Al-Enhanced PECVD SiN_x Induced Hydrogen Passivation in String Ribbon Silicon

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The effectiveness of manufacturable gettering and passivation technologies is investigated for their ability to improve the quality of a promising Si photovoltaic material. The results of this study indicate that a lifetime enhancement of 30 μs is attained when a backside screen-printed aluminum layer and a thin film of SiN_x, applied by plasma-enhanced chemical vapor deposition (PECVD), are simultaneously annealed at 850°C in a lamp-heated belt furnace. Based on the results of this study, a model is proposed to describe the Al-enhanced SiN_x induced hydrogen defect passivation in String Ribbon silicon due to the simultaneous anneal. According to this model, three factors play an important role: i) the release of hydrogen from the SiN_x film into the substrate; ii) the retention of hydrogen at defect sites in silicon; and iii) the generation of vacancies at the Al-Si interface due to the alloying process which increases the incorporation of hydrogen and creates a chemical potential gradient which enhances the migration of hydrogen in the substrate. A PC1D device simulation indicates that screen-printed cell efficiencies approaching 16% can be achieved if the gettering and passivation treatments examined in this study are employed, the substrate thickness is reduced, and a high-quality surface passivation scheme is applied.

Key words: Multicrystalline, ribbon silicon, silicon nitride, hydrogen passivation, defects

INTRODUCTION

While large area high efficiency silicon solar cells have been developed using high quality silicon substrates and long process sequences, the remaining challenge for silicon photovoltaics is to reduce the cost of module fabrication while maintaining an efficiency level that can reduce module cost by a factor of 2–4. An obvious approach towards reducing the cost of silicon photovoltaic modules is to reduce the cost of the silicon substrate, which accounts for 40–55% of the current module cost.[1,2] The PC1D simulation of the effect of bulk lifetime (τ) and back surface recombination velocity (BSRV) in Fig. 1 shows that high efficiency screen-printed solar cells can be fabricated by using relatively low-quality, but thin substrates (100 μm), if a high quality surface passivation treatment is applied. The important PC1D inputs for this simulation are given in Table I. The String Ribbon silicon growth process can reduce the cost of substrate growth because ribbon substrates for solar cells are grown directly from the melt, eliminating the losses associated with slicing and subsequent etching.[3] Another advantage of String Ribbon silicon is that it can be grown to a thickness below 50 μm,[4] resulting in further material and cost savings. While the growth of String Ribbon silicon makes it an attractive material for low-cost silicon photovoltaics, the as-grown minority carrier lifetime in the material is typically 1–10 μs. The results of the simulation in Fig. 1 clearly indicate that, for a 100 μm device, the as-grown lifetime in String Ribbon silicon of 1–10 μs is not suitable for high efficiency (>15%) screen-printed solar cells, and there is only a small impact of back surface passivation. However, the simulation indicates that the cell efficiency will increase sharply as

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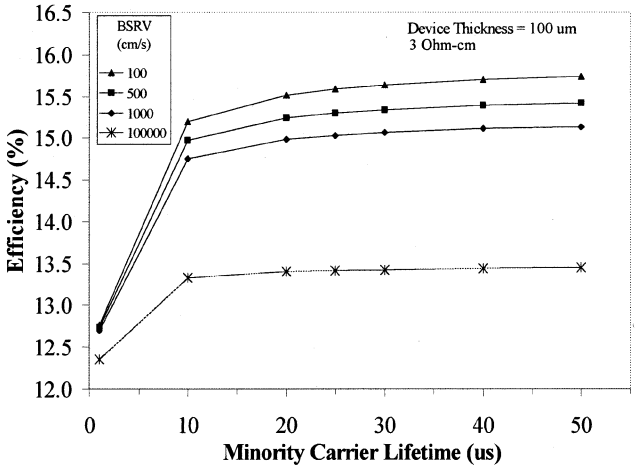

Fig. 1. PC1D simulation of the effect of τ and BSRV on efficiency of string ribbon Si solar cells.

the lifetime increases to 20 μs. For bulk lifetimes greater than 20 μs, improvements in the BSRV are more important than further bulk lifetime improvements in increasing the device efficiency. If the bulk lifetime exceeds 30 μs, high efficiency (>15%) devices can be fabricated even if the BSRV is as high as 1000 cm/s. Therefore, to fabricate high efficiency screen-printed devices on thin String Ribbon silicon we must understand and develop impurity gettering and defect passivation techniques that can improve the minority carrier lifetime in the material without significantly raising the cost.

Several gettering and passivation techniques have been examined for the improvement of silicon photo voltaic (PV) materials including phosphorus and Al gettering and hydrogen passivation. Plasma-enhanced chemical vapor deposition (PECVD) SiN_x films deposited at 200–300°C have a hydrogen concentration between 1.3×10^{22} and 2.0×10^{22} cm^{-3},[5] and have been shown to provide efficient bulk and surface hydrogen passivation when annealed.[6,7] Many authors claim that the release of hydrogen from the PECVD SiN_x film into the silicon substrate and subsequent passivation of bulk defects during thermal anneal is responsible for enhancement in multicrystalline Si (mc-Si) solar cell performance.[7-12] Some authors[9,10] reported the use of a screen-printed Al layer on the back surface of cells which is annealed simultaneously with the PECVD SiN_x film to form a back contact and an Al-back surface field (Al-BSF) while other authors[11,12] have not clearly stated the composition of the device back contact. While there have been many reports of hydrogen passivation from the post-deposition anneal of PECVD SiN_x films in conjunction with Al processing, there has been no effort to isolate the role of Al, if any, in the hydrogen passivation process.

The aim of this study is to evaluate the effectiveness of PECVD SiN_x hydrogenation individually and in combination with phosphorus and Al gettering in String Ribbon silicon. A combination of Fourier transform infrared spectroscopy (FTIR) measurements of PECVD SiN_x films and quasi-steady state photoconductance (QSSPC) bulk minority carrier lifetime measurements is used to improve the fundamental understanding of the hydrogenation process. Finally, a model is proposed which relates the high temperature (850°C) PECVD SiN_x induced hydrogenation of defects in String Ribbon silicon to the release of hydrogen into the substrate, the retention of hydrogen at defect sites at high temperatures, and the injection of vacancies from backside Al alloying.

Table I. Inputs for PC1D String Ribbon Silicon Solar Cell Performance Simulation

Device Parameter	PC1D Input
thickness	100 μm
N	3 Ω-cm, p-type
FSRV	5×10^5 cm/s
Front surface reflectance	PECVD SiN single layer ARC
Rear internal reflectance	70% diffuse
R	0.3 Ω-cm²
Rsh	14925 Ω-cm²
Jo2	154 nA/cm²
n2	2.15
Et	0.0 eV

EXPERIMENT

Sample Cleaning

String Ribbon substrates, grown by *Evergreen Solar* with a thickness of 300 μm and base resistivity of 3 Ω-cm, and 300 μm thick, 1 Ω-cm float zone silicon wafers were cleaned using the following sequence before processing:

1. Rinse in DI H_2O for 5 min
2. HF:H_20 (1:10) for 1:30 min
3. Rinse in H_20 for 3 min
4. H_2O:H_2SO_4:H_2O_2 (2:1:1) for 5 min
5. Rinse in H_2O for 3 min
6. HNO_3:CH_3COOH:HF (15:5:2) for 3 min (String Ribbon only)
7. Rinse in H_2O for 3 min
8. H_2O:HCl:H_2O_2 (2:1:1) for 5 min
9. Rinse in H_2O for 3 min
10. HF:H_2O (1:10) for 3 min
11. Rinse in H_2O 30 sec

Thick String Ribbon substrates were used in this study to avoid breakage of samples during the rough cleaning and etching steps for lifetime measurements.

Phosphorus Gettering Treatment

A commercially available phosphorus spin-on dopant film was applied to the front surface of selected String Ribbon substrates. Substrates were annealed in a lamp-heated belt furnace for 6 minutes in air at a set-point temperature of 925°C to obtain the target sheet resistance of 45 Ω/sq, which is desirable for screen-printed solar cells. After diffusion, the phosphorus-doped glass layer was etched in dilute hydrofluoric acid (HF) and substrates were again cleaned. Multiple samples were processed and the n^+ layer was removed from one sample before the life-

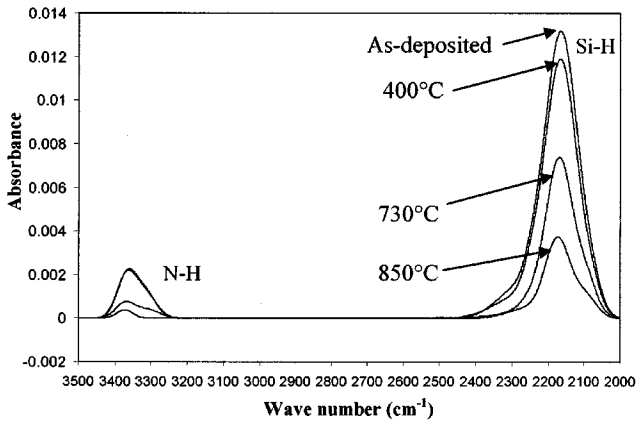

Fig. 2. FTIR spectrum of PECVD SiN_x films after post-deposition anneal.

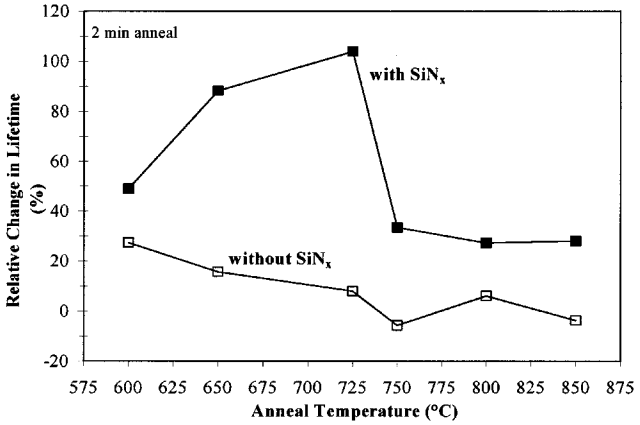

Fig. 3. Defect passivation from post-deposition anneal of PECVD SiN_x.

time measurement.

PECVD SiN_x Deposition and Anneal for Hydrogen Bulk Defect Passivation

SiN_x films were deposited onto String Ribbon substrates for bulk defect hydrogenation and float zone wafers for the analysis of the hydrogen content in PECVD SiN_x films. SiN_x films were deposited at 300°C using a direct, parallel plate PECVD reactor operating at 13.56 MHz with a flow rate of 320 and 1.55 sccm for SiH_4 (2% in N_2) and NH_3, respectively. The SiN_x films had a thickness of 860 Å and a refractive index of 1.94. SiN_x was deposited on both surfaces of the String Ribbon samples. Samples were annealed to drive hydrogen from the film into the bulk of the substrates and evaluate the hydrogenation from PECVD SiN_x. Anneals were performed in a belt furnace for two minutes in air at set-point temperatures in the range of 650–850°C and the films were removed using a dilute HF solution for subsequent lifetime measurement. In a selected instance, SiN_x was deposited only on the front surface of a String Ribbon sample. This sample was reserved for the Al gettering treatment, described below, to study the effect of simultaneous SiN_x hydrogenation and Al alloying. In this case the substrate was not annealed after deposition of SiN_x.

Al Gettering Treatment

A thick film of Al was applied to the back surface of String Ribbon substrates by screen-printing a commercially available Al paste. Then substrates were annealed in a belt furnace for two minutes in air at a set-point temperature of 850°C. The Al layer and underlying p^+- doped layer were then removed.

FTIR Measurements

FTIR measurements were performed on PECVD SiN_x coated, chemically polished 1 Ω-cm, float zone silicon wafers. After SiN_x deposition, described above, selected samples were annealed in a tube furnace at 400°C in forming gas (10% H_2 in N_2) and at 730°C and 850°C in air in a belt furnace. Room temperature FTIR measurements and analysis was performed on each sample after the heat treatments.

Minority Carrier Lifetime Measurements

All substrates were cleaned prior to the post-process lifetime measurement. Lifetime measurements were made using the QSSPC technique[13] with samples immersed in an I_2/methanol solution that has been shown to effectively passivate silicon surfaces.[14] Lifetime values were recorded at an injection level of 1×10^{15} cm^{-3} to avoid recording erroneously high recombination lifetimes at lower injection levels caused by shallow traps.[15] Four lifetime measurements were made on each ~4 in^2 sample and the average lifetime value was used to characterize the entire substrate. After the initial lifetime measurement, substrates were again cleaned and subjected to one or a combination of the above gettering or passivation treatments. All of the gettering and passivation heat treatments in this study were performed in a lamp heated, continuous belt furnace.

RESULTS AND DISCUSSION

Temperature Dependence of Hydrogen Release from PECVD SiN_x and Corresponding Bulk Defect Passivation

The FTIR spectra in Fig. 2 show that upon post-deposition anneal of the PECVD SiN_x film, the bonded hydrogen content in the film, proportional to the total area under the Si-H and N-H peaks, decreases as the anneal temperature is increased. The total bonded hydrogen content in the as-deposited PECVD SiN_x film is 2.7×10^{22} cm^{-3} and decreases by a factor of 4.3, 2.1, and 1.2 for the 850°C, 730°C, and 400°C anneals respectively,[16] indicating that more hydrogen is released from the film as the anneal temperature is increased. If the degree of defect passivation depends only on the release of hydrogen from the PECVD SiN_x film, one would expect the passivation effect to increase as the anneal temperature is increased. To establish the effect of an increased release of hydrogen from the PECVD SiN_x film on defect passivation, the lifetime of String Ribbon substrates was mea-

sured after post-deposition anneal at temperatures in the range of 600–850°C. Figure 3 illustrates the effect of the post-deposition anneal temperature on the bulk passivation in String Ribbon silicon. Because there is some variability in the as-grown lifetime in various String Ribbon samples, the average of the lifetime measurements in four different regions of each sample, measured before and after the gettering and passivation treatment, is used to evaluate the effectiveness of the gettering and passivation treatments in this study. Due to the variability in the as-grown lifetime of String Ribbon samples, the relative change in lifetime ($\tau_{final} - \tau_{as\text{-}grown}/\tau_{as\text{-}grown} \times 100$) that results from PECVD SiN_x induced hydrogenation is plotted in Fig. 3 as a function of the post-deposition anneal temperature. For all anneal temperatures investigated, the relative change in lifetime for samples annealed without SiN_x is small with respect to the change in lifetime for samples annealed with SiN_x. Therefore, the difference between the two curves in Fig. 3 is attributed to SiN_x induced bulk defect hydrogen passivation. The lifetime measurements in Fig. 3 indicate that with SiN_x, defect passivation increases as the post-deposition anneal temperature approaches 725°C commensurate with the decrease in the bonded hydrogen content in the PECVD SiN_x film with temperature shown in Fig. 2. This result supports the hypothesis that the degree of hydrogen passivation is dependent on the amount of hydrogen released from the PECVD SiN_x film.

Competition Between the Release of Hydrogen from the SiN_x Film and the Retention of Hydrogen at Defects

Figure 3 shows that at anneal temperatures between 600°C and 725°C, the relative improvement in lifetime is greater than 50%, while in the temperature range of 750–850°C, the relative improvement drops to near 30%. It should be noted that the degree of defect passivation from the anneal of PECVD SiN_x films, without prior Al or phosphorus treatments, is highly material dependent due to the variety of defects in different silicon PV materials.[17,18] The dramatic decrease in the effectiveness of hydrogen passivation above 725°C shown in Fig. 3 may be due to the high temperature instability of hydrogen at defect sites in silicon. While hydrogen is known to diffuse out of silicon above 500°C during prolonged anneals,[19] this result suggests that defect passivation from SiN_x induced hydrogenation may be more generally dependent on the thermal budget of the post-deposition anneal. Other investigators have observed a dependence of the hydrogen passivation of silicon surfaces from a rapid thermal oxide (RTO)/SiN_x stack on the thermal budget of post-deposition anneals.[16] These results suggest that the hydrogen passivation of silicon defects may be proportional to the release of hydrogen from the PECVD SiN_x film as well as the retention of hydrogen at defect sites in silicon.

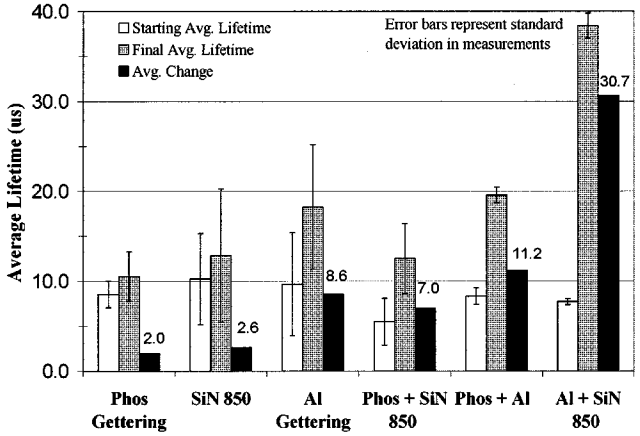

Fig. 4. Effectiveness of gettering and passivation treatments.

Al-Enhanced SiN_x Induced Hydrogenation of Defects

Although Fig. 3 shows that SiN_x induced hydrogenation of defects can improve the lifetime in String Ribbon silicon by as much as 100%, the final lifetime in the substrates does not exceeded 30 μs, which is required for high efficiency devices with BSRV of less than 1000 cm/s (Fig. 1). To further enhance the lifetime of String Ribbon silicon and investigate the interaction of the hydrogenation process with phosphorus and Al gettering, we examined the combinations of phosphorus and Al gettering with PECVD SiN_x hydrogenation using a 850°C post-deposition anneal. Though Fig. 3 indicates that there is little bulk passivation from the PECVD SiN_x post-deposition at 850°C, this anneal temperature was chosen so that Al gettering and the formation of a high quality Al-BSF occur simultaneously along with hydrogenation. The results of the gettering and passivation treatments shown in Fig. 4 illustrate that phosphorus gettering, PECVD SiN_x hydrogenation, and Al gettering are each moderately effective in improving the bulk lifetime but are unable to improve the measured lifetime to over 20 μs. The combination of phosphorus gettering and PECVD SiN_x hydrogenation at 850°C improves the lifetime by 7 μs, which is nearly equal to the sum of the enhancement provided by individual phosphorus gettering and hydrogenation treatments. A similar additive effect is observed in the combination of phosphorus and Al gettering in which the lifetime improved by 11 μs. Still the 30 μs threshold is not exceeded by any of the above combinations. In contrast, a noteworthy average lifetime of 38 μs, an improvement of 30 μs, is observed when the PECVD SiN_x hydrogenation treatment and Al gettering treatment are combined in one heat treatment at 850°C for 2 min. This improvement in lifetime is far greater than the sum of the 850°C hydrogenation and Al treatments alone suggesting that there may be a positive synergistic interaction between the hydrogenation from the front surface and the Al alloying process simultaneously occurring at the back surface of the substrate.

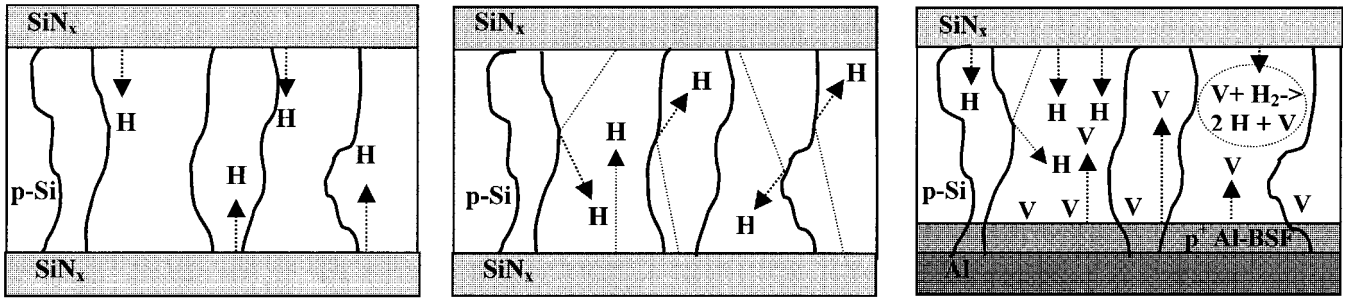

Fig. 5. A schematic representation of the three-step model for SiN_x induced defect passivation: (a) release of hydrogen during SiN_x anneal, (b) decrease in the retention probability of hydrogen at defects during high temperature SiN_x anneal, and (c) vacancy induced dissociation of molecular hydrogen and enhanced migration of atomic hydrogen.

Proposed Model for the Al-Enhanced Hydrogenation of Defects

This gettering and passivation study indicates that PECVD SiN_x induced hydrogenation of String Ribbon silicon is most effective when it occurs simultaneously with backside Al alloying. A three step physical model describing the Al alloying aided hydrogenation is proposed based on the above results and recent theoretical calculations of the interaction of hydrogen and vacancies in silicon and is illustrated in Fig. 5. The results in Figs. 2 and 3 show that hydrogen is released from the SiN_x film and passivates defects in String Ribbon silicon during a post-deposition anneal at temperatures below 725°C when anneal time is 2 min, as depicted in Fig. 5a. The data in Fig. 3 indicate that during higher temperature anneals (>725°C), the ability of hydrogen to stick to defect sites in silicon decreases, as represented in Fig. 5b, and the SiN_x induced passivation is significantly reduced. In contrast, as shown in Fig. 5c, vacancies generated at the back surface during Al-Si alloying can promote hydrogenation of defects by enhancing the dissociation of hydrogen molecules and the migration of atomic hydrogen deep into the bulk Si. During the Al-Si alloying, Si is dissolved in the Al melt, generating voids and vacancies in Si,[20] the latter of which can rapidly diffuse through silicon. These vacancies are now available to participate in the dissociation of molecular hydrogen into rapidly diffusing atomic hydrogen. In a perfect Si lattice, hydrogen is proportioned nearly equally between the molecular and atomic species at high temperatures (>626°C).[21] Estreicher's recent *ab initio* tight binding molecular dynamics calculations have shown that the H_2 molecule is stable in a perfect Si lattice, but disassociates into atomic hydrogen at high temperature (>500 K) in the presence of vacancies or interstitials.[22] Atomic hydrogen is more mobile in silicon than the molecular species by a factor of 10^{10} to 10^{12} at 850°C.[21] Even though grain boundaries may assist in the diffusion of hydrogen,[23] hydrogen may not be retained at grain boundaries during prolonged annealing at high temperatures. Theoretical calculations have shown that the binding energy for hydrogen to vacancies (V_n^0) is high, 3 to 3.5 eV greater than the binding energy for hydrogen at the bond center.[24] This affinity of hydrogen to vacancies can increase the flux of hydrogen in silicon. The flux of hydrogen atoms can be described in terms of a chemical potential as shown in the equation below.[25]

$$J_H = -M_H C_H \frac{\partial \mu_H}{\partial x}$$

where J_H is the flux of hydrogen into the wafer, M_H is the atomic mobility of hydrogen, C_H is the concentration of hydrogen, and μ_H is the chemical potential of hydrogen in the wafer.

In the absence of vacancies, the chemical potential gradient is dictated by the concentration gradient. In the presence of vacancies, the attraction of hydrogen to vacancies at the backside of the wafer increases the chemical potential gradient in the above equation, increasing the flux of hydrogen in the silicon. In this model backside Al alloying generates vacancies, which enhance the dissociation of molecular hydrogen into mobile atomic hydrogen and increase the driving force for the migration of atomic hydrogen in the material, enabling and promoting the passivation of defects deep in the material. While Fig. 3 shows that passivation cannot be attained during the post-deposition anneal of SiN_x films at 850°C, Fig. 4 illustrates that when Al alloying and hydrogenation are performed simultaneously at 850°C, a synergistic effect results in a significant lifetime enhancement of about 400%. At high temperatures, even if the retention of hydrogen at defect sites decreases, we are able to obtain significant passivation because, as shown in Fig. 5, the passivation is proportional to the release of hydrogen from the SiN_x film, the retention of hydrogen at defect sites, and the concentration of vacancies generated at the back surface.

CONCLUSIONS

This paper shows that commercially viable gettering and hydrogen passivation treatments investigated in this study are effective in improving the bulk quality of String Ribbon silicon substrates. A PC1D simulation indicates that the lifetime improvement of 30 μs achieved in this study should be sufficient for the fabrication of low-cost, high efficiency (>15%) thin,

screen-printed solar cells. The results of this study also indicate that hydrogen defect passivation from PECVD SiN_x during a high -temperature post-deposition anneals depends on three processes: i) the release of hydrogen from the PECVD SiN_x film; ii) the retention of hydrogen at defect sites in silicon; and iii) the generation of vacancies at the back surface of the wafer. This conclusion is based on the observed decrease in bonded hydrogen content in PECVD SiN_x films with increasing anneal temperature, decrease in the passivation effect from the anneal of PECVD SiN_x films above 725°C, and the synergistic interaction of PECVD SiN_x hydrogenation and backside Al alloying. As a result of the generation of vacancies by Al alloying, the combination of hydrogenation and Al gettering is effective in improving the lifetime in String Ribbon silicon beyond 30 μs even though the retention of hydrogen to defects in silicon is low at high temperatures. The *PC1D* simulation indicates that the combination of a bulk lifetime of over 30 μs and a BSRV of less than 300 cm/s can result in cell efficiencies approaching 16% using 100 μm thick silicon.

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