 15 deposition runs to dial in a good process for titanium oxide.



Figure 4: Picture of the Leadmicro ALD tool in UNSW's Solar Industrial Research Facility and operated by PhD student Mr Xinyuan Wu.

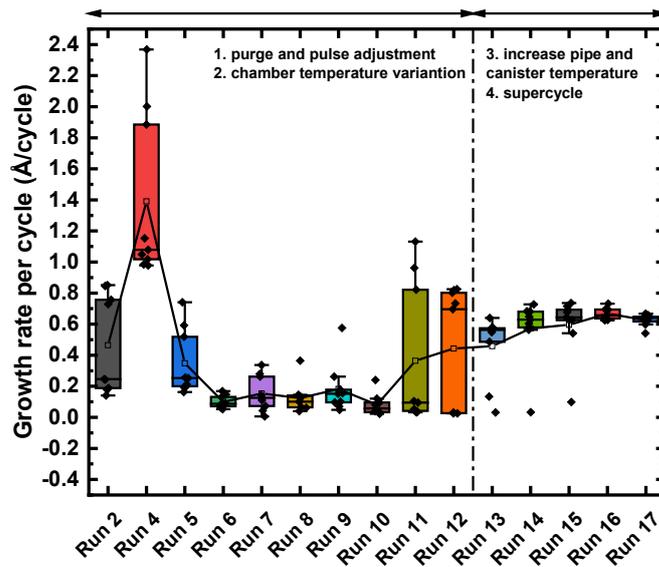


Figure 5: Growth rate per cycle of the titanium oxide process as a function of the experimental runs.

During the project, we were also already able to test a few processes at pilot-production lines of Tier-1 solar cell manufacturers. In Figure 6 we show the result of one such high-volume test where we tested the application of aluminium oxide on both sides of the solar cell. The application of aluminium oxide on the rear side of the PERC solar cell is now the industry standard [11]. However, the application of an additional aluminum oxide on the front side of the solar cell was proposed by our group to reduce the contact resistance of screen-printed silver contacts on phosphorous diffused emitters [12]. As ALD is capable of depositing films uniformly on both sides of a solar cell, we could grow the aluminium oxide layer on both sides of the solar cell in one single process step and we demonstrated that this process gives ALD a further edge over plasma-enhanced chemical vapour deposited (PECVD) aluminum oxide [13].

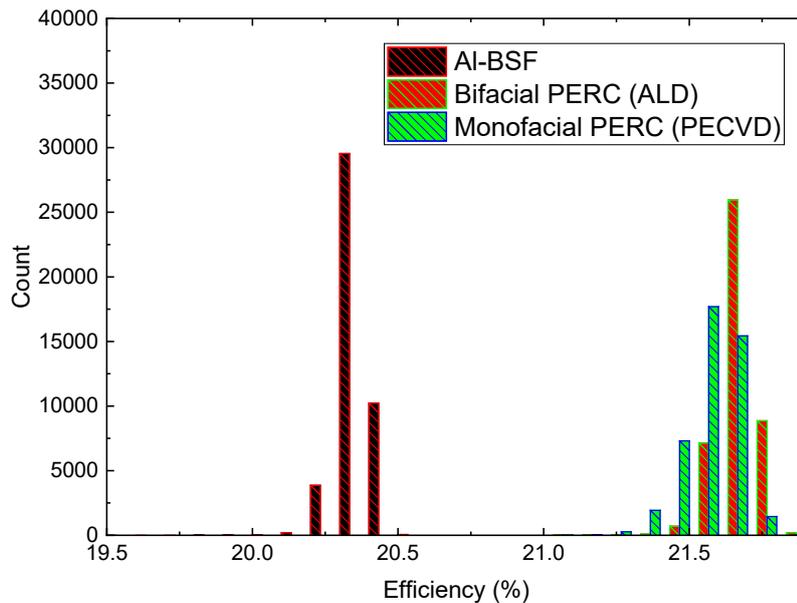


Figure 6: Solar cell efficiency data for Al-BSF solar cells, PERC solar cells with a monofacial aluminium oxide layer on the rear surface, and PERC solar cells with a bifacial ALD layer.

1.5 Conclusion and next steps

Overall, we are very pleased with the outcomes of this project. All milestones were met, one process has already been commercialised, and a second process is currently being trailed with an industry partner. The project has also resulted in some unexpected results that will, hopefully, be investigated further in future projects. A lasting legacy of the project will also be the pilot-scale ALD reactor at UNSW, making it one of the best-equipped universities in this fascinating research area.

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Lessons Learnt

Lessons Learnt Report: Doping of transition metal oxides can significantly improve their performance

Project Name: *Advanced high-efficiency silicon solar cells employing innovative atomic scale engineered surface*

Knowledge Category:	Technical
Knowledge Type:	Technology
Technology Type:	Solar PV
State/Territory:	NSW

Key learning

The intrinsic properties of transition metal oxides are often not ideal for being used in passivating contacts. In this project, we have shown that doping the transition metal oxide with the correct material can significantly improve their properties such as conductivity, thermal stability, and level of crystalline silicon surface passivation. We found that density functional theory (DFT) is a great tool to predict which dopants will provide the desired change in material properties of the transition metal oxides.

Implications for future projects

We will see an increased use of doped transition metal oxides in future solar cells and other devices as doping allows the finetuning of the desired functional properties. This will also significantly increase the parameter space for process optimisation, and there DFT can play an important role in informing the development.

Knowledge gap

Most thin film materials are grown and subsequently used in the solar cell device in the amorphous state. Amorphous materials are quite different from their crystalline counterparts and this has to be taken into account when doing DFT calculations. Typical DFT is done on crystalline structures, as performing these calculations on amorphous systems is significantly more challenging. More work needs to be done to fully understand the impact of doping on amorphous transition metal oxides.

Background

Figure 1 shows how the density of states (DOS) of nickel oxide (NiO) is changed by doping the film with various substitutional dopants. Typically doping results in the addition of defect states in the bandgap of the material and the defect position will determine if these defects provide a beneficial change to the material properties. In Figure 2 we show that Al-doping significantly improves the contact properties of NiO on c-Si. Intrinsic atomic layer deposited NiO has an

inferior contact on c-Si and this significantly improves when the NiO film is doped with Al. Annealing further improves the contact performance up to an annealing temperature of 400 °C when the contact resistance increases significantly again.

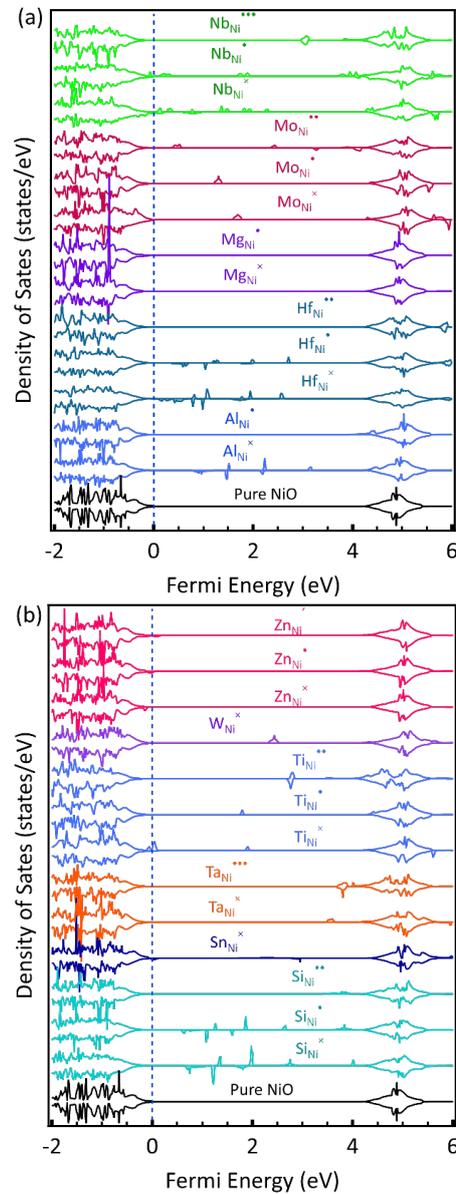


Figure 1: Spin-polarised total density of states (DOS) of the nickel oxide (NiO) with dopants in various charged states (a, b). Total DOS of a pristine NiO is also provided for comparison. As the core states are not affected by the presence of defects, all energies are aligned to the 0 (s) core states of the pristine NiO structure (figure taken from M. A. Hossain et al., "Doped Nickel Oxide Carrier-Selective Contact for Silicon Solar Cells," *IEEE Journal of Photovoltaics*, vol. 11, no. 5, pp. 1176-1187, 2021-09-01 2021).

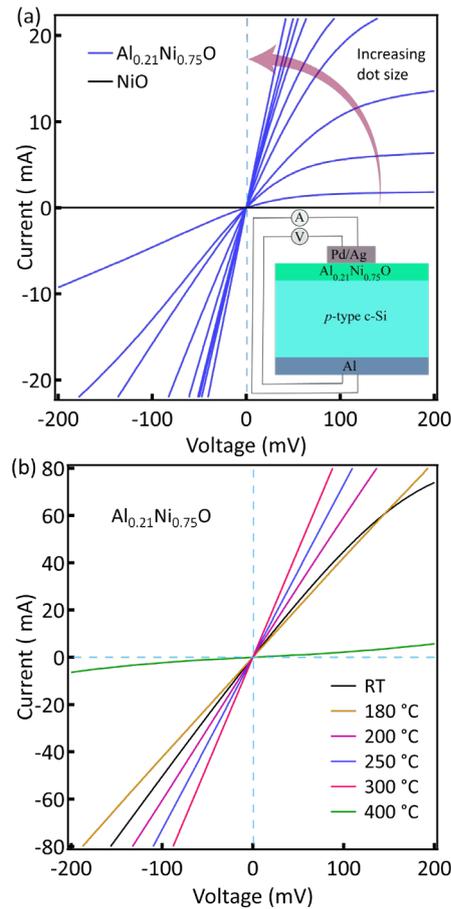


Figure 2: Dark I-V characteristics of p -type c -Si/ $\text{Al}_{0.21}\text{Ni}_{0.75}\text{O}$ contacts (solid lines) measured in the Cox and Strack structure (inset image) using (a) Pd/Ag bilayer pads of 0.1 – 1 cm diameter circles, and (b) post-deposition annealing of the stack layer with 1 cm diameter Pd/Ag bilayer pad at 180 – 400 °C, indicating a stable ohmic c -Si/ $\text{Al}_{0.21}\text{Ni}_{0.75}\text{O}$ contact, where RT is room temperature (figure taken from M. A. Hossain et al., "Doped Nickel Oxide Carrier-Selective Contact for Silicon Solar Cells," *IEEE Journal of Photovoltaics*, vol. 11, no. 5, pp. 1176-1187, 2021-09-01 2021).

Lessons Learnt

Lessons Learnt Report: Atomic layer deposition can enable low cost solar cell manufacturing

Project Name: *Advanced high-efficiency silicon solar cells employing innovative atomic scale engineered surface*

Knowledge Category:	Technical
Knowledge Type:	Technology
Technology Type:	Solar PV
State/Territory:	NSW

Key learning

In this project we have shown that atomic layer deposition (ALD) is a very appealing low cost manufacturing technology for current and future generation solar cells. The recent developments in both spatial and temporal ALD have solved the main drawbacks of ALD with respect of throughput without sacrificing its intrinsic benefits such as atomic layer composition and thickness control as well as excellent uniformity on challenging substrates. We investigated the techno-economic potential of a range of ALD thin films and compared it to their plasma-enhanced chemical vapour deposited (PECVD) and physical vapour deposited (PVD) counterparts and showed that ALD will be the lowest costs process in many circumstances as long as the learning rate of manufacturing the precursors at scale are comparable to what we have seen for trimethyl aluminum (used for aluminium oxide) and diethyl zinc (used for zinc oxide) in the past.

Implications for future projects

This key learning means that ALD can be considered for high-volume manufacturing low-cost solar cells for a wide range of thin films, even with thicknesses up to 100 nm. Process development can be done on single wafer lab scale reactors and subsequently the process can rapidly be transferred to a high-volume reactor.

Knowledge gap

Chemical vapour deposition (CVD) processes like ALD are strongly dependent on the chemical nature of the underlying film. This typically results in significant differences between initial growth and steady state growth. For devices with functional nanoscale thin films, this initial growth can have a significant impact on the device performance and thus should be carefully considered when optimising devices.

Background

We performed a Monte-Carlo based techno-economic analysis of a range of ALD thin films and compared them to their PECVD and PVD counterparts. In Figure 1, we show the result we obtained when we compared ALD MoO_x to p-type doped amorphous silicon. It can be seen that ALD MoO_x is significantly cheaper than p-type doped amorphous silicon in most scenarios that we simulated, as long as the solar cells with MoO_x would have a similar performance (which is currently not yet the case).

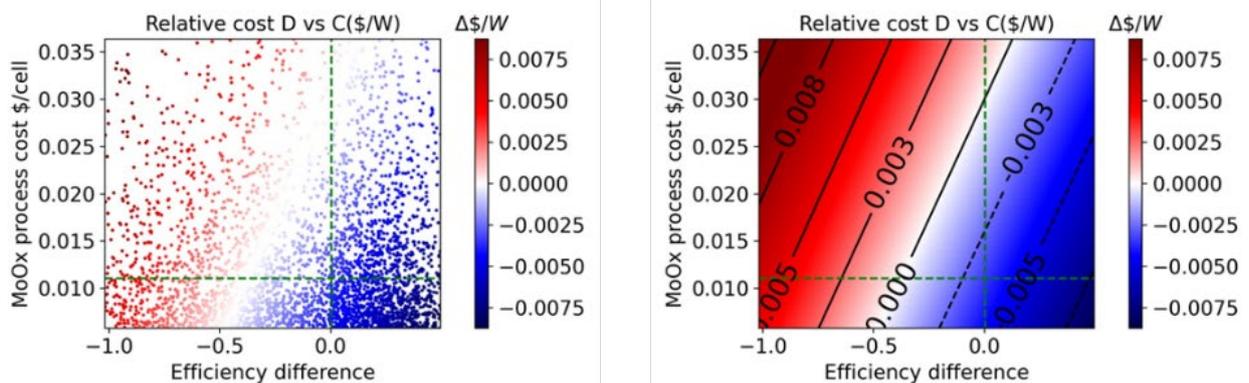


Figure 1: Comparison in $\$/W$ of using ALD MoO_x to replace p-doped a-Si. This shows that solar cells with ALD MoO_x can even be slightly less efficient while still having an edge in terms of $\$/Wp$. Figure taken from (N. L. Chang et al., "Techno-economic analysis of the use of atomic layer deposited transition metal oxides in silicon heterojunction solar cells," *Progress in Photovoltaics: Research and Applications*, 2022-03-11 2022).

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For more information and technical details on the contents above, the reader is referred to the scientific literature. Below is the complete list of papers published during this project.

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