

Surface passivation of silicon solar cells using industrially relevant Al_2O_3 deposition techniques

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ABSTRACT

The next generation of industrial silicon solar cells aims at efficiencies of 20% and above. To achieve this goal using ever-thinner silicon wafers, a highly effective surface passivation of the cell front and rear is required. In the past, finding a suitable dielectric layer providing a high-quality rear passivation has been a major challenge. Aluminium oxide (Al_2O_3) grown by atomic layer deposition (ALD) has only recently turned out to be a nearly perfect candidate for such a dielectric. However, conventional ALD is limited to deposition rates well below 2nm/min, which is incompatible with industrial solar cell production. This paper assesses the passivation quality provided by three different industrially relevant techniques for the deposition of Al_2O_3 layers, namely high-rate spatial ALD, plasma-enhanced chemical vapour deposition (PECVD) and reactive sputtering.

Introduction

In high-efficiency laboratory silicon solar cells, surface recombination is very effectively suppressed by means of silicon dioxide (SiO_2) grown in a high-temperature ($\geq 900^\circ\text{C}$) oxidation process [1]. Very low surface recombination velocities (SRVs) are in particular realized at the lightly doped rear surface, where the combination of a thermally grown SiO_2 layer with an evaporated film of Al gives – after an additional annealing treatment at $\sim 400^\circ\text{C}$ (the so-called ‘alneal’) – SRVs below 20cm/s on low-resistivity ($\sim 1\Omega\text{cm}$) p-type silicon wafers [2]. In addition, the SiO_2/Al stack at the cell rear acts as

an excellent reflector for near-bandgap photons, significantly improving the light-trapping properties and, hence, the short-circuit current of the cell. One of the main reasons why high-temperature oxidation has not been implemented into the majority of industrial cell processes so far is the high sensitivity of the silicon bulk lifetime to high-temperature processes. Particularly in the case of multicrystalline silicon wafers, thermal processes above 900°C typically lead to a significant degradation of the bulk lifetime [3]. Hence, low-temperature surface passivation alternatives are required for future industrial high-efficiency silicon solar cells.

One intensively investigated low-temperature surface passivation alternative to thermal oxide is silicon nitride (SiN_x) grown by PECVD at $\sim 400^\circ\text{C}$, which has proven to give comparably low SRVs as thermal SiO_2 on low-resistivity p-type silicon [4,5]. However, when applied to the rear of PERC (passivated emitter and rear cell)-type solar cells on a p-type substrate, the short-circuit current density is strongly reduced compared to the SiO_2 -passivated cell rear [6]. This effect has been attributed to the large density of fixed positive charges within the SiN_x layer, inducing an inversion layer in the p-type silicon underneath the SiN_x . The coupling of this inversion layer to the base metal contact leads to a significant loss in the short-circuit current density and the fill factor, a detrimental effect that is known as ‘parasitic shunting’ [7].

“Aluminium oxide (Al_2O_3) has proven capable of providing an excellent level of surface passivation.”

Fortunately, the negative-charge-dielectric aluminium oxide (Al_2O_3) has proven capable of providing an excellent level of surface passivation on low-resistivity p-type and n-type silicon wafers as well as on boron- and aluminium-doped p⁺-emitters [8–21]. Al_2O_3 can be deposited by various techniques, such as ALD, PECVD and reactive sputtering. In particular, it was demonstrated that it is ideally suited to the rear passivation of

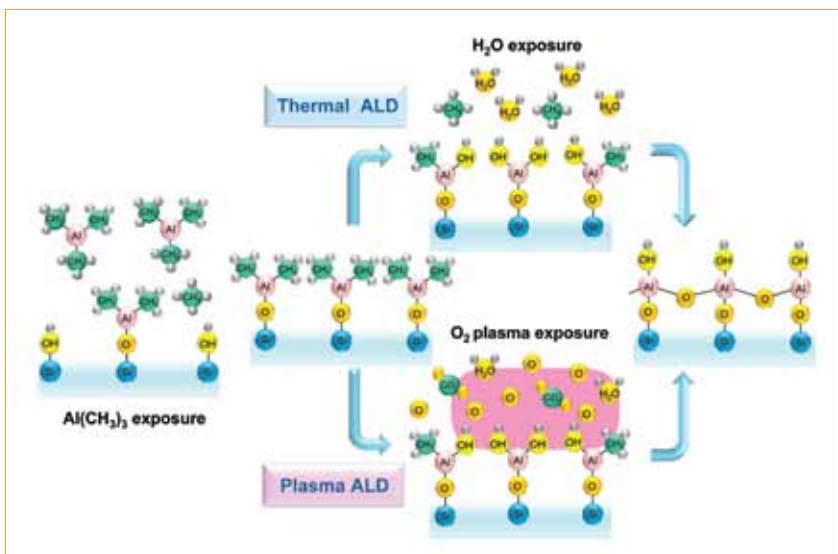


Figure 1. Schematic of one cycle of a thermal and a plasma-assisted ALD process. Each cycle consists of two half-steps: first, the trimethyl aluminium (TMA) molecules attach to the hydroxyl groups bound to the silicon surface; second, the molecules are oxidized by H_2O (thermal ALD) or an O_2 plasma (plasma ALD).

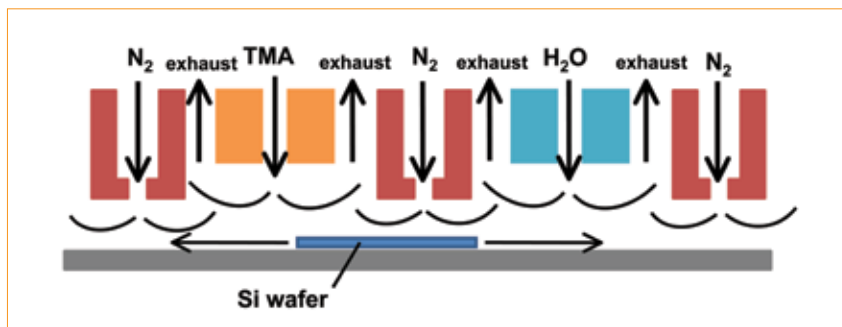


Figure 2. Schematic of the spatial ALD concept [22]. The TMA and water half-reaction zones are separated by N_2 gas bearings.

PERC solar cells, as parasitic shunting is completely absent thanks to the fixed negative charges [11]. In this contribution, we systematically compare the passivation quality of Al_2O_3 films deposited by various deposition techniques. Atomic layer deposition performed in lab reactors (plasma-assisted as well as thermal ALD) provides an outstanding surface passivation quality; however, it is limited to very low deposition rates ($<2\text{nm}/\text{min}$), making conventional, temporal ALD reactors unsuitable for industrial solar cell production. On the other hand, we will demonstrate that high-rate spatial ALD, PECVD, and reactive sputtering have an enormous potential for a transfer of Al_2O_3 into industrial cell production in the near future.

Atomic layer deposition

In the ALD process, one monolayer of Al_2O_3 is grown per cycle, with each cycle consisting of two half-reactions, as depicted in Fig. 1. In the first half-reaction, trimethyl aluminium (TMA) molecules react with hydroxyl (OH) groups attached to the surface. At the end of the first half-reaction, Al atoms and methyl groups cover the surface and the remaining TMA molecules in the deposition chamber are no longer able to react with the surface. After purging the deposition chamber with inert or oxygen gas, the second half-reaction of the ALD cycle starts. One can apply two different realization forms for the second half-reaction: in the thermal ALD process, water vapour is injected into the deposition chamber. The H_2O molecules react very fast with the Al- CH_3 complex attached to the surface. Hydrogen reacts with the methyl group to form methane, and oxygen reacts with aluminium to form aluminium oxide. In the plasma-assisted ALD ('plasma ALD') process, an oxygen plasma is ignited above the substrate, generating oxygen radicals which effectively react with the methyl groups and the aluminium at the surface. Oxford Instruments' FlexAL[™] deposition system, applied in this study, uses a remote inductively coupled plasma (ICP) source, which means that the oxygen

plasma is not in direct contact with the silicon wafer during Al_2O_3 deposition. This type of remote-plasma deposition technique is known to create almost no plasma damage at the surface, and is hence well suited for an excellent silicon surface passivation.

“High-rate spatial ALD, PECVD, and reactive sputtering have an enormous potential for a transfer of Al_2O_3 into industrial cell production.”

In a conventional ALD process, the separation of the half-reactions is implemented by alternate dosing of the process gases. Exposure times of only a few milliseconds are sufficient to ensure complete saturation of the growth surface. In between both precursor doses, however, the reactor chamber is purged

by an inert gas and subsequently pumped to remove the residual process gas and reaction products. In order to prevent parasitic CVD processes and ensure a true ALD process, pumping times of a few seconds are required, severely limiting the growth rate to approximately $2\text{nm}/\text{min}$. This renders conventional ALD unsuitable for high-throughput industrial solar cell production.

Recently, Poedt et al. proposed a high-rate ALD concept based on spatially separated ALD ('spatial ALD') [22], enabling deposition rates of $70\text{nm}/\text{min}$. In contrast to the conventional sequential separation, both half-reactions are spatially separated (see Fig. 2), thus eliminating the need for intermediate pumping steps. In a first proof-of-principle lab tool developed at TNO, the spatial separation was achieved by rotating the wafer underneath a round reactor head incorporating gas inlets for TMA and water vapour, separated by gas bearing planes formed by a flow of pressurized nitrogen. Since both reaction zones are sealed off by nitrogen flow, any unintentional interaction of the process gases is prevented and the deposition can be performed under atmospheric conditions, an additional advantage concerning the industrial applicability. The spatial ALD concept has recently led to excellent surface passivation results on p- and n-type silicon wafers [23].

Note that high-throughput (up to 3,000 wafers per hour) reactors based on the spatial ALD approach are currently under development at two different companies, namely SoLayTec and Levitech, and will be commercially available in the

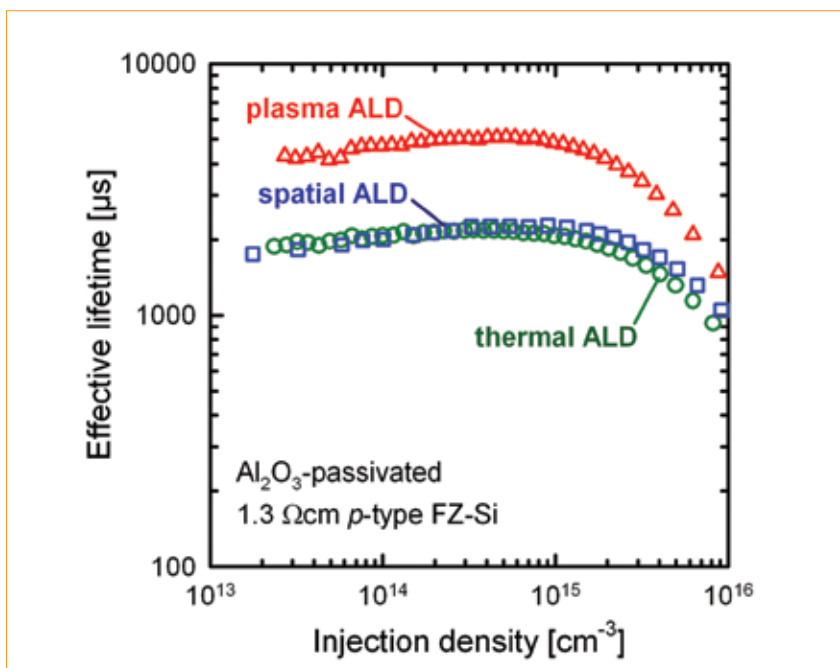


Figure 3. Effective lifetime τ_{eff} as a function of the injection density Δn measured on $1.3\ \Omega\text{cm}$ p-type FZ-Si passivated by Al_2O_3 deposited by plasma, thermal and spatial ALD.

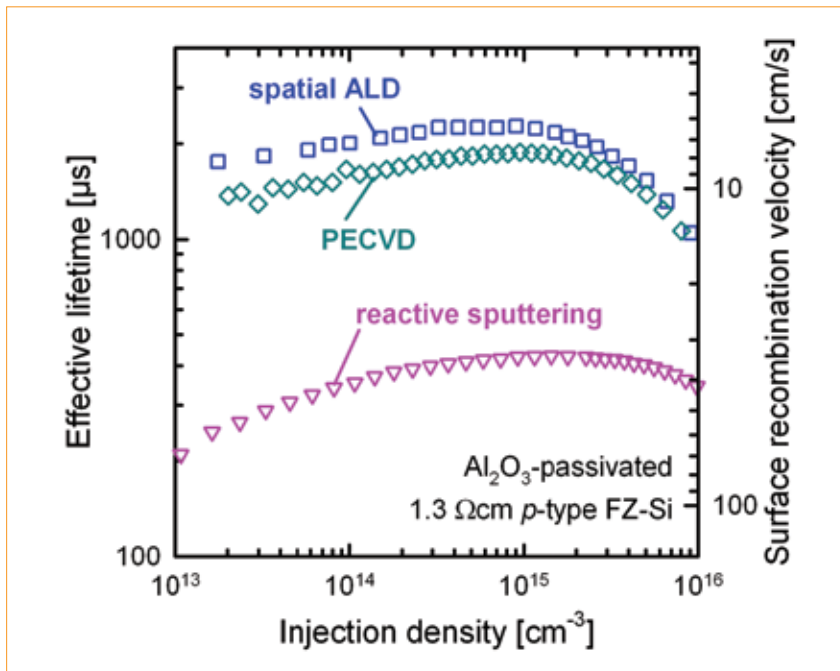


Figure 4. Effective lifetime (left scale) and corresponding surface recombination velocity (SRV) (right scale) as a function of the injection density, measured on 1.3Ωcm p-type FZ-Si passivated by Al₂O₃ deposited by spatial ALD, PECVD, and reactive sputtering.

near future with the extra option of an additional gas bearing at the wafer back side, thus enabling double-floating wafer transport in a reciprocating manner or in a single direction.

Fig. 3 shows the effective lifetimes τ_{eff} measured as a function of the injection density Δn for 1.3Ωcm p-type float-zone silicon (FZ-Si) wafers passivated using Al₂O₃ deposited by plasma-assisted, thermal and spatial ALD. Lifetimes were

measured by the photoconductance decay (PCD) method using a Sinton lifetime tester. All Al₂O₃ films received a post-deposition anneal at (400±50)°C for ~15 min to activate the surface passivation [19]. As can be seen from Fig. 3, all three ALD techniques result in Al₂O₃ films of outstanding surface passivation quality, which shows an extremely weak injection dependence over the complete relevant injection range between 10¹³

and 10¹⁵cm⁻³. Al₂O₃ deposited by plasma ALD provides effective lifetimes between 3 and 4.8ms in the relevant injection range. The measured lifetime of 4.8ms at $\Delta n = 10^{15}$ cm⁻³ lies well above the commonly used empirical expression for the intrinsic lifetime limit for crystalline silicon [24], indicating a nearly perfect surface passivation, even better than that previously achieved with 'annealed' thermally-grown SiO₂. Assuming an infinite bulk lifetime, we can calculate an upper limit to the SRV S_{max} using the simple relation $S_{\text{max}} = W/2\tau_{\text{eff}}$, where $W = 290\mu\text{m}$ is the measured wafer thickness. Using this relation, $\tau_{\text{eff}} = 4.8\text{ms}$, corresponds to an upper SRV limit of $S_{\text{max}} = 3\text{cm/s}$. Given that the measured lifetime is above the previous intrinsic limit for silicon, the lower limit is $S_{\text{min}} = 0\text{cm/s}$, and the real SRV is in between those two values – too low to be noticed by a normal solar cell. In this work, all SRVs reported are S_{max} values.

Most importantly, it can be concluded from Fig. 3 that both traditional thermal ALD as well as spatial ALD provide Al₂O₃ films with an extremely high level of surface passivation, as indicated by lifetimes of 2ms, corresponding to an upper SRV limit of $S_{\text{max}} = 7\text{cm/s}$, for both techniques and a practically negligible injection dependence over the relevant injection range. It is quite remarkable that the high-rate (in our example 14nm/min) spatial ALD produces exactly the same excellent level of surface passivation as the slow (< 2nm/min) conventional thermal ALD [23].

Spatial ALD, PECVD and sputtering

In addition to spatial ALD, two other techniques have recently been demonstrated to be suitable for depositing surface-passivating Al₂O₃ layers. PECVD [14,15,21] has been shown to provide SRVs of only 10cm/s on 1Ωcm p-type FZ-Si, whereas reactive material has resulted in SRVs down to 55cm/s. In addition to the Al₂O₃ films deposited by spatial ALD, we have examined the passivation quality of Al₂O₃ films deposited in an in-line microwave-remote PECVD (Roth&Rau, SiNA) system and in an RF magnetron sputtering lab system [19]. The sputtering uses an aluminium target, which is reactively sputtered in an O₂/Ar atmosphere, while the PECVD uses TMA and nitrous oxide as process gases.

Fig. 4 compares the effective lifetimes measured on 1.3Ωcm p-type FZ-Si wafers passivated by Al₂O₃ films deposited using, in our opinion, the most promising industrial Al₂O₃ deposition techniques: (i) spatial ALD, (ii) PECVD, and (iii) RF magnetron sputtering. The direct lifetime comparison in Fig. 4 shows that both spatial ALD and PECVD provide S_{max}

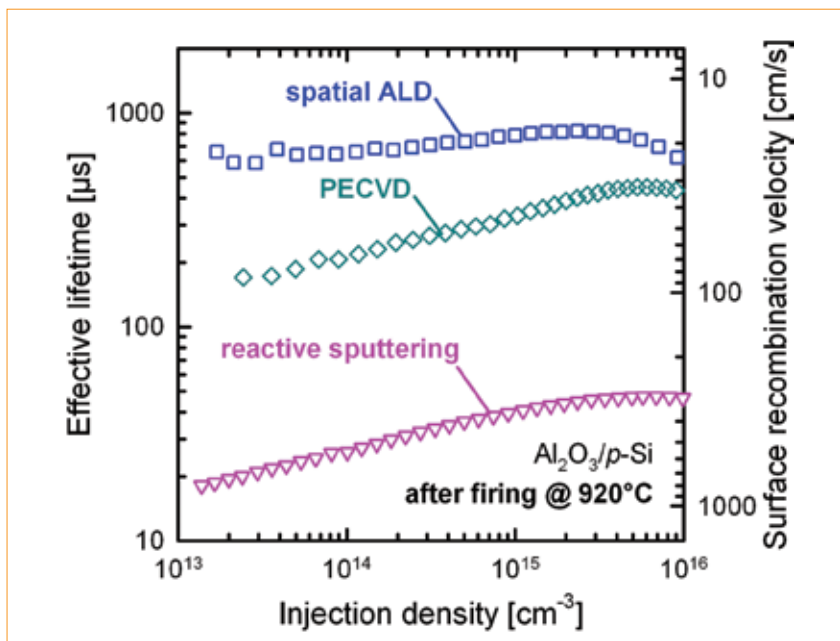


Figure 5. Injection-dependent effective lifetime (left scale) and corresponding surface recombination velocity (SRV) (right scale) after firing at a peak set temperature of 920°C (measured peak temperature ~800°C). The samples are 1.3Ωcm p-type FZ-Si wafers passivated by Al₂O₃ deposited using spatial ALD, PECVD, and reactive sputtering.

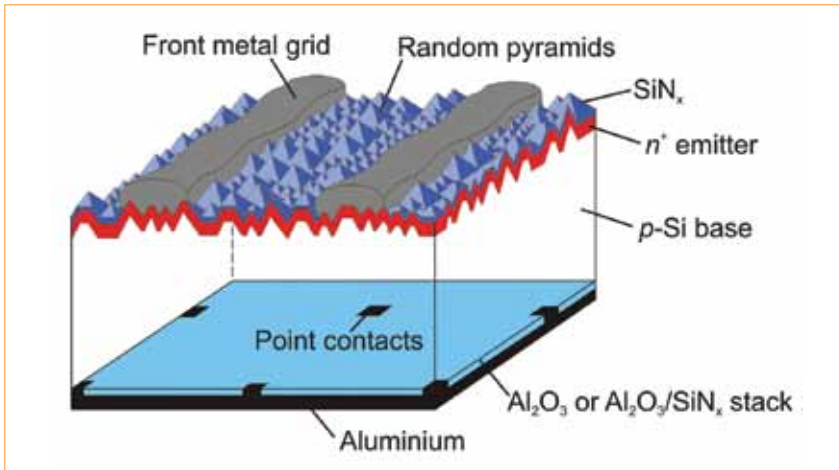


Figure 6. PERC-type solar cell structure used to demonstrate the applicability of different rear surface passivation schemes.

values $< 10\text{cm/s}$, clearly outperforming the sputtered Al_2O_3 . Nevertheless, the sputtered Al_2O_3 passivation layer results in a surface recombination velocity between 35 and 70cm/s in the relevant injection range, which would still be acceptable for the next generation of industrial high-efficiency solar cells.

“The PERC solar cells with sputtered Al_2O_3 as rear passivation achieve an independently confirmed efficiency of 20.1%.”

Another very important property is the stability of the surface passivation during a firing step as it is typically applied in the screen-printing metallization of solar cell production lines. We annealed lifetime samples in an industrial infrared conveyor-belt furnace (Centrotherm Contact Firing Furnace DO 8.600-300-FF) at a set temperature of 920°C (measured peak temperature $\sim 800^\circ\text{C}$). Fig. 5 shows the injection-dependent lifetimes and corresponding surface recombination velocities measured after firing. The Al_2O_3 deposited by spatial ALD shows clearly the best firing stability, providing SRVs of

$\sim 20\text{cm/s}$ after firing over the entire relevant injection range. The Al_2O_3 layer deposited by inline PECVD also results in a good passivation quality after firing, providing S_{max} values between 30 and 80cm/s in the injection range of relevance. The sputtered Al_2O_3 shows the strongest increase in the surface recombination after firing, leading to SRVs between 300 and 800cm/s . Obviously, the sputtered Al_2O_3 needs further optimization, while the PECVD- Al_2O_3 and in particular the spatial ALD- Al_2O_3 layers can be directly implemented in a screen-printing solar cell process.

As large-area in-line PECVD systems are already available on the market, PECVD seems to be the preferred short-term deposition technique for Al_2O_3 . The preferred medium-term and long-term deposition technique might be ultrafast spatial ALD due to its reduced TMA gas consumption compared to PECVD, the absence of parasitic deposition at the reactor wall and a smaller footprint of the deposition systems currently under development. Also, ALD provides highest-quality pinhole-free Al_2O_3 films and allows conformal film deposition, which might prove useful for advanced solar cell concepts.

PERC solar cells

We have implemented our Al_2O_3 rear passivation layers deposited by plasma ALD, thermal ALD and sputtering into

passivated emitter and rear cells using the process sequence described in [11]. Fig. 6 shows the cell structure featuring a PECVD- SiN_x -passivated $100\Omega/\text{sq}$ phosphorus-diffused n^+ front emitter and a rear surface passivated by the dielectric layer systems shown in the first column of Table 1. The front grid is made by shadow-mask evaporation of aluminium and the rear is fully metallized by aluminium evaporation ($\sim 4\%$ rear metal contact fraction) after point contact openings have been generated. Table 1 summarizes the one-sun parameters of the best PERC solar cells, as measured under standard testing conditions (25°C , $100\text{mW}/\text{cm}^2$, AM 1.5 G). The measured open-circuit voltages (V_{oc}) and short-circuit current densities (J_{sc}) of the ALD-passivated cells are clearly superior to the V_{oc} and J_{sc} values of the cell with sputtered Al_2O_3 .

V_{oc} values of ALD-passivated cells are all $> 660\text{mV}$ and J_{sc} values are $> 40\text{mA}/\text{cm}^2$, demonstrating the huge potential of ALD for the rear surface passivation of PERC-type cells. We deposited thicker PECVD- SiO_x or SiN_x layers on top of the very thin ALD- Al_2O_3 layers, mainly to improve the internal rear reflection of the cell. The independently confirmed conversion efficiencies are 21.4% for the plasma ALD- Al_2O_3 rear passivation and 20.7% for the thermal ALD- Al_2O_3 passivation. The passivation quality of the sputtered Al_2O_3 is clearly inferior to that of the ALD- Al_2O_3 films, as indicated by an $\sim 10\text{mV}$ lower V_{oc} and an $\sim 1.5\text{mA}/\text{cm}^2$ reduced J_{sc} . Nevertheless, the PERC cells with sputtered Al_2O_3 as rear passivation achieve an independently confirmed efficiency of 20.1% – the first 20% -efficient solar cell made using a sputtered Al_2O_3 passivation layer.

Conclusions

Despite their lower passivation quality compared to Al_2O_3 films deposited by ALD and by PECVD, we have demonstrated that sputtered Al_2O_3 layers are suitable for the fabrication of 20% efficient PERC cells, while Al_2O_3 deposited by ALD resulted on the same cell structure in efficiencies up to 21.4% . After firing in a conveyor-belt furnace, the SRV provided by Al_2O_3 films deposited by high-rate spatial ALD was found to be below 20cm/s and that of PECVD- Al_2O_3 was in the range 30 – 80cm/s , indicating a very good firing stability of the layers deposited by spatial ALD as well as PECVD. On the other hand, sputtered Al_2O_3 passivation layers degraded to SRVs larger than 300cm/s after firing. We conclude that spatial ALD and PECVD are already compatible with screen-printing, while the firing stability of sputtered Al_2O_3 needs further optimization, e.g. by deposition of hydrogen-rich SiN_x on top of the sputtered Al_2O_3 . As high-throughput PECVD systems are already well introduced in the market, PECVD will, in our opinion,

Rear side passivation	V_{oc} [mV]	J_{sc} [mA/cm^2]	FF [%]	η [%]
Plasma ALD $\text{Al}_2\text{O}_3/\text{SiO}_x$	664	40.7	79.4	21.4*
Thermal ALD $\text{Al}_2\text{O}_3/\text{SiN}_x$	662	40.6	76.9	20.7*
Sputtered Al_2O_3	651	39.1	79.1	20.1*

*Independently confirmed at Fraunhofer ISE Callab

Table 1. One-sun parameters measured under standard testing conditions of the best PERC silicon solar cells with three different rear surface passivations: (i) plasma ALD Al_2O_3 (30nm)/PECVD- SiO_x (200nm), (ii) thermal ALD Al_2O_3 (30nm)/PECVD- SiN_x (100nm), and (iii) sputtered Al_2O_3 (110nm). All cells were fabricated on $0.5\Omega\text{cm}$ FZ p-Si wafers. The aperture cell area is 4cm^2 .

be the preferred short-term deposition technique for Al₂O₃ passivation layers. If the firing stability of sputtered Al₂O₃ layers can be further improved (e.g. by using SiN_x capping layers), this could become another option for the short term. Spatial ALD might be the most interesting medium- to long-term option due to the superior overall precursor use and material properties of atomic-layer-deposited Al₂O₃.

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Andrew Li has recently submitted his Ph.D. thesis for examination at The Australian National University, Canberra, Australia, which investigated the sputtering method for the deposition of aluminium oxide for the surface passivation of silicon solar cells. His research interests include cell design and characterization, particularly of high efficiency solar cells. He is now working for REC, Singapore.

Andres Cuevas has contributed to the development of silicon solar cell technology since 1976, first at the Universidad Politecnica de Madrid, Spain, where he obtained his Ph.D. degree in 1980 and then at ANU, where he now is Professor of Engineering. His current research interests include the characterisation and understanding of the fundamental properties of silicon, the passivation of silicon surfaces using dielectric coatings, and the development of new technologies for n-type silicon solar cells.

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