

Research

Thermal Oxidation for Crystalline Silicon Solar Cells Exceeding 19% Efficiency Applying Industrially Feasible Process Technology

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Thermal oxides are commonly used for the surface passivation of high-efficiency silicon solar cells from mono- and multicrystalline silicon and have led to the highest conversion efficiencies reported so far. In order to improve the cost-effectiveness of the oxidation process, a wet oxidation in steam ambience is applied and experimentally compared to a standard dry oxidation. The processes yield identical physical properties of the oxide. The front contact is created using a screen-printing process of a hotmelt silver paste in combination with light-induced silver plating. The contact formation on the front requires a short high-temperature firing process, therefore the thermal stability of the rear surface passivation is very important. The surface recombination velocity of the fired oxide is experimentally determined to be below $S \leq 38$ cm/s after annealing with a thin layer of evaporated aluminium on top. Monocrystalline solar cells are produced and 19.3% efficiency is obtained as best value on 4 cm^2 cell area. Simulations show the potential of the developed process to approach 20% efficiency. Copyright © 2008 John Wiley & Sons, Ltd.

KEY WORDS: silicon oxide; crystalline silicon solar cells; high-efficiency cells; metallisation

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INTRODUCTION

In order to improve the cost-effectiveness of solar cells from mono- and multicrystalline silicon, the efficiency has to be increased and wafer thickness has to be reduced. One very important component of the cell influencing the performance especially on thin wafers is the rear surface. The increasing demands for optical quality require a higher internal reflection; the electrical quality can be improved applying a more sophisticated surface passivation compared to a full

area aluminium back-surface-field (Al-BSF). Therefore, all designs for high-efficiency silicon solar cells today use a dielectric passivation layer on the rear. Provided that the refractive index of this layer is low enough, this results in a very good optical mirror with high internal reflection which can be further increased by evaporation of a thin layer of aluminium. To extract the carriers from the cell, a point-contact pattern has proven sufficient for good carrier collection.¹ Both cell structures are sketched in Figure 1. A very good surface passivation is required in order to realise an efficiency gain of the passivated rear in comparison to a full area contacted screen-printed Al-BSF.

A common technology meeting the requirements of passivation quality and optical properties is a thick

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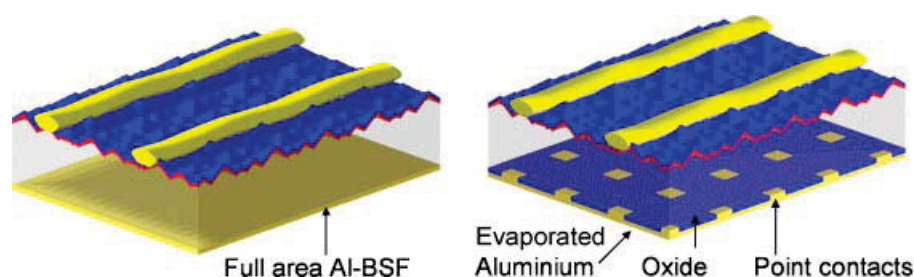


Figure 1. Solar cells structure with screen-printed front metallisation and full area Al-BSF (left) and passivated rear with point contacts (right)

thermal oxidation (≥ 100 nm) grown at high temperatures. This has led to the highest conversion efficiencies on monocrystalline silicon so far.² A technological modification of the standard oxides grown at 1050°C in dry ambience is the use of pyrogenic steam.³ This increases the growth rate and/or reduces the oxidation temperature. The latter is especially beneficial for high-temperature-sensitive material and enabled the first cells in excess of 20% efficiency on multicrystalline silicon using a process at 800°C .⁴ This wet oxidation⁵ is about one order of magnitude faster than the dry oxidation process and therefore a viable technology for industrial silicon solar cell processing.

Following the development of high-efficiency cells using photolithography technology, in the present study cells are manufactured employing an industrial front structure with screen-printed contacts. The front surface consists of a phosphorus doped emitter passivated by a single layer antireflection coating of silicon nitride (SiN_x). The contact grid is screen-printed hotmelt silver paste,⁶ which needs to be fired through the sputtered SiN_x layer. This short high-temperature process of 800 – 900°C can potentially degrade passivation layers. Therefore, a good process design needs to take into account the passivation quality at the end of the process sequence, that is the rear passivation layer has to be temperature stable.

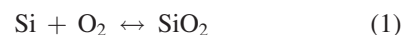
The question to be answered is: Can a wet thermal oxide yield low rear surface recombination velocities and withstand the firing process?

EXPERIMENTAL

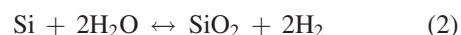
Wet thermal oxidation

Oxidation of the silicon surface is well known to passivate defects at the silicon surface and reduce the

density of interface states (D_{it}).⁷ Since oxidation usually takes place at very high temperatures and needs a considerable long time, it is not broadly used in industrial solar cell mass production nowadays. However, a wet oxidation process can close the gap between laboratory and industrial application due to the strong increase in growth rate.^{3,4,8} This reduces the necessary time/temperature budget. The reason for the increase in the growth rate can be found in the reaction equations:



in the case of dry oxidation and



for wet oxidation. With the H_2O molecule being smaller than the O_2 molecule its diffusion through the already existing oxide layer to the Si/SiO_2 interface is much faster^{9,10} and consequently about an order of magnitude in processing time can be gained, see Figure 2. This makes wet oxidation a viable technology for industrial application.

However, in order to enable highly efficient solar cells it is of utmost importance that the silicon surface is well passivated. The effective surface recombination velocity S_{eff} including recombination at the laser-fired contacts (LFC) on the rear side of the cell¹¹ was determined from carrier lifetime measurements.

Therefore, FZ wafers with specific base resistivities in the range of $\rho \approx 0.5 \Omega \text{ cm}$ to $\rho \approx 7 \Omega \text{ cm}$ were oxidised at 800°C under pyrogenic steam or at 1050°C without steam but additional DCE (trans-1,2-dichloroethylene). Oxides of about 100 nm thickness were grown and $1 \mu\text{m}$ of aluminium evaporated on both surfaces. Three wafers of the same base resistivity were divided into four quarters. On both sides, LFC were applied with three different spacings (250, 500 and $1220 \mu\text{m}$ distance between contact points in a rectangular pattern), the fourth quarter was not

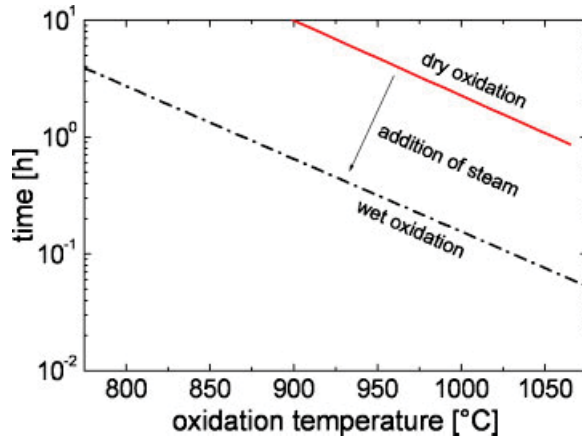


Figure 2. Time/temperature budget necessary to grow a 100 nm oxide on $\langle 100 \rangle$ oriented silicon calculated with data given in [10]

contacted (no LFC). After a forming gas anneal (25 min at 425°C) the aluminium was etched off in hydrochloric acid. The effective carrier lifetime of these *annealed*¹² wafers was measured with the QssPC method.¹³ The excess carrier density for data evaluation was chosen to be $\Delta n = 1 \times 10^{15}/\text{cm}^3$ in order to circumvent any trapping artefacts.¹⁴ The effective surface recombination velocity S_{eff} was calculated applying Equation (3)¹⁵

$$\frac{1}{\tau_{\text{eff}}} \cong \frac{1}{\tau_{\text{bulk}}} + \frac{2S}{W} \quad (3)$$

assuming a high bulk lifetime of 1800 μs .¹⁶ Therefore, the calculated S_{eff} -values represent the upper limit. Each data point in Figure 3 represents the arithmetic average of three samples. Very low surface recombination velocities show the high level of surface passivation of the oxidised silicon. The obtained data were fitted by minimising the deviation between the measured values and the calculations according to

$$S_{\text{eff}} = S_0 \left(\frac{N_A}{N_{\text{onset}}} \right)^\alpha \quad (4)$$

S_0 shifts the calculated curve in vertical direction, N_A is the base doping, N_{onset} is the first evaluated doping concentration to normalise the fraction in brackets and α is the exponent which determines the slope. Such parameterisation was also used by Cuevas,¹⁷ Dicker¹⁸ and Kray¹⁹ who found similar values. The parameters for the wet oxide are given in Table I.

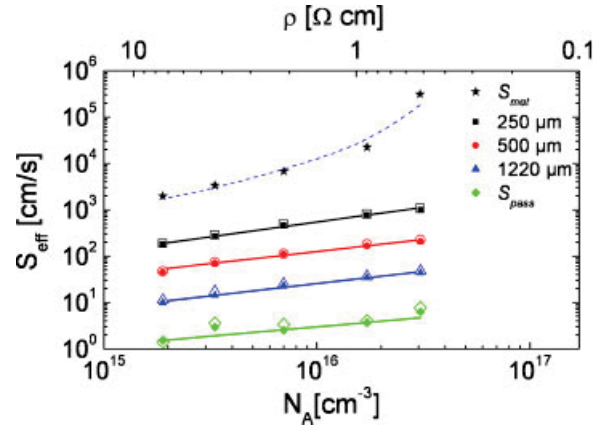


Figure 3. Upper limit of effective rear surface recombination velocity of wafers passivated with 105 nm of thick oxide grown under dry (hollow symbols) and wet (solid symbols) conditions. Different contact spacings were applied. The lines are best fit according to Equation (4) except for S_{met} , where the dashed line is a guide-to-the-eye only

Fischer²⁰ published an analytical model to calculate the surface recombination velocity of solar cells with point-contacts on the rear.

$$S_{\text{eff}} = \frac{D_e}{W} \left(\frac{L_p}{2W\sqrt{\pi f}} \arctan \left(\frac{2W}{L_p} \sqrt{\frac{\pi}{f}} \right) - \exp \left(-\frac{W}{L_p} \right) + \frac{D_e}{fWS_{\text{met}}} \right)^{-1} + \frac{S_{\text{pass}}}{1-f} \quad (5)$$

D_e denotes the diffusion constant of minority carriers, W the wafer thickness, L_p the contact pitch, f the metallisation fraction, S_{met} and S_{pass} the surface recombination velocity at the metallised and passivated areas of the rear, respectively. Plagwitz²¹ experimentally verified the model via the diode saturation current on carrier lifetime samples for amorphous silicon as passivation layer, Kray and Glunz²² applied the model to silicon oxide passivated

Table I. Fit parameters according to Equation (4) for S_{eff} as a function of base doping and LFC pitch L_p

Oxide	Pitch (μm)	S_0 (cm/s)	N_{onset} (cm^{-3})	α
Wet	∞	2	1.9×10^{15}	0.47
Wet	1220	12	1.9×10^{15}	0.46
Wet	500	56	1.9×10^{15}	0.47
Wet	250	267	1.9×10^{15}	0.42

rear surfaces with photo-lithographically defined contacts and LFC. We used the model to extract the surface recombination velocity under the metal contacts S_{met} . This required the fraction of metallised area that can easily be calculated when the contact radius of the LFC point is known. Kray²² determined this contact radius to be 46 μm for the same set of laser parameters used in this study. Also for S_{met} a clear dependence on the base doping concentration was observed (Figure 4). This is explained by the aluminium alloy under the contact point which creates a local back-surface-field and is more effective for lowly doped substrates.²³

Passivation after firing process

The previous experiment shows the excellent passivation quality of the wet oxide to be equal to the dry oxide after the aluminium annealing. But: Does the firing process for the screen-printed front metallisation degrade the passivation of the thick thermal oxide?

To answer this question, the following experiment was set up: 330 μm thick boron doped FZ silicon wafers of 1 Ωcm were cleaned and oxidised in steam ambience at 950°C. The wafers were exposed to the

firing step at about 810°C peak temperature in a belt furnace. Subsequently 2 μm of aluminium were evaporated on the surfaces and the wafers were annealed for 15 min at 350°C in forming gas. This anneal procedure is believed to hydrogenate the silicon surface, the hydrogen being the product of a reaction of aluminium with residual water in the silicon oxide layer.¹² The atomic hydrogen passivates the dangling bonds at the silicon/silicon oxide interface created during the firing process which improves surface passivation. The metal layers were etched off in hydrochloric acid before measuring the minority carrier lifetime using the QssPC method. The average effective carrier lifetime was 335 μs which, after evaluation according to Equation (3), results in an upper limit of $S \leq 38\text{ cm/s}$. Using Equation (4) this can be converted to 82 cm/s for 0.5 Ωcm material. Applying LFC with a contact pitch of 750 μm , this results in effective surface recombination velocities of $S_{\text{eff}} = 112$ and 143 cm/s, respectively (Equation 5). Table II summarises the conversion calculations.

Although the recombination velocities are significantly higher than the values derived for the wet oxide without the firing process, this is low enough to provide excellent surface passivation and allows for highly efficient solar cells. Therefore, we implemented the

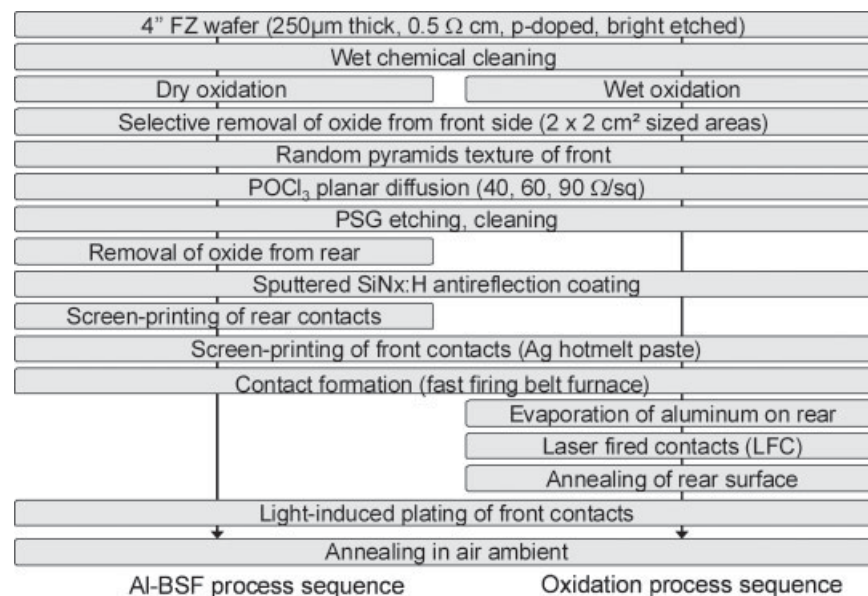


Figure 4. Process flow diagram of manufactured solar cells featuring screen-printed front contacts thickened by light-induced plating. The left flow shows the Al-BSF process, on the right-hand side the rear side is passivated by wet thermal oxide

Table II. Effective rear surface passivation S_{eff} in the case of wet oxide with and without a metallisation firing process. Contact pitch is $L_p = 750 \mu\text{m}$. The calculations use Equations (4) and (5)

Base resistivity ($\Omega \text{ cm}$)	Not fired		Fired at 810°C	
	0.5	1.0	0.5	1.0
S_{pass} (cm/s)	8	5	54	38
S_{met} (cm/s)	2×10^5	2×10^4	2×10^5	2×10^4
S_{eff} (cm/s)	96	78	143	112

oxide in the process sequence as is described in the following section.

Solar cells results

The base material used for the solar cells processed in this study was $250 \mu\text{m}$ thick boron-doped, high-quality FZ silicon of $0.5 \Omega \text{ cm}$ base resistivity. Both, cells with a standard industrial Al-BSF and cells with an oxide passivation, were processed in order to experimentally determine the benefit of the point-contacted oxide passivated rear compared to the full area Al-BSF. Recently developed technologies for crystalline silicon solar cell production like wet oxidation,⁴ sputtering of SiN_x ,²⁴ screen-printing of hotmelt silver paste,⁶ LFC¹¹ and light-induced silver plating²⁵ were implemented in an industrial process sequence shown in Figure 4 resulting in the cell structures illustrated in Figure 1.

The results of the IV-measurements of the best cells under standard testing conditions are shown in Table III. The series resistance R_s was determined by comparison of illuminated and dark IV-measurement^{18,26} under operating conditions. For all emitter diffusions the V_{oc} and j_{sc} values are significantly increased for the oxide passivated cells. The reason can be seen in the quantum efficiency measurements shown in Figure 5.

In the short wavelength region and therefore linked to the properties of the front surface of the cells, the

difference in the quantum efficiency is due to the different emitter doping profiles and thus the two types of cell structure show equal performance. In the long wavelength region, and therefore showing the rear surface properties, the Al-BSF and the oxide passivated rear exhibit distinct differences. The superior rear surface passivation and the high internal reflectance lead to a significant gain in the short-circuit current density and open circuit voltage compared to the Al-BSF cells. Additionally, differences in the reflectance between 1150 nm and 1200 nm can be observed for the three different emitters. The reason can be found in free carrier absorption²⁷ of the long wavelength photons in the highly doped emitter layers.²⁸ This effect is only seen for the cell structure with good light-trapping, where the photons are reflected several times inside the cell. Therefore, the application of the lightly doped emitter on the front leads to slightly higher quantum efficiencies also in the long wavelength region.

The fill factor (FF) of the point-contacted cell is slightly lower than FF of the full area contacted Al-BSF. This can partially be explained by increased series resistance on the rear due to the point-contact structure and increased spreading resistance.^{20,29} An additional contribution of the point-contact structure of $\Delta R_s = 0.14 \Omega \text{ cm}^2$ (see Table III) was measured for the $40 \Omega/\text{sq}$ emitter. Therefore the differences in FF are

Table III. IV-parameters of the best solar cells with Al-BSF or oxide passivated rear

Wafer. cell-ID	R_{sh} (Ω/sq)	Rear side	V_{oc} (mV)	j_{sc} (mA/cm^2)	FF (%)	η (%)	R_s ($\Omega \text{ cm}^2$)
32_6-4	40	Al-BSF	632	35.2	81.2	18.1	0.46
33_7-1		Oxide passivation	639	36.7	80.0	18.8	0.6
32_15-4	60	Al-BSF	632	35.6	81.2	18.3	0.49
33_13-1		Oxide passivation	647	37.1	79.6	19.1	n.a.
32_24-4	90	Al-BSF	640	36.5	79.8	18.7	0.74
33_19-2		Oxide passivation	655	38.2	76.9	19.3*	1.19

*Calibrated measurement at Fraunhofer ISE calibration laboratory.

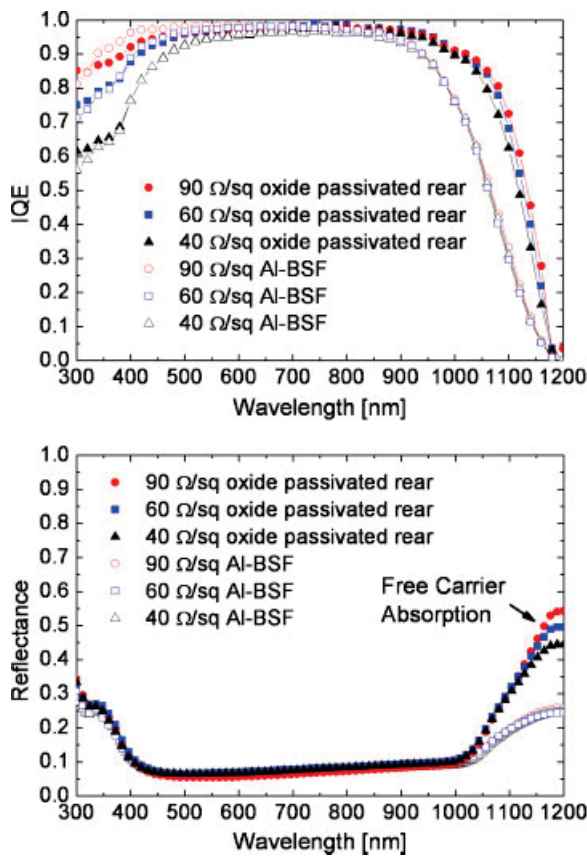


Figure 5. Measurement of internal quantum efficiency IQE and reflectance for the Al-BSF and for the oxide passivated rear

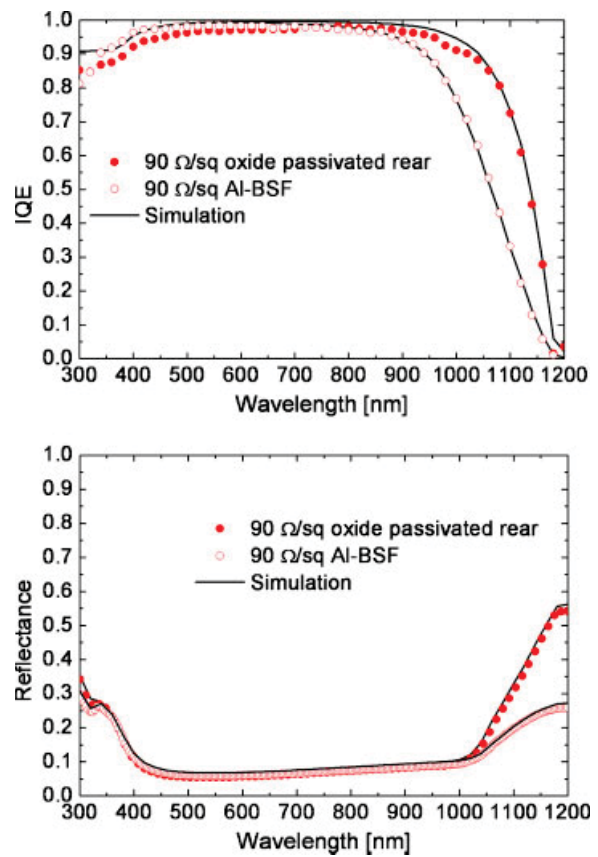


Figure 6. Simulated internal quantum efficiency and reflectance (lines) fitted to the measurements (dots)

small for the 40 and the 60 Ω/sq emitter, for the 90 Ω/sq emitter the difference in series resistance was determined to be $\Delta R_s = 0.45 \Omega \text{ cm}^2$. This resulted in about 3% absolute loss in fill factor, which cannot be explained by the cell design but must be due to another technological problem. However, the high gain in j_{sc} and V_{oc} still allow for a gain of 0.6% absolute. An electrical conversion efficiency of 19.3% was measured in a calibrated measurement at Fraunhofer ISE CalLab. This efficiency is, to the knowledge of the authors, the highest value reported for a screen-printed front metallisation pattern to date.³⁰

Modelling

In order to assess the potential of the cells structure and the process, one-dimensional modelling using PC1D³¹ was performed. The doping profile of the 90 Ω/sq

emitter was measured by SIMS and used for the Al-BSF and the oxide passivated rear. The simulation results can be seen in Figure 6.

The fit parameters used are summarised in Table IV, Sim 1 and Sim 2 model the measured cells of Table III. For the rear surface an effective surface recombination velocity of $S_{eff} = 1200 \text{ cm/s}$ for the Al-BSF cell was used for a good fit, the calculated $S_{eff} = 143 \text{ cm/s}$ for the oxide passivated cell well suits the experimental data. Internal reflectance values used for modelling of the internal reflectance at the rear surface were 72% diffuse for Al-BSF and 94% specular for oxide passivation. Also given in Table IV is the simulation for the potential of the oxide passivated cell for optimum performance (Sim 3). This simulation assumes that the front surface is identical to the Al-BSF cell and that the series resistance is $R_s = 0.88 \Omega \text{ cm}^2$. The latter assumption is obtained by adding $\Delta R_s = 0.14 \Omega \text{ cm}^2$ to the Al-BSF cell data.

Table IV. Internal reflectivities (R_{back}) and effective rear surface recombination velocities at the back (S_{eff}) as extracted from solar cell modelling ((d) is diffuse and (s) is specular reflection). In addition the IV-parameter simulation results are presented

Wafer. Cell-ID	R_s ($\Omega \text{ cm}^2$)	Rear side	R_{back} (%)	S_{eff} (cm/s)	V_{oc} (mV)	j_{sc} (mA/cm ²)	FF (%)	η (%)
Sim 1	0.74	Al-BSF	72 (d)	1200	644	36.3	80.0	18.7
Sim 2	1.19	Oxide	94 (s)	143	651	38.4	77.4	19.4
Sim 3	0.88	Oxide	94 (s)	143	651	38.4	79.2	19.8

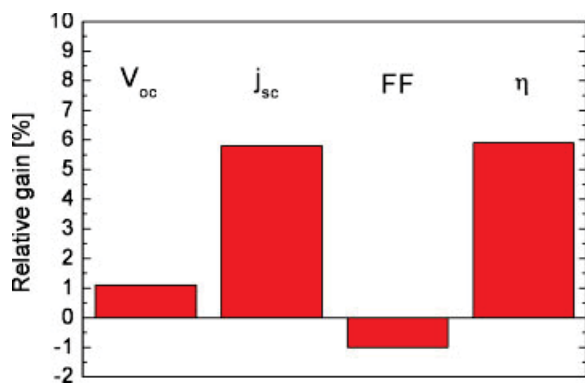


Figure 7. Simulation results illustrating the relative gain of solar cell parameters by using the oxide passivation instead of a full area Al-BSF. Modelling refers to a 240 μm thick solar cell with a base doping of 0.5 Ωcm and an emitter sheet resistance of 90 Ω/sq

This results in a FF of 79.2% and therefore 19.8% efficiency.

The relative gain by using the oxide passivation instead of a full area Al-BSF is shown in Figure 7. According to the simulation, the improved rear surface passivation and higher internal reflectance increase V_{oc} and j_{sc} by 1% and 6%, respectively. FF decreases by 1% due to spreading resistance. This sums up to an overall relative gain in efficiency of nearly 6%.

Summary

The wet thermal oxide process shows the same electrical performance as the standard dry oxidation used for high-efficiency monocrystalline silicon solar cells. The oxidation was successfully implemented in an industrially feasible production sequence with fine-line screen-printed and plated front contacts on a low sheet resistance emitter of 90 Ω/sq . Although passivation quality degraded during the firing step, the effective surface passivation was still on a high level. The point-contacted rear side was experimentally

compared to the standard industrial rear surface contact, the full area Al-BSF. Best cell efficiencies were 18.7% for Al-BSF and 19.3% for the oxide passivated rear surface. This is due to superior surface passivation and light-trapping as could also be shown in one-dimensional modelling. The potential of the process was calculated to approach 20% efficiency.

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