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Analysis of the Silicon Dioxide Passivation and Forming Gas Annealing in Silicon Solar Cells

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Abstract

The passivation of silicon solar cells by the deposition of SiN_x anti-reflection coating is usual in the industry. However, materials such as SiO_2, TiO_2 and Al_2O_3 may be a cost-effective alternative and its analysis is mainly reported in silicon wafers. The goal of this paper is to present the development and analysis of silicon solar cells passivated with a thin layer of SiO_2 as well as the evaluation of the effectiveness of the annealing step in forming gas. The dry oxidation was performed before the TiO_2 anti-reflection coating deposition and the annealing in forming gas was performed in the same furnace. The temperature and time of the oxidation and the annealing step were experimentally optimized. The efficiency of 15.9 %was achieved. The highest average efficiency was found in the oxidation temperature range from 750 ºC to 800 ºC, during 7 minutes, caused by the increasing of open circuit voltage and fill factor. At short wavelengths, the internal quantum efficiency decreases slightly with the increasing of the oxidation temperature. The minority carrier diffusion length (L_D) of the solar cells processed with the oxidation temperature of 800 °C was around 1890 μm. The open circuit voltage shows a slight trend of increasing with the oxidation time. The annealing step in forming gas did not improve the average efficiency of the solar cells. Solar cells processed with oxidation and annealing presented higher internal quantum efficiency at short wavelengths than cells with only annealing.

Keywords: silicon solar cells, passivation, silicon oxide

1. Introduction

The industry of silicon solar cells is mainly based on conventional screen printing metallization and passivation of the phosphorus emitter with silicon nitride anti-reflection (AR) coating. The passivation reduces the recombination of minority charge carriers in the surface of the solar cells. The surface passivation with silicon nitride using plasma-enhanced chemical vapor deposition (PECVD) at low temperature (< 450 ºC) results in low surface recombination velocity on both p-type and n-type c-Si, as well as low absorption at short wavelengths (Aberle, 2001). Silicon nitride passivation for n^+ emitter provides positive interface charges, causing a field effect passivation (Rahman and Khan, 2012). Low surface recombination velocity of 1.6 cm/s on p-type c-Si with passivation by SiN_x deposited at 290 ºC was achieved (Wan et al., 2013).

The passivation with Al_2O_3 is also under investigation. The aluminum oxide has advantages over SiN_x when used to passivate the p^+ emitter, because Al_2O_3 presents a high density of fixed negative charges (Pawlik et al., 2014; Saynova et al., 2013). Surface recombination velocities below 2.9 cm/s and low interface state density were achieved with Al_2O_3 layers, performed by atomic layer deposition (ALD) (Werner et al., 2011). The surface recombination velocity of 90 cm/s was reported with Al_2O_3 as rear surface dielectric layer and this value was similar to that achieved with thermally grown SiO_2 (Schmidt et al., 2008). Duttagupta et al. (2013) reported that good passivation can be obtained with the AlO_x/SiN_x dielectric stack, deposited by PECVD and an AlO_x thickness of 5 nm is sufficient.

Thermally grown SiO_2 is the most effective c-Si surface passivation technique, but silicon dioxide has a low

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refractive index and, consequently, it is not good to form the anti-reflection coating. In this way, silicon dioxide is usually complemented by other dielectric layers. The interface defect density may be low after the growth of a thermal SiO_2 film on c-Si and a subsequent annealing in forming gas. The surface passivation behavior has also been investigated for SiO_2/SiN_x stack and the surface recombination velocity of 2.4 cm/s was found in n-type Cz silicon wafers passivated by SiO_2 thermally grown (Ye et al., 2010). Low dark current densities of <20 fA/cm^2 on textured n-type FZ silicon wafers were found. In this substrate, both surfaces were phosphorus doped and the sheet resistance was of 140Ω/□ (van Erven et al., 2008). In the frontal surface field solar cells with very low surface phosphorous concentrations, the passivation with a SiO_2/SiN_x stack demonstrated an increase in cell efficiency of around 0.5 % (absolute), compared to the standard PECVD-SiN_x passivation. For instance, n-type CZ-Si solar cell passivated by the SiO_2/SiN_x stack achieved the efficiency of 19.4% (Book et al., 2011). Another technique investigated was a stack of hydrogenated amorphous silicon and SiO_x deposited by PECVD and the surface recombination velocity of 120 cm/s on p-type FZ-Si substrate was achieved (Hofmann et al., 2008).

Titanium oxide may not provide efficient surface passivation, but it is used to form the AR coating, because presents low absorption in short wavelengths and has a high refractive index. This material is more suitable to passivate boron doped surfaces, but the technique used to deposit the layer influences the passivation. A layer of TiO_2 performed by atmospheric pressure chemical vapor deposition leads to a surface recombination velocity less than 30 cm/s, on a 200-Ω/□ boron emitter (Thomson and McIntosh, 2012). On the other hand, titanium oxide films performed by thermal atomic layer deposition provide the surface recombination velocity of 2.8 cm/s and 8.3 cm/s on n-type and p-type FZ-Si substrate, respectively (Liao et al., 2014).

As presented above, surface velocity recombination can be reduced by passivation using a dielectric film and by performing a surface electric field that repels the minority charge carriers. In this way, Bonilla and Wilshaw (2014) developed a technique using positively charged alkali ions (Na or K) embedded in a silicon oxide grown in a dry dichloroethylene (DCE) environment. Surface recombination velocities in the range of 6–15 cm/s were obtained in n-type FZ-Si.

Many studies were reported on the surface velocity recombination in Si-wafers, but the experimental analysis of the passivation on the performance of Si solar cells needs more investigation. Otherwise, the passivation with a thin layer of silicon dioxide, growth at temperature below 800 °C and combined with the TiO_2 AR coating, may be an alternative to the silicon solar cell industry. The goal of this paper is to present the development and analysis of the surface passivation with a thin layer of SiO_2, with and without annealing in forming gas, on the electrical parameters of silicon solar cells. The AR coating was formed with TiO_2 deposited by e-beam evaporation.

2. Solar cell processing and characterization

Solar cells were developed in p-type CZ-Si solar grade wafers with thickness of 200 μm and base resistivity from 1 Ω.cm to 20 Ω.cm. The process sequence to produce solar cells consists of the following steps: texture etching, RCA cleaning, phosphorus diffusion, phosphorus silicate glass (PSG) etching and RCA cleaning, dry oxidation and forming gas annealing, anti-reflection coating deposition, screen printing metallization on the frontal and rear face, firing of the metal pastes and laser edge isolation.

The n^+ layer was performed by phosphorus diffusion at 845 ºC using $POCl_3$ as source, resulting in the sheet resistance around 60 Ω/□. The thermal oxidation was carried out in a specific quartz tube furnace and the forming gas annealing was performed in the same furnace. The thickness of the TiO_2 anti-reflection coating, deposited by the e-beam technique, was of 60 nm. The depth of aluminum back surface field (Al-BSF) of around (5 ± 1) μm was estimated taking into account the peak temperature firing of 840 °C and the Al paste surface density of about 3.5 mg/cm^2 (Zanesco et al., 2014).

The temperature during thermal oxidation was ranged from 650 ºC to 900 °C and the short time of the oxidation was from 4 to 15 minutes, resulting in a thin layer of silicon dioxide of around 10 nm. The forming gas annealing temperature was ranged from 350 °C to 450°C and the processes were performed during 20 and 30 minutes.

Solar cells were characterized under standard conditions (100 mW/cm^2, AM1.5G and 25°C) in a solar simulator calibrated with a solar cell previously measured at CalLab - FhG-ISE (*Fraunhofer-Institut für Solare Energiesysteme*), Germany. The two-dimensional distribution of minority carrier lifetime and diffusion length was measured using the WT-2000PV device of Semilab, by µ-PCD (microwave induced photoconductivity decay) and LBIC (light beam induced current) technique, respectively. The minority carrier lifetime was measured after the thermal oxidation and with a SiO_2 layer in order to compare the passivation. The internal quantum efficiency was calculated starting from the measured external quantum efficiency and reflectance.

3. Results and analysis

3.1. Influence of the oxidation temperature

Tab. 1 presents the average values of the open circuit voltage (V_{oc}), short-circuit current density (J_{sc}), fill factor (FF) and efficiency (η) of the solar cells processed with different oxidation temperature (T_{oxi}), during 7 minutes. The highest average efficiency was found in solar cells processed with the oxidation temperature range from 750 ºC to 800 ºC. Comparing solar cells with and without passivation in Tab.1, we observe that the average efficiency increases from (14.3 ± 0.3)% to (15.6 ± 0.1)%, due to the SiO_2 passivation. The increasing of the efficiency of 1.3 % (absolute) is caused by the behavior of V_{oc} and FF. Moreover, Tab. 1 and Fig. 1-d show that the optimization of the SiO_2 passivation reduced also the standard deviation.

The electrical parameters of the best solar cells are reported in Tab. 2. The highest efficiency of 15.9 % was achieved in a solar cell processed at the oxidation temperature of 800 °C, due to mainly the increasing of the V_{oc} and FF, caused by the passivation.

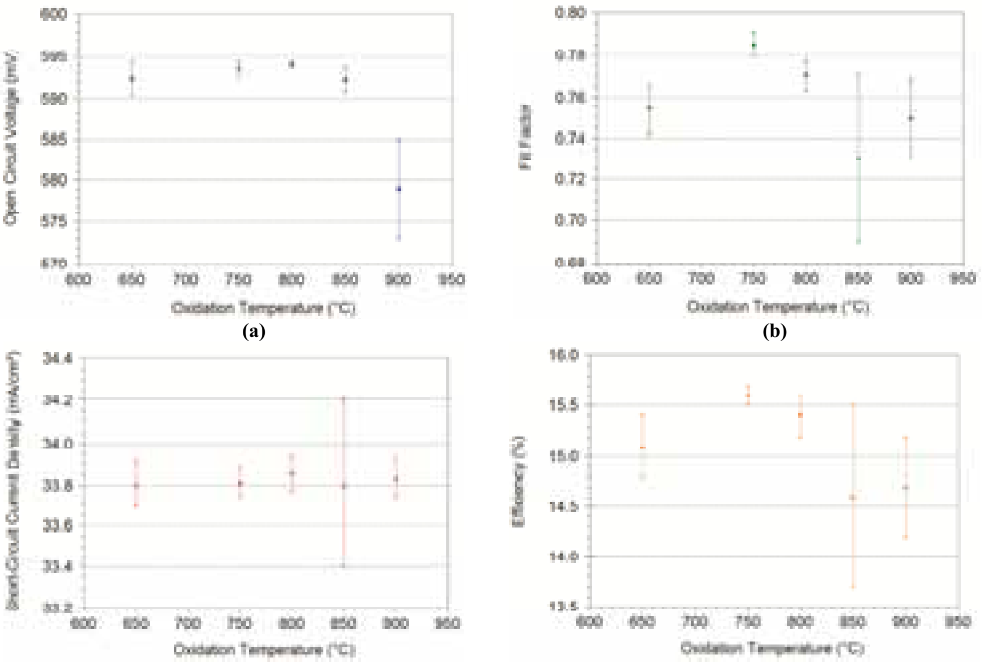
Tab. 1: Average electrical characteristics of the silicon solar cells as a function of oxidation temperature (T_{oxi}).

T_{oxi} (°C)	Nº of Cells	V_{oc} (mV)	J_{sc} (mA/cm^2)	FF	η (%)
Without oxidation	18	586.9 ± 2.3	33.85 ± 0.06	0.719 ± 0.013	14.3 ± 0.3
650	18	592.3 ± 1.9	33.80 ± 0.11	0.754 ± 0.012	15.1 ± 0.3
750	18	593.4 ± 0.9	33.81 ± 0.07	0.785 ± 0.006	15.6 ± 0.1
800	18	593.9 ± 0.3	33.85 ± 0.08	0.770 ± 0.007	15.4 ± 0.2
850	17	592.2 ± 1.4	33.8 ± 0.4	0.73 ±0.04	14.6 ± 0.9
900	18	579 ± 6	33.83 ± 0.09	0.749 ± 0.019	14.7 ± 0.5

Tab. 2: Electrical characteristics of the silicon solar cells with higher efficiency as a function of oxidation temperature.

T_{oxi} (°C)	V_{oc} (mV)	J_{sc} (mA/cm^2)	FF	η (%)
Without oxidation	589.3	33.8	0.75	15.0
650	593.0	33.7	0.76	15.5
750	595.0	33.8	0.79	15.6
800	593.3	33.9	0.78	15.9
850	593.4	33.8	0.76	15.2
900	582.7	33.9	0.76	14.7

The Fig. 1 (a) and (b) show that higher values of open circuit voltage and fill factor were achieved at temperature oxidation of 800 °C and 750 °C, respectively. The V_{oc} increased about 7 mV when passivation step was implemented. The passivation affects also the fill factor. This parameter augments from 0.72 to 0.78 with the oxidation at temperature of 750 ºC – 800 ºC. The oxide growth at 900 °C reduces the V_{oc}, when compared to the results without oxidation. This temperature may cause a degradation of the substrate. Fig. 1 (c) indicates that the J_{sc} was not affected by the oxidation temperature and the SiO_2 passivation, because of the efficient aluminum back surface field (Al-BSF). Consequently, the higher efficiencies occur at the oxidation temperature of 750 °C and 800 °C, as Fig.1 (d) confirms.



(a) (b)

(c) (d)

Fig. 1: (a) Open circuit voltage, (b) fill factor, (c) short-circuit current density and (d) efficiency of the solar cells as a function of temperature oxidation.

The Fig. 2 indicates that at short wavelengths, the internal quantum efficiency (IQE) decreases slightly with the increasing of the oxidation temperature because the SiO_2 passivation is less effective. With the increase of the oxidation temperature from 850 °C to 900 °C, the surface recombination velocity enhances of about two orders of magnitude, indicating that the SiO_2 passivation is worst.

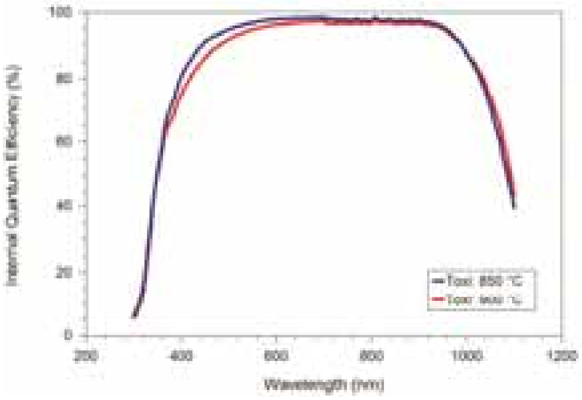


Fig. 2: Internal quantum efficiency of the silicon solar cells with the highest efficiency and processed at temperature oxidation of 850 °C and 900 ºC.

The effective minority carrier lifetime (τ_{eff}) was low for the oxidation temperatures evaluated and depends on the oxidation temperature, as Fig. 3 shows. The highest average values of 40 μs - 50 μs were found at temperature oxidation from 750 °C to 850 °C.

It's worth mentioning that the average initial minority carrier lifetime measured in 60 solar grade silicon wafers was of 39 μs and the final effective value takes into account the double pn junction formed by phosphorus diffusion, passivated with SiO_2 growth at different temperatures. Then, the process to fabricate the solar cells with the oxidation temperature range of 750 - 850 °C leads to the minority carrier lifetime slightly higher than the initial value. Fig. 3 shows also that low values of the minority carrier lifetime occur in the center of the wafers. For the range of oxidation temperature from 750 °C to 850 °C, the highest values of the lifetimes are of around 110 μs.

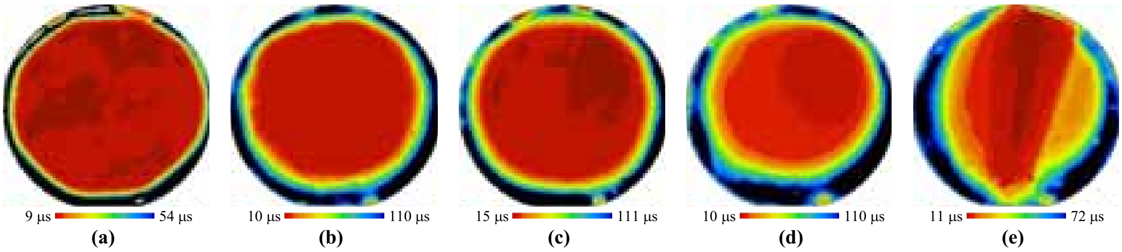


Fig. 3: The two-dimensional distribution of the minority carrier lifetimes measured by μ-PCD technique afterwards the oxidation during 7 minutes and temperature of (a) 650 ºC (τ_{eff} = 21 μs), (b) 750 ºC (τ_{eff} = 42 μs), (c) 800 °C (τ_{eff} = 43 μs) (d) 850 °C (τ_{eff} = 49 μs) and (e) 900 °C (τ_{eff} = 33 μs).

The Fig. 4 shows the comparison of the minority carrier diffusion length (L_D) of the solar cells processed with the oxidation temperature of 800 °C and 900 °C and without oxidation. The average L_D of the solar cells without oxidation step is around 1380 μm and this parameter enhances to 1890 μm, when the oxidation is performed at 800 °C. However, when the oxidation temperature is of 900 °C, the average minority carrier diffusion length decreased to 960 μm, indicating that the substrate was degraded during the oxidation process.

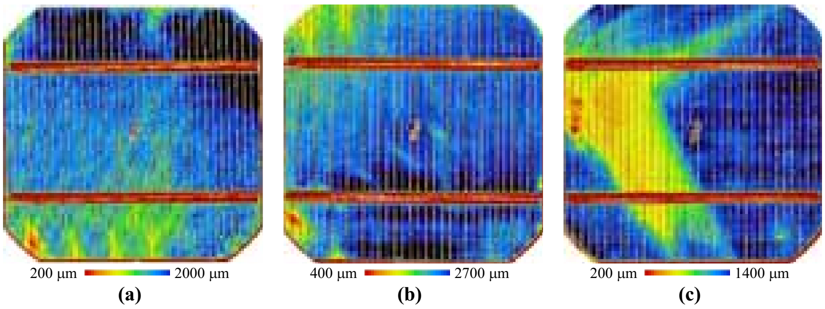


Fig. 4. The two-dimensional distribution of the minority carrier diffusion length of solar cells processed (a) without oxidation (L_D = 1380 μm) and (b) with oxidation temperature of 800 °C (L_D = 1890 μm) and (c) of 900 °C (L_D = 960 μm).

3.2. Influence of the oxidation time

The electrical characteristics of the solar cells processed by the oxidation time from 4 minutes to 15 minutes at the temperature of 800 ºC are presented in Tab.3. The highest average efficiency was found when cells were thermally oxidated during 7 minutes. The V_{oc} shows a slight trend of increasing with the oxidation time. Nevertheless, the J_{sc} decreases slightly when the oxidation time is increased, as Tab. 4 confirms. The Tab. 4 indicates that the highest efficiency of 15.9 % is obtained in solar cells processed at the oxidation temperature of 800 °C during 7 minutes.

Tab. 3: Average electrical characteristics of the silicon solar cells as a function of oxidation time (t_{oxi}).

t_{oxi} (min)	Nº of Cells	V_{oc} (mV)	J_{sc} (mA/cm^2)	FF	η (%)
4	17	593.2 ± 1.0	33.88 ± 0.11	0.751 ± 0.011	15.0 ± 0.3
7	18	593.9 ± 0.3	33.85 ± 0.08	0.767 ± 0.007	15.4 ± 0.2
15	15	594.1 ± 0.9	33.82 ± 0.10	0.753 ± 0.014	15.0 ± 0.3

Tab. 4: Electrical characteristics of the fabricated silicon solar cells with the highest efficiency as a function of oxidation time.

t_{oxi} (min)	V_{oc} (mV)	J_{sc} (mA/cm^2)	FF	η (%)
4	593.9	34.0	0.76	15.3
7	593.3	33.9	0.78	15.9
15	594.0	33.7	0.77	15.4

3.3 Analysis of the annealing step

Batches of solar cells were processed at temperature oxidation of 800 °C during 7 minutes and the annealing temperature and time were ranged. Starting from the results summarized in Tab.5, we observe that the annealing in forming gas at 400 °C did not improve the average efficiency of solar cells when compared to the results presented in Tab. 1. Moreover, the annealing temperature of 350 °C and 450 °C lead to lower efficiencies than that presented in Tab. 1 for the oxidation temperature of 800 °C, indicating that the annealing process at these temperatures did not improve the efficiency of the solar cells.

The open circuit voltage tends to increase with long annealing time. For annealing temperature of 400 °C and 450 °C, the V_{oc} increased about 2.5 – 3.0 mV, when the annealing time was increased from 20 minutes to 30 minutes. This result is confirmed in Tab. 6 that reports the electrical characteristics of the solar cells with the highest efficiency. For the best cells, the V_{oc} was improved up to 3 mV when the annealing time is augmented of 10 minutes.

With the annealing without the previous oxidation, an average efficiency of (15.3 ± 0.2) % was achieved, as shown in Tab. 5. This value is only 0.3 % (absolute) lower than the value obtained in devices with oxidation and annealing in forming gas and the reduction of the efficiency was caused by the decrease of V_{oc}, J_{sc} and FF.

Tab. 5: Average electrical characteristics of the solar cells processed with different parameters of forming gas annealing.

T_{An} (°C)	t_{An} (min)	Nº of Cells	V_{oc} (mV)	J_{sc} (mA/cm^2)	FF	η (%)
350	20	7	591.2 ± 1.3	33.84 ± 0.09	0.756 ± 0.017	15.0 ± 0.3
400	20*	6	590.7 ± 0.4*	33.73 ± 0.06*	0.774 ± 0.003*	15.3 ± 0.2*
	20	3	593.8 ± 0.8	33.88 ± 0.10	0.782 ± 0.007	15.6 ± 0.1
	30	2	596.7 ± 1.1	33.81 ± 0.11	0.800 ± 0.004	15.6 ± 0.1
450	20	5	591.5 ± 1.4	33.75 ± 0.10	0.766 ± 0.008	15.1 ± 0.3
	30	4	593.9 ± 0.5	33.78 ± 0.07	0.757 ± 0.022	15.1 ± 0.1

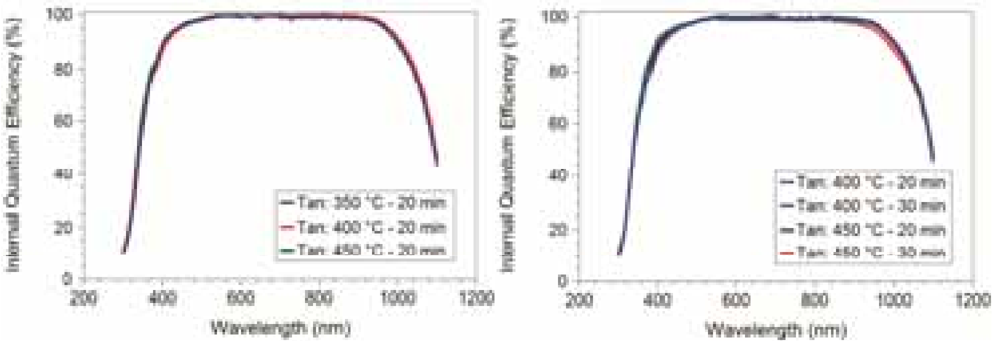
* with annealing and without previous oxidation.

Tab. 6. Electrical characteristics of the best silicon solar cells processed with different parameters of forming gas annealing.

T_{An} (°C)	t_{An} (min)	V_{OC} (mV)	J_{SC} (mA/cm^2)	FF	η (%)
350	20	591.1	33.7	0.77	15.4
400	20*	591.4*	33.7*	0.78*	15.4*
	20	594.1	33.8	0.79	15.6
	30	595.9	33.9	0.80	15.6
450	20	591.5	33.8	0.78	15.4
	30	594.4	33.9	0.77	15.4

* with annealing and without previous oxidation

The short-circuit current density is slightly affected by the annealing process, as Tab. 5 reports. The annealing temperature and time do not influence the J_{sc} and this conclusion is confirmed in Fig. 5 that compares the internal quantum efficiency of the best processed solar cells as a function of annealing temperature and time. Otherwise, the oxidation step improves the passivation, as shown in Fig 6. In this figure, the internal quantum efficiency of the solar cell processed at 400 °C during 20 minutes with oxidation and annealing steps is compared to the IQE of the cell processed with annealing and without oxidation. At short wavelengths, the IQE of cells with SiO_2 passivation is higher, indicating the effectiveness of the passivation. When the oxidation is implemented, the surface recombination velocity was reduced of about one order of magnitude.



(a) **(b)**

Fig. 5: Internal quantum efficiency of the silicon solar cell with the highest efficiency as a function of (a) annealing temperature and processed during 20 minutes and (b) annealing time at the temperature of 400 °C and 450 °C.

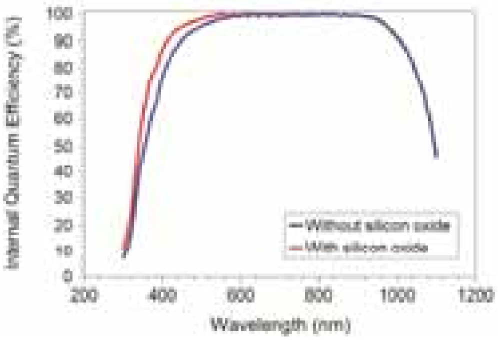
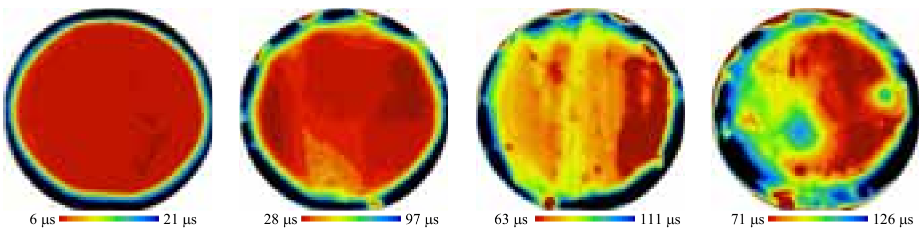


Fig. 6: Internal quantum efficiency of the silicon solar cell processed with and without oxidation step and annealing at the temperature of 400 °C during 20 minutes.

The Fig. 7 illustrates the two-dimensional distribution of the minority carrier lifetimes after the oxidation at the temperature of 800 °C during 7 minutes and annealing step during 20 minutes with different temperatures. In the sample without oxidation (see Fig. 8-a), but with annealing step, the average value of τ_{eff} was of 10 µs. This result confirms that oxidation at 800 °C improves the minority carrier lifetime, as Fig. 7 (c) shows. Values of the average effective minority carrier lifetime of around 80 µs – 95 µs were measured in samples processed at 400 °C and 450 °C. In this annealing temperature range, values of the τ_{eff} up to 125 µs were measured. Again, we can observe that the low values of the effective minority carrier lifetime occur in the center of the sample. Moreover, comparing Fig 3-c (τ_{eff} = 43 µs) to Fig. 7-c (τ_{eff} = 82 µs) and Fig. 7-d (τ_{eff} = 95 µs), we can notice that the average effective minority carrier lifetime may be increased if the annealing step is implemented.



(a) **(b)** **(c)** **(d)**

Fig. 7: The two-dimensional distribution of the minority carrier lifetimes afterwards the oxidation at temperature of 800 °C during 7 minutes and annealing step during 20 minutes and temperature of (a) 400 ºC (without oxidation) (τ_{eff} = 10 µs), (b) 350 ºC (τ_{eff} = 48 µs), (c) 400 ºC (τ_{eff} = 82 µs) and (d) 450 ºC (τ_{eff} = 95 µs).

4. Conclusion

The passivation of silicon solar cells with a thin SiO_2 layer were experimentally optimized and analyzed. The efficiency of 15.9 % was achieved in solar cells processed at the oxidation temperature of 800 ºC, during 7 minutes. The better average efficiencies were obtained in the oxidation temperature range from 750 °C to 800 °C, caused by the increasing of the open circuit voltage and fill factor. This result is related to the highest effective minority carrier lifetime, of 40 μs - 50 μs, measured in wafers processed in this oxidation temperature range. The short-circuit current was not affected by oxidation temperature, but the internal quantum efficiency decreases slightly with the increasing of the oxidation temperature. The minority carrier diffusion length of the solar cells processed with the oxidation temperature of 800 °C was around 1890 μm and decreased to 960 μm with the increasing of the oxidation temperature to 900 °C. The open circuit voltage shows a slight trend of increasing with the oxidation time, but the fill factor decreases. The annealing step in forming gas did not improve the average efficiency of solar cells and the open circuit voltage tends to grow with the increasing of the annealing time. The annealing temperature and time did not affect the short-circuit current density. The highest values of the average effective minority carrier lifetime of around 80 and 100 μs were measured in samples processed at annealing temperature of 400 °C and 450 °C, respectively. Solar cells processed with oxidation and annealing presented higher internal quantum efficiency at short wavelengths than cells with only annealing step.

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