

HIGH SPEED REGENERATION OF BO-DEFECTS: IMPROVING LONG-TERM SOLAR CELL PERFORMANCE WITHIN SECONDS

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ABSTRACT: Boron-oxygen related defects typically limit the efficiency of solar cells made from silicon containing high concentrations of boron as well as oxygen. The detrimental effect of these defects can be eliminated by applying a Regeneration procedure that needs carrier injection at slightly elevated temperatures. The kinetics of this process is influenced by different processing steps like thermal treatment and was found to rely on a high enough hydrogen concentration in the silicon bulk. It is shown here that neither emitter formation nor the use of $\text{Al}_2\text{O}_3/\text{SiN}_x:\text{H}$ or $\text{SiO}_2/\text{SiN}_x:\text{H}$ passivation stacks affect Regeneration in a fundamental way. By contrast, the thickness of a $\text{SiN}_x:\text{H}$ layer acting as hydrogen source during a high temperature firing step has direct influence on Regeneration confirming that better hydrogenation results in faster Regeneration reactions. Condensing different process steps that all accelerate Regeneration allows the application of a high-speed Regeneration process consisting of a combination of relatively high temperature and high carrier injection, resulting in complete Regeneration of BO defects in less than 10 s. This makes Regeneration feasible as an in-line process in solar cell production and thus solves the problem of the boron-oxygen defects. Even further acceleration is achieved by laser induced Regeneration where the substrate is heated and illuminated simultaneously.

Keywords: silicon, boron-oxygen, defects, regeneration, hydrogen

1 MOTIVATION

Solar cells made from boron doped oxygen-rich silicon are typically limited by the detrimental influence of so called boron-oxygen (BO) defects that become recombination active under carrier injection and drastically reduce minority carrier lifetime [1][2][3][4][5]. For PERC-type solar cells, the use of rather highly doped silicon is advantageous in order to fully exploit the concept's efficiency potential, hence boron-oxygen related degradation leads to efficiency losses of around 1.5%_{abs.} for 1 Ωcm boron-doped silicon and oxygen concentrations in the $5\text{-}10\cdot 10^{17}\text{ cm}^{-3}$ range which is a typical value in Czochralski (Cz) grown silicon. Thus, BO defects need to be eliminated to reach high efficiencies in p-type Cz silicon solar cells.

The degradation effect can be reversed by thermal treatment at $\sim 200^\circ\text{C}$ for several minutes in the dark. This causes an annealing reaction leading into the original recombination inactive high lifetime state. The annealed state being instable under carrier injection will result in renewed BO-related degradation. In 2006, the so called Regeneration reaction has been presented by Herguth et al. [6][7]. It occurs under carrier injection (illumination or external biasing of a solar cell) at slightly elevated temperatures (typically $60\text{-}200^\circ\text{C}$) and transforms boron-oxygen defects from the recombination active degraded state into the recombination inactive regenerated state. This state is characterized by high lifetimes and, most importantly, by being stable under solar cell operating conditions [8][9] (Fig. 1). During the last years, a lot of research has been done on understanding how the kinetics of the regeneration reaction depends on sample processing, material properties and regeneration parameters like temperature and illumination intensity.

In this paper, progress in all three fields will be presented resulting in an optimized regeneration process that is now feasible in inline solar cell production, so that the efficiency of boron-doped solar cells is not necessarily limited by the detrimental influence of boron-oxygen related defects anymore.

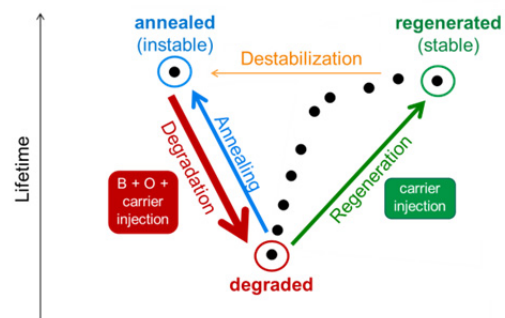


Figure 1: Definition of all transitions between the three states of the boron-oxygen defect. Whereas annealing and destabilization are purely thermally activated reactions, regeneration only occurs under carrier injection and the degradation reaction additionally requires the presence of boron and oxygen.

2 EXPERIMENTAL DETAILS

To compare the influence of different process steps, material features or regeneration parameters on the kinetics of the regeneration process, lifetime samples were made from 1.3 Ωcm boron doped Cz silicon wafers that were POCl_3 gettered and surface passivated using PECVD $\text{SiN}_x:\text{H}$ layers. Finally, all samples were shortly fired at high temperature. The so processed samples were then completely degraded (e.g. 40°C , 0.1 suns, 24 h) before the Regeneration process was applied at 130°C and ~ 1 sun illumination by a halogen lamp. The stability of the resulting defect state was always checked by illuminating the samples for ~ 24 h and finally all samples were annealed at 200°C for 10 min in the dark in order to be able to exclude any non BO related effects (Fig. 2).

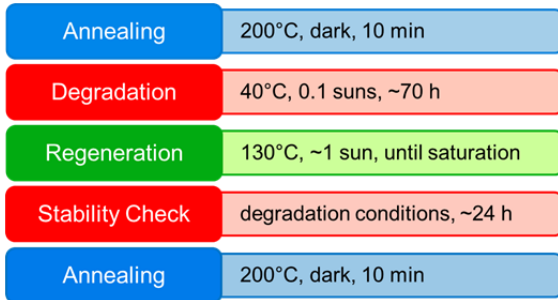


Figure 2: Course of one regeneration sequence.

After every step and several times during regeneration minority carrier lifetime was measured using a Sinton Lifetime Tester and the normalized defect concentration $N^*(t)$ was calculated using

$$N^*(t) = 1/\tau(t) - 1/\tau_{\text{annealed}} \quad (1)$$

with $\tau(t)$ being the minority carrier lifetime at time t and τ_{annealed} being the lifetime in the annealed state where it is not affected by BO defects. When plotted over Regeneration time, the evolution of $N^*(t)$ can be fitted using a single exponential function of which the characteristic time constant corresponds to the regeneration time constant. Its inverse gives the regeneration rate R_R that is used to compare the regeneration kinetics of different samples. Deviations from this scheme are mentioned separately in the respective sections.

3 INFLUENCE OF REGENERATION CONDITIONS

3.1 Temperature

The Regeneration reaction is a thermally activated process [6][10] meaning that it can be accelerated by increasing temperature. One has to keep in mind that the other transitions of the boron-oxygen defect (degradation, annealing and destabilization) are also enhanced at higher temperatures. Hence, temperature can only be increased to a certain extent if one wants the Regeneration reaction to be the dominant process. See [11] for details about interaction of the different transition paths.

3.2 Carrier injection

Carrier injection, typically realized by illumination, is necessary for Regeneration to occur with a higher carrier injection level resulting in higher Regeneration rates meaning a faster Regeneration process [8]. The carrier injection level depends on illumination intensity as well as on minority carrier lifetime of a silicon wafer. Hence, the injection level can only be considered to be the same in two samples if both illumination and lifetime are the same. This means that anything causing recombination within a wafer will result in decreased injection and thus in decreased Regeneration rates even though the illumination intensity is kept constant. This is crucial when analyzing the influence of a diffused emitter region as well as of different surface passivation stacks on Regeneration kinetics.

3.2.1 Influence of a diffused emitter layer

Two sets of parallel processed lifetime samples are made to investigate a possible influence of a diffused emitter layer on Regeneration. In comparison to the samples processed as described in section 2, the $50 \Omega/\square$ diffused layer is not removed for some of the samples.

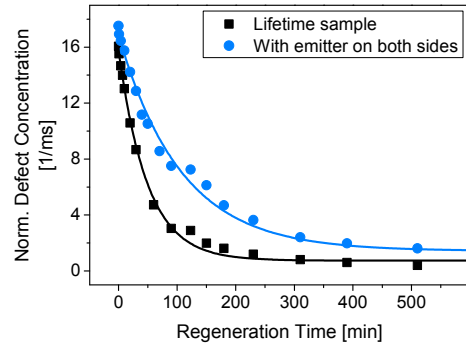


Figure 3: Evolution of the normalized defect concentration during Regeneration of a standard lifetime sample compared to a sample with diffused emitter layers on both sides.

The measurement of the Regeneration process results in the data shown in Fig. 3. The sample containing diffused emitter layers on both sides regenerates more slowly ($R_R = 0.6 \pm 0.2 \text{ h}^{-1}$) than the reference sample, in which the diffused layer have been removed ($R_R = 1.2 \pm 0.1 \text{ h}^{-1}$). But whereas the first sample has an effective minority carrier lifetime of $\sim 40 \mu\text{s}$ in the annealed BO state, the equivalent value is $\sim 200 \mu\text{s}$ in the reference sample. This means that recombination in the emitter region cannot be ignored and even though both samples were illuminated in parallel during Regeneration, the carrier injection level differed. If this is taken into consideration (see [12]), no difference in Regeneration rate can be found between the two samples anymore. Hence we find that a diffused region has no fundamental influence on the Regeneration of BO defects, unless reducing the injection level generated within a wafer by a given illumination intensity and thereby indirectly reducing the Regeneration rate.

3.2.2 Influence of surface passivation stacks

Surface passivation stacks like thin Al_2O_3 or SiO_2 interlayers covered by thick $\text{SiN}_x\text{:H}$ are typically used in PERC-type solar cells for optimizing surface passivation, hydrogen passivation of bulk defects and optical light trapping. Whereas thick interlayers were found to decelerate the BO Regeneration process [13][14], we investigate here the influence of such thin interlayers (8-14 nm Al_2O_3 and 5-10 nm thermal SiO_2 , respectively).

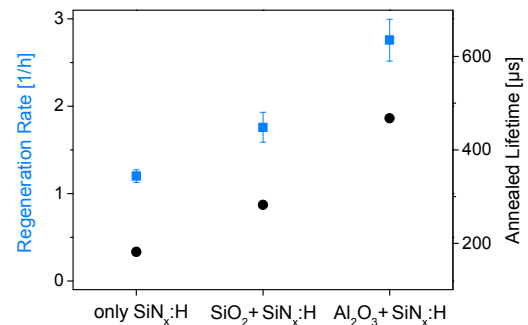


Figure 4: Regeneration rates of differently coated lifetime samples regenerated under the same conditions. Reduced recombination activity at the sample surface resulting in higher annealed lifetime values seems to enhance Regeneration.

No differences in Regeneration rate are found within the thickness ranges investigated for each layer system, but the Regeneration rates seem to increase with increasing surface passivation quality represented by the annealed lifetime values as given in Fig. 4. Hence, thin Al_2O_3 interlayers do not seem to have a fundamental influence on Regeneration but—in opposite to the diffused layer—they are able to reduce recombination in the silicon wafer and thus increase the carrier injection level generated by a certain illumination intensity. Therefore, they can indirectly have a positive effect on Regeneration kinetics. When a factor is applied that corrects for the differences in injection level [12], all three sample sets have the same Regeneration rate within measurement uncertainty. This suggests that hydrogenation can be assumed to be similar in all cases and all differences in Regeneration rate can be explained by the differences in injection level.

4 INFLUENCE OF HYDROGEN SOURCES

First studies on the question of why silicon samples regenerate with different regeneration rates even though temperature and carrier injection level are kept constant revealed that hydrogen could be of major importance for the regeneration process: whereas no regeneration has ever been shown in non-hydrogenated samples, the Regeneration effect is usually found in samples coated with hydrogenated surface passivation layers [15][16]. Hence, we use two different hydrogenation methods in order to separate a possible influence of the surface passivation layer itself from hydrogen related effects.

4.1 $\text{SiN}_x\text{:H}$ as hydrogen source layer

Comparing the Regeneration behavior of samples coated with $\text{SiN}_x\text{:H}$ layers of different thickness (90 nm and 210 nm, respectively) reveals increasing regeneration rates for increasing layer thickness. At the same time, the hydrogen diffusing from the layer towards the silicon bulk is increased for thicker $\text{SiN}_x\text{:H}$ layers as confirmed by NRA measurements of the hydrogen content within the first 80 nm of the silicon bulk [17], see Fig. 5.

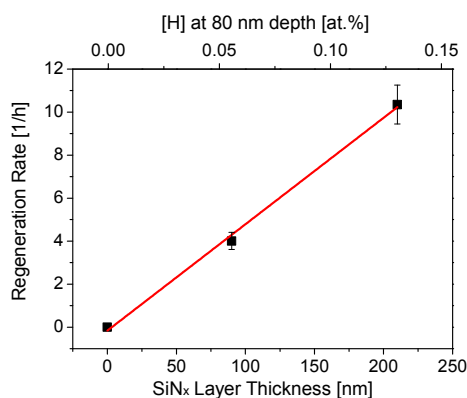


Figure 5: $\text{SiN}_x\text{:H}$ layers of different thickness introduce different amounts of hydrogen into the silicon bulk. The samples containing more hydrogen due to thicker $\text{SiN}_x\text{:H}$ layers also show higher regeneration rates.

Even though the thicker layers also result in slightly better surface passivation, the difference in Regeneration cannot be explained by the layers influencing the injection level: After the Regeneration rates have been corrected for any injection level influence [12], the rates still differ considerably (Fig. 5 shows the corrected Regeneration rates) The reason for the acceleration of the Regeneration reaction could thus rather be the thicker layer providing more hydrogen. Thus, increasing regeneration rates seem to be correlated with increasing hydrogen content of the silicon bulk.

4.2 Direct hydrogenation by remote H plasma

Using hydrogenation of silicon wafers by remote hydrogen plasma offers the possibility of decoupling a possible hydrogen effect from the influence of the surface coating layer. Cyclic hydrogenation was applied to bare wafers as presented by Stutzmann et al. [18]. With every step, the hydrogen content in the silicon wafer is increased. This was confirmed by SIMS measurements on deuterated samples [19]. The wafer surfaces were passivated chemically using a quinhydrone-methanol solution and minority carrier lifetime was measured after annealing, degradation and 20 min Regeneration. The data presented in Fig. 6 reveal that the more hydrogenation steps a sample has seen, the more the lifetime measured after 20 min Regeneration approaches the value measured in the annealed state: Whereas the lifetime value after the Regeneration treatment equals the lifetime after degradation for the non-hydrogenated sample, complete Regeneration is achieved in the samples that had seen 2 hydrogenation steps. In the samples hydrogenated by one hydrogenation step, lifetime after 20 min Regeneration was somewhere between the degraded and the annealed state suggesting incomplete Regeneration within the given timespan. Apart from the Regeneration effect, the data also show an increased quality of the non BO-related background due to the hydrogenation steps as revealed by increasing lifetime values in the annealed and in the degraded state with increasing number of hydrogenation steps.

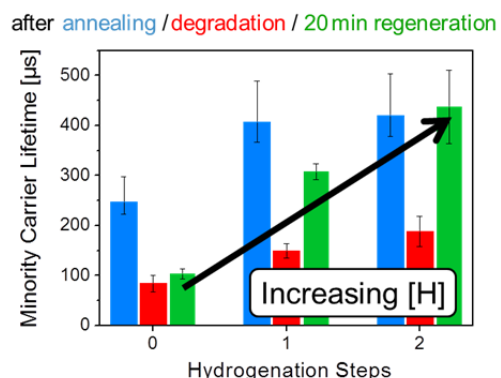


Figure 6: Silicon wafers hydrogenated by 0, 1, or 2 hydrogen plasma steps, respectively. Lifetime was measured after annealing, after degradation and after 20 min Regeneration revealing no Regeneration in the non-hydrogenated samples, partly Regeneration after one hydrogenation step and complete Regeneration after 2 hydrogenation steps. Data taken from [13].

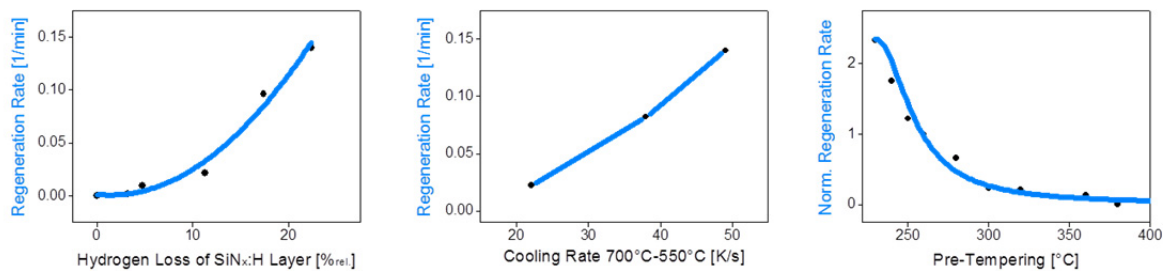


Figure 7: Influence of a hydrogenated surface coating releasing hydrogen during a short high temperature firing step (left), of the cooling phase after firing (middle) and of mid-temperature steps applied prior to Regeneration (right). Regeneration rate was found to be reduced if only little hydrogen is provided by the coating layer (left, [20]), if the sample is cooled down slowly from high temperatures (middle, [21]) or if a pre-tempering step at temperatures approaching 400°C is applied (right, [9]).

This investigation showing the same tendency as samples coated with SiN_x:H, namely better hydrogenation of the silicon bulk resulting in enhanced Regeneration of BO defects, makes us suggest that Regeneration requires a high enough hydrogen concentration in the silicon bulk— independent of the surface passivation or hydrogenation method. These results and the measurement method are described in detail in [13].

5 INFLUENCE OF TEMPERATURE STEPS

Different temperature steps applied to silicon wafers coated with hydrogenated surface passivation layers were found to have major influence on the Regeneration of BO defects. Short high temperature firing steps that are known to make hydrogen diffuse from a SiN_x:H layer into the silicon bulk were found to result in Regeneration rates depending on the actual peak temperature during firing [20]. Increasing Regeneration rates could be correlated with the coating layer releasing more hydrogen during firing (Fig. 7, left). This confirms the hypothesis that hydrogen might be necessary for the Regeneration reaction to occur.

An equally important effect was shown for the cooling process after high temperature firing where high cooling rates allow high Regeneration rates ([21], confirmed by Walter et al. [22]) (Fig. 7, middle). Different hypotheses have been suggested to explain those results including high cooling rates causing high net diffusion of hydrogen from a SiN_x:H layer into the Si bulk [21], advantageous hydrogen binding configurations after cool-down [9] or the creation of nano-precipitates that might influence Regeneration kinetics in a non-hydrogen based mechanism [22].

In addition to those high-temperature related effects, process steps approaching 400°C (applied to fired samples) were also found to affect Regeneration [9]. The influence of such temperature steps on the creation of hydrogen binding configurations that might be unfavorable during Regeneration has been suggested as an explanation: if a considerable amount of hydrogen is bound in a way that it cannot be released under Regeneration conditions (60-200°C + carrier injection), the effective concentration of mobile hydrogen during Regeneration might be reduced. This should result in lower Regeneration rates if mobile hydrogen is somehow involved in the Regeneration process.

6 INFLUENCE OF MATERIAL PROPERTIES

Apart from sample processing, the silicon material itself can also influence Regeneration kinetics by the impurities in usually contains.

6.1 Doping impurities

Studies on p-type silicon compensated with boron, gallium and phosphorus showed that Regeneration is considerably slowed down in these materials. This could be ascribed to a negative influence of the acceptor atoms boron [23] and gallium [24]. Even in compensated n-type silicon, the transformation of BO defects into a stable regenerated state was shown to be successful [24]. The fact that both boron as well as gallium are known to trap hydrogen was suggested as an explanation: Both atoms could either trap hydrogen permanently and thus reduce the concentration of mobile hydrogen in the silicon bulk or they could act as temporary traps reducing hydrogen mobility. If mobile hydrogen is needed during Regeneration, both effects could explain the reduced Regeneration rates observed in differently doped material. See [24] for details.

6.2 Other impurities

In addition to the doping impurities that are deliberately added to silicon used for solar applications, relatively high concentrations of interstitial oxygen O_i are typically found in Cz grown silicon. We can confirm the observation by Lim et al. [25] who have shown that the Regeneration rate increases with decreasing interstitial oxygen concentration. We suggest hydrogen interacting with O_i to be a possible reason for that. In material containing relatively high concentrations of metal impurities that can also trap hydrogen in silicon, a beneficial effect of a POCl₃ gettering step can be expected as it has been shown indeed by Lim et al. [26].

7 HYDROGEN BASED REGENERATION MODEL

The many different experiments reviewed and presented here all lead to the conclusion that hydrogen is a necessary condition for the Regeneration effect to occur and better hydrogenation was found to result in faster Regeneration processes. Using hydrogenated surface passivation layers in combination with short high temperature firing steps was found to be a suitable way of

introducing enough hydrogen into the silicon bulk. The bulk itself also seems to have an influence, either caused by different types of possibly hydrogen trapping impurities or due to temperature steps that might cause a specific distribution of hydrogen within the silicon lattice. Altogether, this brings us to the hypothesis that enough mobile hydrogen is needed during Regeneration, meaning at slightly elevated temperatures and under carrier injection. If this is fulfilled, passivation of recombination active BO defects by mobile hydrogen could cause the lifetime recovery that characterizes the Regeneration process.

Different reasons are thinkable why higher temperatures are beneficial and why carrier injection is needed. As hydrogen is typically bound to impurities, sites of lattice imperfection or to other hydrogen atoms at low temperature [27], slightly elevated temperatures might be needed to free hydrogen from those bindings. In certain important cases, e.g., when hydrogen is bound to acceptor atoms, this process might be assisted by carrier injection because this could enable changes in the charge states of any of the participating species. In sum, the hypothesis saying that the Regeneration reaction is actually hydrogen passivation of BO defects can explain all experimental results known until now and opens the possibility of predicting how different steps during solar cell processing would probably affect Regeneration kinetics. A detailed discussion of the hydrogen-based Regeneration model can be found in [9].

8 HIGH-SPEED REGENERATION PROCESSES

8.1 High-speed Regeneration

The knowledge about what has to be considered in order to manufacture well regenerating samples enables us to push the limits of the established Regeneration process towards a high-speed Regeneration process. As mentioned in section 3, increasing temperature as well as illumination accelerates the Regeneration reaction. Using well hydrogenated samples due to suitable firing after $\text{SiN}_x\text{:H}$ deposition then results in Regeneration time constants around 1.5 s at 230°C and 2.7 suns illumination. This means that 99% of all BO defects can be regenerated in less than 10 s making this a feasible in-line process in solar cell production. All conditions that need to be fulfilled to make this work are already part of standard solar cell processes, so that usually no changes in the production line are needed before the application of a high-speed regeneration process. This process is patent pending and described in detail in [9].

Please note that the Regeneration process can only be accelerated by increasing the Regeneration temperature until a certain limit is reached. This limit might depend on the specific solar cell process and is typically at around 230°C. Higher temperatures will result in incomplete Regeneration with the completeness decreasing drastically when 400°C is approached. Fig. 8 depicts the temperature dependence of the completeness of the Regeneration reaction for lifetime samples processed analogously to standard solar cells when an intensity of 2.7 suns is used during Regeneration.

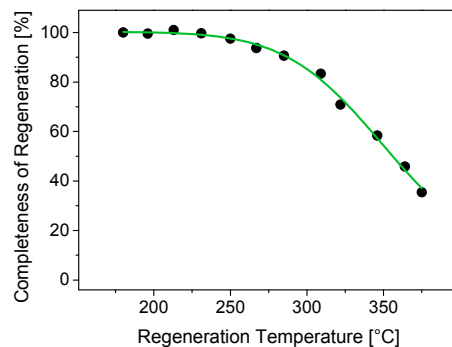


Figure 8: Maximum completeness of the Regeneration reaction at high Regeneration temperatures measured at 2.7 suns illumination. Data taken from [9].

8.2 Laser induced Regeneration

Using a high-intensity light source like a laser heating the sample to a suitable temperature while illuminating it, gives the possibility of further accelerating the Regeneration procedure. For proving this concept, we used a 1064 nm ns laser, scanning a lifetime sample in lines. The lines were illuminated several times in order to monitor the evolution of minority carrier lifetime during the process. Lifetime changes were measured using photoluminescence, and the illumination time is calculated for every line from the scanning velocity of the laser beam. The laser process was applied after complete BO degradation. Fig. 9 shows that an increasing laser illumination time results in increasing PL intensity. The resulting lifetime values were stable under further illumination at low temperature (0.2 suns halogen lamp at 40°C for 24 h) meaning that BO defects have been regenerated successfully.

Transforming the measured PL intensity values into normalized defect concentration gives the typical exponential decay when plotted over laser illumination time (Fig. 10). The Regeneration time constant is at around 0.1 s so that saturation is reached at very low defect concentration within 0.5 s.

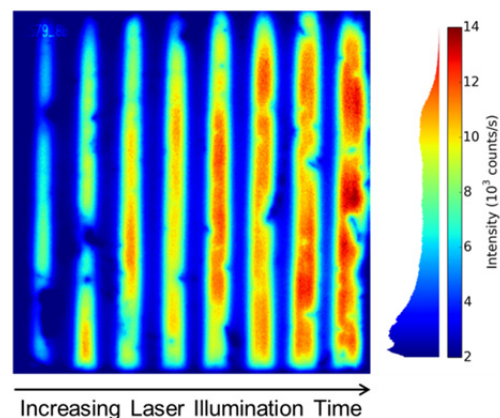


Figure 9: Photoluminescence image of a lifetime sample after laser induced Regeneration process. The BO degraded sample is illuminated and simultaneously heated by laser scanning in vertical lines. The number of line scans and thus the accumulated laser illumination time increases from left to right.

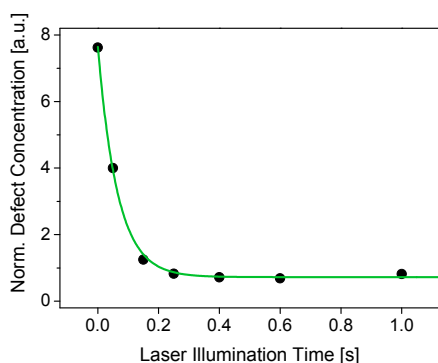


Figure 10: Evolution of the normalized defect concentration during laser induced Regeneration process. Defect concentration saturates at a very low level in less than 0.5 s.

Some optimization concerning heat and carrier injection generated by the laser beam is needed in order to bring the final BO defect concentration down to zero meaning to achieve complete Regeneration within less than 1 s. Details of this laser induced Regeneration process will be published elsewhere.

9 SUMMARY

The influence of different cell processing steps, e.g., the use of different passivation layer systems as well as thermal steps has been investigated and revised. We found that the effect a diffused emitter layer as well as $\text{Al}_2\text{O}_3/\text{SiN}_x\text{:H}$ or $\text{SiO}_2/\text{SiN}_x\text{:H}$ passivation stacks have on Regeneration kinetics could be ascribed to those layers affecting the carrier injection level at constant external illumination. This is true as long as all passivation stacks enable comparable hydrogenation of the silicon bulk. This was in contrary to $\text{SiN}_x\text{:H}$ layers of different thickness providing different amounts of hydrogen during a high temperature firing step, and therefore directly determining the Regeneration rate. Regeneration kinetics depending on the degree of hydrogenation of the silicon bulk could be confirmed on samples hydrogenated by remote hydrogen plasma where enhanced Regeneration was found to correlate with increasing hydrogen content. Different temperature steps were reported to influence Regeneration kinetics as well as do intrinsic material properties like impurity concentrations. Merging all those results leads to our hypothesis that Regeneration of BO defects is actually passivation of recombination active BO defects by mobile hydrogen. It is suggested that this process needs slightly elevated temperatures in combination with carrier injection so that enough mobile hydrogen can be provided during Regeneration.

Hence, 5 criteria should be fulfilled in order to manufacture well regenerating samples:

1. Introduce a lot of H into the sample.
2. Avoid temperature steps $\sim 400^\circ\text{C}$ and slow cooling rates after firing.
3. Avoid H traps (e.g., impurities) that reduce H mobility.
4. Apply carrier injection (e.g. illumination)
5. Choose the Regeneration temperature as high as possible, but avoid incomplete Regeneration.

The progress in developing well regenerating silicon

samples allowed us to design a high-speed Regeneration process consisting of a combination of relatively high temperatures in the range of $200\text{-}230^\circ\text{C}$ and high illumination intensity of ~ 2.7 suns. This results in very fast Regeneration processes that can be completed in less than 10 s making Regeneration feasible as an in-line process in solar cell production. Hence, p-type solar cells made from Cz material are not limited by the detrimental influence of boron-oxygen defects anymore. Finally, we presented for the first time a laser induced Regeneration process that is able to simultaneously heat and illuminate the sample to regenerate BO defects in less than 0.5 s. Hence, the presented procedure proves that the time limit is not yet reached and ultra-fast Regeneration processes are realizable by different technological approaches.

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