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Damp-Heat-Stable, High-Efficiency, Industrial-Size Silicon Heterojunction Solar Cells

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SUMMARY

Silicon heterojunction (SHJ) solar cells employ nanometer-thin stacks of intrinsic and doped hydrogenated amorphous silicon (*a*-Si:H) films as carrier-selective contacts. To achieve excellent carrier selectivity, the *a*-Si:H must be carefully optimized to guarantee an atomically sharp *a*-Si:H/*c*-Si interface. In this work, by combining experiments with molecular dynamics and *ab initio* calculations, we unveil H atoms bonded to internal-void surfaces in *a*-Si:H broaden its optical bandgap via a filamentary effect near the valence-band maximum. The photovoltaic performance of rear-emitter SHJ solar cells can be significantly improved via tailoring the Si–H bonding state in the front *a*-Si:H passivation layer, resulting a power conversion efficiency (PCE) of 23.4% on a 6-inch cell. By implementing double-antireflection coatings (ARCs) of SiN_x and SiO_x, the PCE is further improved to 23.9%. More importantly, the ARCs devices show prominently improved damp-heat stability without encapsulation in 1000-hour aging at 85°C, 85% relative humidity.

Keywords

SHJ solar cells, Tailoring hydrogen states, Damp-heat stability, Mass production

1. INTRODUCTION

Silicon and hydrogen are earth-abundant elements. Their compound hydrogenated amorphous silicon (a -Si:H) is a key functional material in thin-film transistors, lithium-ion batteries, photocathodes and photovoltaics (PV).¹⁻⁴ Thanks to the excellent electronic passivation intrinsic a -Si:H(i) provides to crystalline silicon (c -Si) surfaces, doped/intrinsic a -Si:H stacks have been successfully developed as carrier-selective, passivating contacts for high-efficiency silicon heterojunction (SHJ) solar cells and related tandem PV devices.⁵⁻⁹ Assembling such contacts in an all integrated back-contact (IBC) design has resulted in the current world-record power-conversion efficiency (PCE, 26.7%) for c -Si solar cells.¹⁰ Thin a -Si:H films are usually chemical vapor deposited (CVD) by plasma-enhanced CVD (PECVD) or catalytic CVD (Cat-CVD), featuring a high H concentration present in their bulk network and on internal-void surfaces.¹¹ It has been well established that H atoms chemically passivate Si dangling bonds in a -Si:H films and on c -Si surfaces, which greatly reduces gap states and therefore the carrier recombination velocity.¹² In thin-film solar cells, a -Si:H(i) serves as PV absorber, for which ‘dense’ a -Si:H, lean in voids, yields the highest electronic quality.¹³ However, for effective passivation in SHJ solar cells, an additional criterion is that the a -Si:H/ c -Si interface must be atomically sharp (i.e. without epitaxy) to enable excellent surface passivation.¹⁴ Moreover, due to its relatively narrow bandgap (E_g , typically about 1.6 – 2.0 eV)¹⁵ and the fact that doped a -Si:H films are heavily defective, front SHJ contact stacks parasitically absorb a notable fraction of the short-wavelength solar photons, lowering the short circuit current density. Holman *et al.* found that ~70% carriers generated in a -Si:H(i) and ~100% carriers generated in the a -Si:H(p) suffer from parasitic absorption loss.¹⁶ To resolve this, recent years saw intensive efforts to integrate SHJ technology into an IBC architecture, thereby eliminating any front-contact related optical loss, which comes at increased processing complexity, however.^{2, 17} Another strategy is to replace the doped a -Si:H(p) films by materials with higher transparency and appropriate work function. To this end, metal oxides (e.g. MoO_x, VO_x and WO_x)¹⁸⁻¹⁹ and hydrogenated microcrystalline silicon oxide [μc -SiO_x(C_y):H]²⁰⁻²¹ have been successfully demonstrated as carrier-selective materials. Notably, all these strategies only achieved high PCEs with the presence of thin a -Si:H(i) buffer layers, to assure effective c -Si surface chemical passivation. This raises the question to what extent a -Si:H(i) passivation layers can be tailored to offer simultaneously excellent passivation and high optical transparency without hindering carrier transport. For this, a

detailed microscopic understanding of thin a -Si:H(i) film is required.

Moreover, since developed ~30 years ago by Sanyo (now Panasonic), the SHJ solar cell community has been dedicating vast intensive studies on enhancing the PCE and light-induced degradations/improvements,²²⁻²³ however, much less attention was paid on the device stability in damp-heat environment. Yu *et al.* achieved an impressive 23.08%-efficient 5-inch SHJ solar cell by using SiO_x/IWO double antireflection coatings (DARC).²⁴ Zhang *et al.* conducted optical simulations of SiO_x/ITO DARC by ASA (Advanced Semiconductor Analysis) software, based on which they fabricated a 19%-efficient SHJ solar cell via improving the current density.²⁵ Herasimenka *et al.* simulated optical properties of SiO_x/ITO DARC and SiO_x/SiN_x/ITO antireflection coatings (ARCs) using Opal 2 and Ray Tracer software, on that basis they successfully fabricated a 23.4%-efficient 4 cm² SHJ solar cell.²⁶ Adachi *et al.* reported that SiO_x capping on ITO layer improves the damp-heat stability of SHJ solar cells,²⁷ and they tentatively concluded the ITO/metal contact deterioration is the most possible reason for the damp-heat degradation. Morales-Vilches *et al.* enabled a 23%-efficient 4 cm² SHJ solar cell by using SiO₂/ZnO:Al DARC.²⁸ Moreover, they demonstrated the SiO_x capping impedes humid-induced ZnO:Al degradation at the grain boundaries. For long-term realistic operating environment, we believe the damp-heat stability of PV products is at least of equal importance compared to their PCE. Because the a -Si:H(i) material itself is metastable,²⁹ and therefore the specific regions and climates must be taken into account before installation. In this context, it would be urgent to inspect damp-heat stability of SHJ solar cells, and provide a potential solution.

In this work, we present an in-depth study on how the microscopic Si–H bonding state relates to the E_g of a -Si:H(i) buffer layers of only a few nanometers thin. By combining experiments with molecular dynamics (MD) and *ab initio* calculations, we elucidate that E_g is controlled by void-H-induced filamentary effect near the valence band maximum (VBM), thereby strongly affecting the optical transparency as well as carrier transport in a -Si:H(i) thin films. Based on these findings, we then optimize the E_g via tailoring the microscopic Si–H bonding state in the a -Si:H(i) passivation layer, resulting in 23.4%-efficient SHJ solar cells on 6-inch wafers. Then we clarify the stability of SHJ solar cells in damp-heat environment is closely related to passivation degradation at the a -Si:H/ c -Si interface. After capping with SiN_x and SiO_x ARCs, we find the PCE is further enhanced to 23.9%. The 1000-hour damp-heat testing at 85°C under 85% relative humidity (DH85) demonstrates the stability of the

ARCs devices is dramatically improved compared to the standard-architecture (STD) devices. This work provides a clear roadmap how to gain damp-heat-stable, high PCE, industrial-size SHJ solar cells, thereby underlines important implications for mass production.

2. RESULTS AND DISCUSSION

2.1. Si–H bonding state: impact on E_g

Figure 1A shows the Fourier-transform infrared (FTIR) spectrum of a typical device-relevant a -Si:H(i) thin film deposited by radio-frequency (RF, 13.56 MHz) PECVD. The Si–H vibration modes consist of a wagging mode at $\sim 640\text{ cm}^{-1}$, bending modes at $\sim 845\text{ cm}^{-1}$ and $\sim 890\text{ cm}^{-1}$, and stretching modes from 1900 cm^{-1} to 2200 cm^{-1} .³⁰ The latter modes depend on the precise Si–H bonding state. Notably, the stretching modes in Figure 1A can be de-convoluted by two Gaussians centered at $\sim 2000\text{ cm}^{-1}$ and $\sim 2080\text{ cm}^{-1}$, assigned to Si–H bonds in the bulk network and on internal-void surfaces, respectively (see Figure 1B).¹¹ The relative dominance of one stretching mode over another depends on deposition parameters such as substrate temperature, precursor gas flow rates, chamber pressure, and reactor design. In Figure 1C, the E_g of various a -Si:H(i) thin films with a thickness of $14.5 \pm 1.5\text{ nm}$ is plotted against their $C_{\text{H}}^{\text{tot}}$. Although a general positive correlation is observed, E_g does not scale linearly with $C_{\text{H}}^{\text{tot}}$. To gain further insight, we focus on the samples within a narrow $C_{\text{H}}^{\text{tot}}$ range of $15 \pm 1\text{ at.}\%$ (green band in Figure 1C). Figure 1D shows how their E_g increases with the so-called ‘microstructure factor’ R , defined as $R = C_{\text{H}}^{\text{void}} / [C_{\text{H}}^{\text{bulk}} + C_{\text{H}}^{\text{void}}]$, where $C_{\text{H}}^{\text{bulk}}$ and $C_{\text{H}}^{\text{void}}$ are the hydrogen concentrations associated with the bulk network and void surfaces, respectively. The increasing E_g , especially for $R > 0.5$, unambiguously suggests that void-related Si–H bonds broaden E_g more efficiently than the bulk-related Si–H bonds.

To understand the observed E_g dependency on the Si–H bonding state, we built typical different models of amorphous silicon, with and without bonded hydrogen, *i.e.* a -Si:H and a -Si, using a quenching manner from liquid Si (see EXPERIMENTAL PROCEDURES). Their theoretical radial distribution functions (RDFs) are plotted in supplementary Figure S1; for reference, the experimental neutron-diffraction RDF of pure a -Si, taken from literature,³¹ is also shown therein. Our modelled RDFs display identical characteristics as the experimental data, with peaks at $\sim 2.35\text{ \AA}$, $\sim 3.81\text{ \AA}$ and $\sim 5.81\text{ \AA}$, which suggests the generated models from MD and *ab initio* dynamics are reasonable. As

expected, compared to *c*-Si (supplementary Figure S2), the amorphous models lose many characteristic RDF peaks, due to their absence of long-range order. The additional RDF peak at ~ 1.53 Å in *a*-Si:H models stems from Si–H bonds.

For clarity, we consider Si atoms with interatomic distances away from the closest H atom that are larger than 2, equal to 2 and equal to 1 bonds, which are labeled as bulk-Si, 2-Si, and 1-Si, respectively, as illustrated in supplementary Figure S3. Partial (electronic) density of states (PDOS) of *a*-Si:H models were calculated using the HSE06 hybrid functional,³² an advanced functional for electronic predictions. For *a*-Si:H featuring exclusively bulk Si–H bonds (*i.e.* no Si–H on internal voids), the average PDOS of bulk-Si, 2-Si, 1-Si are shown in Figures 2A–2C. Relative to the Fermi level, the conduction band minimum (CBM) and VBM of Si atoms are hardly affected by H decoration. The CBM and VBM of bulk H in Figure 2D are only slightly shifted compared to bulk *a*-Si, likely due to H-induced band tail reduction.³³ This explains why experimentally *a*-Si:H with small R ($C_{\text{H}}^{\text{void}} \ll C_{\text{H}}^{\text{bulk}}$) shows a narrow E_{g} (Figure 1D). Figures 2E–2H are the average PDOS of bulk-Si, 2-Si, 1-Si and H of *a*-Si:H containing only Si–H bonds present on internal-void surfaces (*i.e.* no bulk Si–H). Contrasting to Figures 2A–2C, the CBM and VBM gradually shift to higher and lower energies, respectively, compared to bulk *a*-Si. This might be attributed to Si-bonding relaxation near surfaces of voids, because the energy and stress of surface Si atoms significantly differ from Si atoms present in the bulk *a*-Si.³⁴ Furthermore, Si–H bonds associated with internal voids (Figure 2H) show the highest CBM and lowest VBM energetic positions. We note that *ab initio* PDOS cannot predict conduction and valence band offsets (ΔE_{c} , ΔE_{v}) because the theoretical state distributions are referred to the Fermi level (which is fixed at 0 eV), rather than to the vacuum level. Therefore, we determined ΔE_{c} and ΔE_{v} experimentally. Figure 2I shows ultraviolet photoelectron spectroscopy (UPS) spectra of two *a*-Si:H films with narrow (1.59 eV) and wide (1.85 eV) E_{g} s, determined by ellipsometry (supplementary Figure S4). Their work functions are 4.33 eV and 4.48 eV, determined from the cutoffs of the UPS spectra. As a consequence, their CBM positions are at 3.54 eV and 3.55 eV, whereas their VBM are at 5.13 eV and 5.41 eV, respectively. These results indicate that the void-related Si–H bonds broaden the E_{g} via shifting VBM to a lower energy, whilst the CBM position is almost unchanged, consistent with the conclusion from Schulze *et al.*³⁵ This founding is of importance for device design. An increase in ΔE_{v} may result in hole-collecting issues. Therefore, employing wider bandgap *a*-Si:H layers at the

front of the SHJ device favors the so-called rear-emitter polarity, collecting electrons at the sunward side. Earlier studies estimated the void density to be $\sim 10^{20} - 10^{21} \text{ cm}^{-3}$ in porous $a\text{-Si}(\text{C}_x)\text{:H}$.³⁶⁻³⁷ Such a high void density gives rise to a filamentary effect,³⁸ i.e., a VBM retreatment that is structure-relaxation induced,³⁵ as illustrated in Figure 2J.

2.2. Tailoring Si–H bonding state enables high PCE SHJ solar cells

Based on the aforementioned discussion, we now experimentally manipulate the E_g of $a\text{-Si:H}$ via tailoring the Si–H bonding state. Matsuda pointed out that H-exchange reactions in the plasma generate local heating, which improves 2-D diffusion of radicals arriving from the plasma (e.g., SiH_3 and SiH_2) on the growth surface of the film under deposition.³⁹ From this viewpoint, H_2 dilution helps to grow the ‘dense’ $a\text{-Si:H}$ films. Figure 3A shows FTIR spectra of nanometer-thin ($14.5 \pm 1.5 \text{ nm}$) $a\text{-Si:H}(i)$ films. The brown line corresponds to a film deposited at a high $[\text{H}_2]/[\text{SiH}_4]$ ratio, whose $C_{\text{H}}^{\text{tot}}$ is lower than the IR spectroscopy limit. Its ellipsometric angles (Ψ , Δ) cannot be satisfactorily analyzed by the Tauc-Lorentz model,⁴⁰ instead, they are perfectly fitted with a combination of two Tauc-Lorentz oscillators and three Gaussian oscillators, indicating its nano-crystalline silicon ($nc\text{-Si:H}$) nature. This ordered microstructure is caused by H-induced crystallization.⁴¹ To obtain $a\text{-Si:H}$, the $[\text{H}_2]/[\text{SiH}_4]$ ratio is gradually decreased to 0, leading to an increase of R from 0 to 100%. Correspondingly, E_g is broadened to 1.65 eV, 1.79 eV, 1.85 eV and 1.96 eV, as indicated by the absorption cutoffs in Figure 3B.

Next, we apply these $nc\text{-Si:H}(i)$ and $a\text{-Si:H}(i)$ films as electron-selective passivation layer contacting the front surface of 6-inch bifacial rear-emitter SHJ solar cells (Figure 3D). Device fabrication details are found in EXPERIMENTAL PROCEDURES. Except for the front intrinsic passivation layer, other layers are kept identical. Figure 3C displays the minority carrier lifetime (τ_{eff}) and the saturation current density (J_0) extracted at an injection level of $1.0 \times 10^{15} \text{ cm}^{-3}$ and $1.0 \times 10^{16} \text{ cm}^{-3}$, respectively, measured by a Sinton tester WCT-120. Apparently, the passivation is improved with increasing E_g . Figure 3D shows their light J – V and P – V curves under standard 1-sun illumination conditions, the parameters are listed in Table 1. Expectedly, the device with $nc\text{-Si:H}$ passivation shows the lowest open-circuit voltage (V_{oc}), short-circuit current density (J_{sc}), fill factor (FF) and maximum output power point (V_{mp} , I_{mp}), resulting in the lowest PCE of 19.61%. This implicates that H plays a

key role in achieving high-quality surface passivation. In PECVD process, extremely poor passivation is observed in case of surface epitaxial growth, in which H atoms are scarce.¹⁴ In Figure 3B, the high absorption of the *nc*-Si:H at low energies <1 eV suggests H scarcity incurs gap states. At the highly defective *nc*-Si:H/*c*-Si interface, an adverse electron barrier is expected at the *c*-Si surface due to Fermi level pinning,⁴³ detrimental to the device performance.

Compared to *nc*-Si:H and *c*-Si, *a*-Si:H(*i*) features a wider E_g , thereby reducing parasitic absorption on device level. In Table 1, we observe a linear correlation between J_{sc} and E_g , which can be empirically described by $J_{sc} = A \cdot E_g + B$, where A and B equal to 2.40 mA/cm²·eV and 33.75 mA/cm², respectively. This linear increase in J_{sc} is most likely due to the improved transparency for the approximate uniform solar irradiance between 1.65 eV and 1.96 eV (supplementary Figure S5), and increasing improvement in *c*-Si surface passivation (Figure 3C).⁴⁴ From Table 1, we also find V_{oc} increases with E_g . Essentially, when field passivation from the capping doped *a*-Si:H is fixed, the V_{oc} is determined by the defect density (D_{it}) at the *a*-Si:H(*i*)/*c*-Si interface via $V_{oc} \propto \ln(J_{ph}/J_0 + 1)$,⁴⁵ where J_0 is the flux of uncollected carriers via recombination, and J_{ph} is the photo-generated current density. The V_{oc} increase can be attributed to the J_0 decrease from 21 fA/cm² to 3.5 fA/cm², as shown in Figure 3C. Figure 4A shows the SEM image of textured surface with random pyramids, which are not rounded using acid solution. These sharp valleys might lead to incomplete contact and poor surface passivation in silicon heterojunction.⁴⁶ Figures 4B and 4C show energy-dispersive X-ray spectroscopy (EDX) mappings of indium and silicon in tungsten-doped indium oxide (IWO), used as transparent electrode in our devices, and the underlying silicon layers and substrate, respectively. The abrupt boundary confirms an intimate conformal contact between IWO and *a*-Si:H. In terms of *a*-Si:H deposition, the 109° valley angle in Figure 4C indicates *a*-Si:H is also uniformly grown at the pyramid valleys. To understand the passivation, Figures 4D and 4E display high-resolution transmission electron microscopy (HR-TEM) cross-sectional images of *a*-Si:H(*i*)/*c*-Si interfaces, featuring *a*-Si:H(*i*) with narrow (1.65 eV) and wide (1.85 eV) E_g s, respectively. Unlike perfect epitaxy by high-temperature molecular beam epitaxy,⁴⁷ the low-temperature RF-PECVD epitaxy have lattice distortions. A previous study reported *a*-Si:H(*i*) epitaxy leads to low V_{oc} and FF.¹⁴ The interface atomic distortion in Figure 4D indicates that the high D_{it} is not necessarily caused by dangling bonds, it is more likely resulted from distortion-induced gap states, analogous to distortion-induced defects in amorphous silicon.⁴⁸ By solving Poisson and

continuity equations on the AFORS-HET device-simulation platform,⁴⁹ we calculated the hetero-interface band alignment at open-circuit condition as a function of the D_{it} . The results are shown in supplementary Figure S6. For a D_{it} value as high as $1.0 \times 10^{13} \text{ cm}^{-2}$, the splitting of electron and hole quasi Fermi levels, referred to as E_{fn} and E_{fp} , in *c*-Si is dragged down due to E_{fp} pinning [Figure S6(A)]. Along with D_{it} decreases to $5.0 \times 10^{11} \text{ cm}^{-2}$, $1.0 \times 10^{10} \text{ cm}^{-2}$ and 0 cm^{-2} , the $E_{fn} - E_{fp}$ splitting increases to 644 meV, 726 meV and 738 meV, respectively [Figures S6(B)–S6(D)]. The best value 738 meV is in good agreement with the experimental highest $V_{oc} \sim 737.9 \text{ mV}$ for STD rear-emitter SHJ solar cell in Table 1. The similar V_{oc} evolution trend between the simulation and experiment qualitatively demonstrates it is controlled by the D_{it} .

In contrast to the continuous increase of J_{sc} and V_{oc} with E_g , the FF reaches a maximum at $\sim 1.85 \text{ eV}$ (see Table 1). As the E_g increases from 1.65 eV to 1.85 eV, the device FF boosts from 78.8% to 82.1%, followed by a decrease to 80.4% with an even wider E_g of 1.96 eV. To understand this, we simulated the dependence of FF on E_g of the front *a*-Si:H(*i*) passivation layer, in which the affinity (CBM) was fixed to satisfy the results in Figure 2 and literature.³⁵ We find FF almost keeps constant as the E_g increases (supplementary Figure S7), excluding the possibility that FF reduction is caused by the electron barrier at *a*-Si:H(*i*)/*c*-Si interface. To gain further qualitative insight, we measured the dark and photo-conductivities of *a*-Si:H(*i*) films grown at different $[\text{H}_2]/[\text{SiH}_4]$ flow ratios, measured by the TLM method (inset in Figure S8(A)).⁵⁰ In conjunction with a decrease of $[\text{H}_2]/[\text{SiH}_4]$ from 7.5 to 0, the silicon film gradually changes from dense *a*-Si:H(*i*) to porous *a*-Si:H(*i*) with $E_g \sim 1.96 \text{ eV}$. As $[\text{H}_2]/[\text{SiH}_4]$ is decreased from 7.5 to 4.5, the dark conductivity exponentially decreases by 2 orders from 10^{-8} S/cm to 10^{-10} S/cm (Figure S8(A)). We could not measure the dark conductivity of samples grown at even lower H_2 dilution. In Figure S8(B), the photo-conductivity also exponentially decreases from $\sim 10^{-5}$ to 10^{-6} S/cm when $[\text{H}_2]/[\text{SiH}_4]$ decreases from 7.5 to 1.5. However, as the H_2 dilution continuously approaches to 0, the photo-conductivity drastically decreases by ~ 2 orders. The conductivity data in Figure S8 is consistent with literature regarding the order of magnitudes.⁵¹⁻⁵² Two factors might account for this lower FF of SHJ solar cell passivated by *a*-Si:H(*i*) with E_g of 1.96 eV (Table 1): first, it may be caused by mid-gap defects in porous *a*-Si:H(*i*), and another is the high resistivity of the *a*-Si:H(*i*) film. The high τ_{eff} and low J_0 of SHJ solar cells passivated by 1.96 eV *a*-Si:H(*i*) exclude the first speculation. According to Yoshikawa *et al.*,² the largest losses in FF of SHJ

solar cells are due to a limited effective carrier lifetime at maximum power point and to series resistance from all resistive materials along the carrier collection path. Taking into consideration the excellent passivation in Figure 3C and the low conductivity in Figure S8, we believe the high resistivity of porous *a*-Si:H(*i*) is a possible bottleneck to the FF.

2.3. ARCs stabilize SHJ solar cells

SHJ solar cells have been developing for ~30 years, and now are on the way to mass production, in competition with aluminum back surface field (Al-BSF) and passivated emitter and rear (PERC) *c*-Si solar cells. However, a deep investigation on damp-heat stability of the SHJ passivation without encapsulation is still missing. According to the relation between V_{oc} and J_0 ,⁵³

$$V_{oc} = \frac{kT}{q} \ln \left(\frac{J_{sc}}{J_0} + 1 \right),$$

it is deduced that

$$\frac{\partial V_{oc}}{\partial J_0} = -\frac{kT}{q} \frac{1}{J_0}.$$

This differential equation indicates that high-efficient SHJ solar cells (with small J_0 values) are more sensitive to J_0 . A small increment in J_0 might cause a large decrement in the V_{oc} , and thus PCE. Furthermore, wide- E_g *a*-Si:H(*i*) passivation layer (used for the optimized SHJ solar cells in this work) is metastable featuring highly disordered microstructure, which has a risk of being oxidized in damp-heat environment. Therefore, it becomes necessary to check the stability of our optimized SHJ solar cells in real environment and DH85 testing, as a prerequisite for mass production. We use 5-nm-thick *a*-Si:H(*i*₂)/5-nm-thick *a*-Si:H(*i*₁) stacks to passivate both surfaces of (100) float-zone (FZ) Si wafers (thickness: 280 ± 20 μm ; resistivity: 1–6 $\Omega \cdot \text{cm}$), forming *a*-Si:H(*i*₂)/*a*-Si:H(*i*₁)/*c*-Si/*a*-Si:H(*i*₁)/*a*-Si:H(*i*₂) symmetric passivated structures. For all samples, E_g of *a*-Si:H(*i*₂) is fixed at 1.65 eV, only E_g of *a*-Si:H(*i*₁) is varied between 1.71 eV and 1.85 eV. We stored these passivated SHJ samples in a black box, and then put the box in the desert close to the *red sea* (39°36'E 22°17'N). In this black box, moisture is accessible, while sunlight is blocked. By this means, we can monitor the hot-dry & cool-humid (HDCH) stability without disturbance from light soaking. Figures 5A and 5B display the relative humidity and temperature in the box during the experiment. The night relative humidity reaches as

high as $> 90\%$, and the diurnal temperature can reach as high as $\sim 50^\circ\text{C}$. This environment is anticipated to exert harsh impact on our SHJ passivating samples. In Figure 5C, the red and blue triangles represent samples passivated by 1.85 eV $a\text{-Si:H}(i_1)$. Clearly, such a porous $a\text{-Si:H}(i_1)$ successfully achieves a high $\tau_{\text{eff}} \sim 16$ ms, however, this value decreases to ~ 5 ms after 4-day exposure (red triangles). It is interesting to find that a brief annealing (15 min; 190°C in N_2) can fully recover the τ_{eff} to the initial value (blue triangles), but then followed by a quick decay to ~ 5 ms within only 2 days. The faster τ_{eff} decrease after annealing (compared to initial state) suggests the annealing state differs from the initial state despite they have identical τ_{eff} . After the τ_{eff} decreases to a saturated value, the SHJ passivation manifests a “memory” behavior, where the “high” state is written by annealing and can be erased to “low” state by damp-heat degradation. From Figure 5C, we also notice that, after 90 hours, wafers passivated by $a\text{-Si:H}(i_1)$ with E_g of 1.85 eV and 1.71 eV degrade to an identical τ_{eff} value (red triangles versus green squares). This indicates the porous $a\text{-Si:H}(i_1)$ will lose the passivating advantage in the HDCH environment.

Figure 6A shows τ_{eff} evolutions of the $a\text{-Si:H}(i_2)/a\text{-Si:H}(i_1)/c\text{-Si}/a\text{-Si:H}(i_1)/a\text{-Si:H}(i_2)$ passivated samples after annealed at 55°C , 85°C , and 105°C for 50 hours in dry N_2 atmosphere. The τ_{eff} keeps constant under different temperatures, excluding the possibility that passivation degradation in Figure 5C was caused by hydrogen dispersive diffusion. After 8-day exposure in the desert, a conspicuous Si-O mode at $\sim 1000\text{ cm}^{-1}$ is observed on the FTIR spectrum (Figure 6B), suggesting oxidation of the $a\text{-Si:H}$ film. This result indicates moisture plays a key role in the $a\text{-Si:H}$ oxidation, and hence in the passivation degradation. Figure 6C displays carrier density N_e , mobility μ_e , and resistivity of 80-nm-thick IWO and IWO:H films on quartz glass substrates before and after 1000-hour DH85 testing. No obvious degradations are observed in these transparent conductive films, excluding the possibility that SHJ solar cell degradation in damp-heat condition is caused by the degradation of IWO layer. Photoluminescence (PL) images of an optimized 4-inch STD rear-emitter SHJ solar cell before and after 1000-hour DH85 testing are shown in Figure 6D. As the dash lines indicate, milk-like dark areas (degradations) emerged from edges. This once again demonstrates the DH85 degradation mainly arises from degradation of the $a\text{-Si:H}/c\text{-Si}$ passivation, rather than the IWO coating, because dark area on the whole wafer is expected on the PL image if the degradation is caused by the IWO layer. In comparison to the demonstrated ITO/metal contact degradation²⁷ and ZnO:Al grain-boundary degradation²⁸, this

is a new degradation mechanism (relating to the *a*-Si:H/*c*-Si interface) for SHJ solar cell in damp-heat environment. However, Figure 6D also finds sporadic small dark points in the center area of the STD SHJ solar cell after 1000-hour DH85 testing. These degradation points are likely attributed to ionic contaminations during experimental process, e.g. Na⁺, whose diffusion is detrimental to the passivation.²⁷ In contrast, the PL image of ARCs cell exhibits negligible changes after 1000-hour DH85 testing (Figure 6E), indicating a significantly improved stability, most probably mainly due to isolation of moisture by the ARCs at the edges.

Finally, we test the damp-heat stability of SHJ solar cells passivated by 1.85 eV *a*-Si:H(*i*) under DH85 testing. In Figure 7, the triangles represent the PV parameters of SHJ solar cells during 20-hour 100 mW/cm² light soaking, demonstrating an improved PCE from 22.76% to 23.36% (independent certificated value of 23.18%, as shown in Figure 3E), thanks to the improved V_{oc} and FF (Figures 7A and 7C) by interface relaxation.²² Following the light soaking, the devices experience 1000-hour DH85 testing. The V_{oc} , J_{sc} and FF degrade substantially after 100 hours, as demonstrated by the red circles, resulting in an absolute PCE loss of ~3.1% at 1000 hours. Photoluminescence (PL) images demonstrate the *a*-Si:H(*i*)/*c*-Si interface degradation starts from the wafer edge because the shadow mask during IWO capping expose ~0.5 mm *a*-Si:H to the air (supplementary Figure S9 and Figure S10). To overcome the passivation degradation, double ARCs consisting of 40-nm-thick SiN_x and 85-nm-thick SiO_x are capped on front side of SHJ solar cells after metallization. The thickness of SiN_x and SiO_x dielectrics are based on optical simulations using Wafer Ray Tracer, as illustrated in Figure S11. Compared to the STD cells, it is exciting to observe that the PCE is further improved to 23.9%, as presented by the squares in Figure 7D, thanks to a significantly improved J_{sc} . The ARCs demonstrate we can reduce >50% of the expensive IWO materials for SHJ solar cells, but the additional depositions in the PECVD chamber are prejudicial to the cost saving. However, it is interesting to note that Issa and co-workers recently reported a cost-effective atmospheric pressure chemical vapor deposition (APCVD) for growing high-quality SiO_x thin films without voids, pinholes or cracks.⁵⁴ Optical simulations demonstrate the ARCs couple more sunlight into the *c*-Si absorber (supplementary Figure S12). However, the improvement in J_{sc} will be limited in module level because of the refractive index of ethylene vinyl acetate (EVA) and glass.⁵⁵ Interestingly, V_{oc} also increases slightly, most likely due to the H penetration into the *a*-Si:H/*c*-Si interface during SiN_x capping, analogy to the dynamics

observed by Cruz *et al.*⁵⁶ The ARC devices show a slight lower FF because thinner front IWO layer (~35 nm) are applied, in order to maximize the light coupling. However, the important thing is that the absolute PCE of the ARC devices only degrades ~0.49% after 1000-hour testing at DH85, demonstrating a dramatically improved stability. Furthermore, the PCE tends to stabilize after 600-hour damp-heat exposure. A weak logarithmic decay predicts the PCE of ARC devices still reach 23.3% after 30-year installment, under the assumption that every day has 5-hour DH85 condition. Taking into account that SHJ solar cells exhibit lower temperature coefficients than that of Al-BSF and PERC *c*-Si solar cells,⁵⁷ we believe SHJ solar cell is a competitive candidate for next-generation high-efficiency *c*-Si PV product, provided that the cell edge must be meticulously protected to avoid suffering from *a*-Si:H(*i*)/*c*-Si interface degradation. These findings should also capture interest in module-level PVs, because moisture can diffuse into the PV module through the back sheet and the EVA sheet,⁵⁸ and the EVA itself also degrades after a long-term installation.⁵⁹ In Figure 7, stability data of mini modules in DH85 environment is also presented. Compared to STD raw cells (without encapsulation), degradations of V_{oc} , J_{sc} and FF are effectively slowed down. Nevertheless, the PCE degradation of mini modules is not completely suppressed, which is still faster than the ARC cells, indicating moisture can penetrate through the encapsulation. From this viewpoint, we expect the ARCs take effect even in the module level. Another possible way to enhance the stability of SHJ cells is to replace the deposition technique of the *a*-Si:H(*i*) passivation layer with Cat-CVD, as illustrated in Figure S13. This once again confirms the damp-heat degradation of our devices is dominated by the *a*-Si:H(*i*) passivation layer. Detailed discussions on this topic are beyond this work.

3. Conclusions

In this work, we manipulated E_g of the front *a*-Si:H(*i*) passivation layer for rear-emitter SHJ solar cells via tailoring the Si–H bonding state. With increasing E_g of *a*-Si:H(*i*), the J_{sc} and V_{oc} increase continuously, however, the FF reaches the best value at an optimal E_g value of 1.85 eV. The high resistivity of porous *a*-Si:H(*i*) with $E_g \sim 1.96$ eV is probably caused by the disordered microstructure due to appearance of high-density large voids. By implementation of the optimal *a*-Si:H(*i*) passivation layer, 6-inch STD rear-emitter SHJ solar cells with an efficiency of 23.4% have been achieved. However, damp-heat testing at DH85 unveils such an *a*-Si:H(*i*)/*c*-Si interface is metastable, which can

be overcome by using the double ARCs of SiN_x and SiO_x, achieving an improved efficiency of 23.9% in the meantime. This work provides significant implications for developing damp-heat-stable high-efficiency SHJ solar cells for mass production. Moreover, it also outlines a clear guidance to design other stable *a*-Si:H electronics in damp-heat climates.

EXPERIMENTAL PROCEDURES

Full experimental procedures are provided in the Supplemental Information.

SUPPLEMENTAL INFORMATION

Supplemental Information can be found online at [xxx](#).

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AUTHOR CONTRIBUTIONS

W.L., S.D.W., and Z.L. conceived the idea. W.L., L.Z., X.Y., and J.S. carried out the device fabrication, electrical characterization and analysis. W.L. carried out the MD, *ab initio* and device simulations. L.Y., L.X., Z.W., R.C., J.P., J.K., K.W., and F.M. assisted with materials preparation and characterizations. W.L. wrote the paper, and all other authors provided feedback.

DECLARATION OF INTERESTS

The authors declare no competing interests.

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Figure titles and legends

Figure 1. Internal-Void Hydrogen Atoms Broaden Optical Bandgap of Nanometer-Thin *a*-Si:H Films.

(A) FTIR spectrum of a typical *a*-Si:H(*i*) thin film fabricated using RF-PECVD.

(B) Si–H stretching mode de-convoluted by two Gaussians centered at $\sim 2000\text{ cm}^{-1}$ and $\sim 2080\text{ cm}^{-1}$.

(C) E_g of *a*-Si:H(*i*) as a function of the C_H^{tot} ; E_g is obtained from ellipsometry, and C_H^{tot} from FTIR; samples within the green band have a C_H^{tot} of 15 ± 1 at.%.

(D) E_g versus R plot for samples within the green region of (C). R values were calculated with different A_{2080} of $9.0 \times 10^{19}\text{ cm}^{-2}$ (circles) and $2.2 \times 10^{20}\text{ cm}^{-2}$ (stars). The vertical line represent $R \sim 0.5$.

Figure 2. *Ab Initio* Interpretation of Valence Band Retreatment.

(A – D) Average PDOS of bulk-Si, 2-Si, 1-Si and H in *a*-Si:H with only bulk Si–H bonds.

(E – H) Average PDOS of bulk-Si, 2-Si, 1-Si and H in *a*-Si:H with only void Si–H bonds.

(I) UPS spectra of *a*-Si:H films with narrow (1.59 eV) and wide (1.85 eV) E_{gS} , using He I excitation (21.2 eV) after surface etching.

(J) Schematic of VBM retreatment caused by structure-relaxation induced filamentary effect.

Figure 3. Device Optimizations via Tailoring Hydrogen States in Front-Side *a*-Si:H passivation Layer.

(A–B) FTIR spectra and absorption coefficients of *nc*-Si:H(*i*) and *a*-Si:H(*i*) films with different E_g .

(C) τ_{eff} and J_0 of 1–6 $\Omega\cdot\text{cm}$ *n*-type *c*-Si wafers with different passivation layers before RPD deposition.

(D) Medium light $J-V$ and $P-V$ curves of SHJ solar cells with different front passivation layers, standard architecture of a rear-emitter SHJ solar cell.

(E) Certificated PV parameters of an STD 6-inch SHJ solar cell, passivated by intrinsic *a*-Si:H(*i*) layer with $E_g \sim 1.85$ eV.

Figure 4. Microstructural Characterizations of *a*-Si:H/*c*-Si Interfaces.

(A) SEM image of textured surface with random pyramids on SHJ solar cell.

(B–C) EDX mappings of In and Si at a pyramid valley.

(D–E) HR-TEM cross-sectional images of a -Si:H(i)/ c -Si interfaces passivated by a -Si:H(i) with narrow and wide E_g s.

Figure 5. Passivation Evolutions in Hot-Dry & Cool-Humid Real Environment.

(A–B) Relative humidity (RH) and temperature (T) of the desert close to the *red sea* (39°36'E, 22°17'N), where the passivated samples were exposed.

(C) τ_{eff} evolution of three groups of passivated samples as a function of exposure time in the desert. The passivated structure is a -Si:H(i_2)/ a -Si:H(i_1)/ c -Si/ a -Si:H(i_1)/ a -Si:H(i_2). Thickness and E_g of a -Si:H(i_2) are 5 nm and 1.65 eV for all samples. Thickness of a -Si:H(i_1) is 5 nm for all samples, but with different E_g of 1.85 eV (blue and red triangles) and 1.71 eV (green squares).

Figure 6. Moisture-Induced a -Si:H/ c -Si Interface Degradations.

(A) τ_{eff} evolution of a -Si:H(i_2)/ a -Si:H(i_1)/ c -Si/ a -Si:H(i_1)/ a -Si:H(i_2) passivated samples annealed at different temperatures in N_2 atmosphere.

(B) FTIR spectra of an a -Si:H(i_2)/ a -Si:H(i_1)/ c -Si/ a -Si:H(i_1)/ a -Si:H(i_2) passivated sample before and after 8-day exposure in the desert close to the *red sea* (39°36'E, 22°17'N).

(C) Carrier density N_e , mobility μ_e , and resistivity of 80-nm-thick IWO and IWO:H films on quartz glass substrates before and after 1000-hour DH85 testing via hall-effect measurement.

(D–E) PL images of STD and ARCs bifacial rear-emitter SHJ solar cells before and after 1000-hour DH85 testing.

Figure 7. DH85 Aging Characteristics of STD and ARCs SHJ Solar Cells, and Corresponding Mini Modules.

(A–D) Relative evolutions of V_{oc} , J_{sc} , FF and PCE of STD (triangles ▲ and circles ●), ARCs (squares ■) bifacial rear-emitter SHJ solar cells, and STD mini modules (stars ★) encapsulated by 0.5 mm EVA and 3 mm glass, and back sheet. The first 20 hours were subjected to light soaking under 100 mW/cm² at 25°C. After that, the cells and mini modules experienced 1000-hour DH85 testing in dark environment.

Table 1. Average PV parameters of 6-inch rear-emitter SHJ solar cells with different *nc*-Si:H(*i*)/*a*-Si:H(*i*) passivation layers. Thickness of the passivation layers on pyramids was estimated to be 5 ± 0.25 nm according to $d_{\text{pyramid}} = d_{\text{flat}}/1.7$.⁷

E_g (eV)	C_H^a (at.%)	R (%)	V_{oc} (mV)	J_{sc} (mA/cm ²)	FF (%)	V_{mp} (mV)	I_{mp} (mA/cm ²)	PCE (%)
<i>nc</i> -Si:H	–	–	680.12	37.43	77.03	569.18	34.45	19.61
1.65	7.9	0	726.43	37.72	78.76	618.02	34.92	21.58
1.79	18.4	72	731.40	38.02	80.83	632.41	35.54	22.48
1.85	20.8	97	736.60	38.15	82.07	647.83	35.60	23.06 ^b
1.85	20.8	97	738.10	38.46	82.28	650.81	35.89	23.36 ^c
1.85	20.8	97	741.01	40.30	79.96	647.11	36.90	23.88 ^d
1.96	24.2	100	737.90	38.47	80.42	638.73	35.74	22.82

^a $A_{2000} = 9.0 \times 10^{19} \text{ cm}^{-2}$, $A_{2080} = 2.2 \times 10^{20} \text{ cm}^{-2}$ from literature⁴²

^bThe average PCE in the STD batch

^cThe best PCE in the STD batch

^dThe best PCE in the ARCs batch