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ARTICLE

Improved amorphous/crystalline silicon interface passivation with two-step intrinsic layers

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A novel way of two-step intrinsic (*i*) layers growth process was provided to improve amorphous/crystalline silicon (a-Si:H/c-Si) interface passivation. This effort was guided by the research of the relationship between the bulk photo-electrical properties of the a-Si:H film using a variable H₂-dilution gas flow ratio $R=[\text{H}_2/\text{SiH}_4]$ and their passivation results of amorphous/crystalline silicon interface. The results demonstrated that the a-Si:H/c-Si interface had more dominating influence on the passivation than the bulk of the a-Si:H films. However, when the a-Si:H/c-Si interface did not occur the epitaxial growth, the bulk quality of the a-Si:H film had significant effect on the passivation. The optimum two-step process was designed as follows: the initial stage of the *i* layer was deposited at lower *R* than the bulk to ensure that the interface remained within the amorphous phase, the second stage was deposited using the optimum bulk a-Si:H film process to get the best quality of the passivation film. Although only 5nm thick passivation film was deposited on polished Cz-Si wafers, the optimum effective lifetime was up to 1.7ms. after the annealing, the effective lifetime could further raised to 2.5ms and the corresponding implied V_{oc} was up to 0.724V.

silane by H₂ produce the good quality a-Si:H film without risking epitaxy growth at the c-Si interface is the question needed to solve.

Introduction

As one of the most promising solar cell structures, it was reported that recently the efficiency of the silicon heterojunction (SHJ) solar cell has been reached 25.6%, with the total area of 143.7 cm². It is the highest efficiency among the silicon based solar cells to date. To obtain high V_{oc} and high efficiency, excellent surface passivation of c-Si is a key prerequisite.^{1,2} Generally, there are many ways of passivating the surface of c-Si such as wet-chemical, hydrogen plasma, thermal annealing treatment, or thin film deposition.³⁻⁸ It has been shown that a ultra-thin intrinsic hydrogenated amorphous silicon (a-Si:H) layer demonstrated superior passivation capability as well as adequate carrier transport ability in high-efficiency silicon heterojunction solar cells.^{9,10} High quality passivation for c-Si by means of direct PECVD a-Si:H(*i*) deposition must have an abrupt a-Si:H/c-Si interface.¹¹ In addition, the optimized a-Si:H material is produced from deposition regimes close to the crystalline transition both for thin film silicon solar cells¹² and for c-Si wafer passivation.¹³ Highly diluted or highly depleted silane plasmas are the two traditional PECVD techniques depositing this material. Because highly diluted silane plasma growth process will result in epitaxy growth at the c-Si interface, many groups use the second one, i.e. pure silane plasma growth process.^{11,13,14,15} However, to our best knowledge, the a-Si film deposited by pure silane without the participation of H₂ has many defects, and highly depleted silane has the risk of producing large number of polymer radicals which are harmful to the film in the depositing chamber. So employing diluted

In this study we proposed two-step strategy of depositing *i* layers by H₂ dilution to alleviate these problems, and achieved better performance.

Experimental

Film prepared and characterization

The films were deposited in a capacitively-coupled parallel plate reactor using a plasma excitation frequency of 13.56 MHz at a low temperature of 130°C with the H₂ dilution ratio $R(R=[\text{H}_2/\text{SiH}_4])$ values from 1 to 4, total gas pressure 1 Torr. Double sided mirror polished 250 μm thick relatively low resistivity 3.0 Ω·cm n-type Czochralski silicon (111) CZ-Si wafers were used. Before film deposition, the Si wafers were cleaned by the standard RCA cleaning process. After that the wafers were immersed in 1% HF solution for 3 minutes to remove the native oxide layer and then transferred into the load-lock of the deposition system immediately. To quantify the quality of passivation, the i-a-Si:H films were symmetrically deposited on the c-Si substrate and then effective minority carrier lifetimes and the corresponding implied V_{oc} at specific injection level of minority carrier density (MCD) of 10¹⁵cm⁻³ were measured by the quasi-steady-state photo-conductance (QSSPC) method using a Sinton Consulting WCT-120 lifetime tester. The QSSPC

measurements established them by measuring the photoconductivity with the variation of illuminating light intensities. Implied V_{oc} was calculated at 1 sun illumination without contact formation and reported the linear dependency on real V_{oc} of solar cells.^{16–17} As is well known, the implied V_{oc} which is the theoretical maximum open-circuit voltage is given by the splitting of the quasi-Fermi levels Φ_n and Φ_p . It is given by:

$$\text{impl}V_{oc} = \frac{kT}{q} \ln \left[\frac{(n_0 + \Delta n)(p_0 + \Delta p)}{n_i^2} \right] \quad (1)$$

Where K is Boltzmann's constant, T is the absolute temperature, q is the electric charge, n_0 and p_0 are the electron and hole densities at thermal equilibrium, Δn and Δp are the excess electron and hole densities, n_i is the intrinsic carrier density. In a semiconductor under steady-state illumination, effective minority carrier lifetime τ_{eff} is given by:

$$\tau_{eff} = \frac{\Delta n_{av}}{G_L} \quad (2)$$

Where Δn_{av} is the average excess carrier density, G_L is the generation rate. The a-Si:H films deposited on c-Si substrate were characterized ex-situ by Fourier transform infrared (FTIR) spectroscopy. And the a-Si:H films deposited on glass substrate were used for conductivity, reflectance–transmittance and thickness measurements. Post-deposition annealing was conducted in a PECVD chamber at 180°C under H_2 atmosphere with 2 Torr pressure for half an hour.

Result and discussion

Fig. 1 shows the lifetimes of the a-Si:H films, the a-Si:H films' microstructure factors' complement of 1 (R'_{as-dep}) with thickness of 20nm deposited on the c-Si substrates and the photosensitivity of the a-Si:H films with thickness of 100nm deposited on the glass substrates as functions of R . With the increase of the R values, the lifetime increased to a peak at $R=2$ and then decreased. The photosensitivity has the similar tendency but the peak existed at $R=3$. Why is not the R where the best passivation obtained the same as that where the best a-Si:H bulk film quality obtained? These two series were deposited using the same process. The biggest difference was that their substrates: one used glasses, and the other used silicon wafers. So the a-Si:H/c-Si interface might be the origin of this difference. As is known, the microstructure factor of the a-Si:H film, marked as R , i.e. the ratio of the SiH_2 peak area to the SiH peak area ($R = I_{SiH_2} / (I_{SiH_2} + I_{SiH})$) reflects the film's compactness and bulk quality. The smaller the value of R , the better the bulk quality of the a-Si:H film i.e. the more compact the structure of the a-Si:H film.¹⁸ Here we set R' as $R'=1-R$, for the convenience of the following discussion. Obviously, the larger the value of R' , the better the bulk quality of the a-Si:H film i.e. The more compact the structure of the a-Si:H film.

As Fig. 1 shown, the R'_{as-dep} and the photosensitivity both have the similar tendency with the increase of the R values. It demonstrated that the bulk quality of the a-Si:H films deposited on the c-Si substrates had the similar tendency as the a-Si:H films deposited on the glass substrates with the increase of the R values. So we could

confirm the a-Si:H/c-Si interface was the origin of the difference mentioned above. Indeed, Schulze et al.¹⁹ have showed that the as-deposited interface defect density is determined by the local and n-

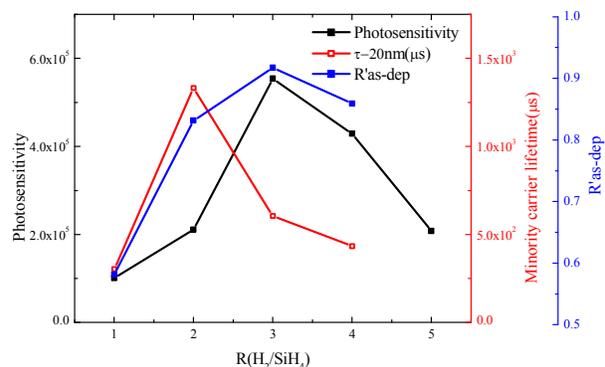


Fig. 1 The lifetimes of the a-Si:H films, the a-Si:H films' microstructure factors' complement of 1 (R'_{as-dep}) with thickness of 20nm deposited on the c-Si substrates and the photosensitivity of the a-Si:H films with thickness of 100nm deposited on the glass substrates as functions of R , (lines are guides to the eye).

onequilibrated network structure at the a-Si:H/c-Si interface, while the interface defect density after annealing is governed more by the bulk properties of the film, due to subsequent hydrogen equilibration.¹⁹ The results from Fig. 1 agree with their conclusion about the passivation at the as deposited state. Therefore, we proposed a hypothesis that both the bulk of the a-Si:H film deposited on the c-Si substrates and the a-Si:H/c-Si interface were important to passivation. Since the process obtaining the best bulk quality of the a-Si:H film may make the epitaxy growth occurs at the a-Si:H/c-Si interface, we must deal with them respectively.

According to the hypothesis mentioned above, we firstly cut the thickness of the a-Si:H films from 20nm to 5nm to further exclude the influence of the bulk a-Si:H film and investigated the a-Si:H/c-Si interface's effect on passivation. Fig. 2 shows the lifetimes of the as-deposited and post-annealing a-Si:H films with thickness of 5nm and 20nm deposited on the c-Si substrates as functions of R . With the increase of R , despite of annealing or not, thick or thin, they all had the similar tendency. That indicated that the a-Si:H/c-Si interface had more dominating influence on the passivation than the bulk of the a-Si:H films. However, through annealing, the lifetimes of the samples passivated by the 5nm thick a-Si:H films decreased when the R is larger than 2 compared to that of the samples at the as-deposited state, which suggested that when the R is larger than 2 the a-Si:H/c-Si interface has grown epitaxy phase, Annealing is detrimental for the passivation in such a case.²⁰ But when the thickness of the a-Si:H film increased to 20nm, all the lifetimes of the samples increased after annealing, indicating the bulk of a-Si:H film had more influence on the passivation of the c-Si when the thickness of the a-Si:H film increased. Indeed, Similar report has been also presented by Yang et al.²¹: even though a deposition condition establishing superior passivation quality with tens of nanometer-thick layers, the passivating behavior totally differs in 10nm thick layers before and after annealing. The explanation for that the high quality passivation could be obtained regardless of the existence of epitaxial layer was the suppression of detrimental effect from epitaxial growth by sufficiently thick, high quality amorphous layer. As the epitaxial growth occurs in early stage of a-Si:H deposition, short deposition time for thin layer led to insufficient

growth of amorphous bed on top of polycrystalline epitaxy. Consequently, the volume fraction of crystalline of the amorphous layer increased with decreasing layer thickness. Therefore the thin samples showed deteriorated passivation quality even the deposition condition once accomplished high quality passivation in thick layer.²¹

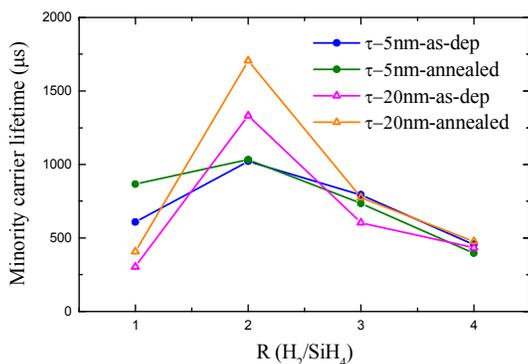


Fig. 2 The lifetimes of the as-deposited and post-annealing a-Si:H films with thickness of 5nm and 20nm deposited on the c-Si substrates as functions of R , (lines are guides to the eye).

Fig. 3 shows the a-Si:H films' microstructure factors (R_{as-dep}) with thickness of 20nm and 5nm deposited on the c-Si substrates as functions of R . With the increase of R , the microstructure factors of the 20 nm thick a-Si:H films firstly decreased and then increased, and when $R=3$, the R_{as-dep} had the lowest value, indicating that the film quality was best. This tendency was similar as that of films deposited on the glass substrates shown in Fig. 1. However, the tendency of the 5nm thick films' R_{as-dep} was obviously different from that of the 20 nm thick films, the R_{as-dep} monotonically increases with the increase of R , indicating that the film's microstructure was becoming more and more uncompact and porous. Therefore, the microstructure of the a-Si:H film closing to the a-Si:H/c-Si interface is different from that of the thicker film, especially with the variation of R . However, by observation of Fig. 2 and Fig. 3, we have found that although the tendency of the bulk quality of the different thick a-Si:H films was different with the variation of R , the tendency of their passivation was similar. It suggested that the amount of SiH_2 and the microstructural defect in the i-layer was not the key factors effecting the passivation quality of the i-layers. U. K. Das et al has got the similar results.²² For further observation and analysis, Table 1 shows the lifetimes and the corresponding microstructure factors of the as-deposited i-a-Si:H films with thickness of 5nm and 20nm deposited on the c-Si substrates as functions of R . At $R=1$, the lifetime of the sample with 20nm was lower than that of the sample with 5nm (303 μs Vs 608 μs). The microstructure factors of the a-Si:H films with thickness of 20nm and 5nm were 0.42, 0.18 respectively. It indicated that the 20nm a-Si:H bulk was less compact than the 5nm a-Si:H bulk. Similarly, at $R=2$, with the larger H_2 dilution than that of $R=1$, the lifetime of the sample with 20nm was higher than that of the sample with 5nm (1330 μs Vs 1021 μs), the 20nm a-Si:H

bulk was more compact than the 5nm a-Si:H bulk (values of the microstructure factors: 0.17 Vs 0.23). The results which presented above indicated that when the a-Si:H/c-Si interface did not occur the epitaxial growth, the bulk quality of the a-Si:H film had obvious influence on the passivation. But at $R=3, 4$, although the 20nm a-Si:H bulks were both compact than the 5nm a-Si:H bulks (values of the microstructure factors: 0.08 Vs 0.56 at $R=3$, 0.14 Vs 0.71 at $R=4$), the lifetimes all decrease with the increase of the thickness. These indicated that when the a-Si:H/c-Si interface occurred the epitaxial growth, the bulk quality of the a-Si:H film had no obvious influence on the passivation. So our hypothesis mentioned above was right: The bulk quality of the a-Si:H film deposited on the c-Si substrates and the a-Si:H/c-Si interface were both important to passivation. The a-Si:H/c-Si interface had more dominating influence on the passivation than the bulk of the a-Si:H film. But when the a-Si:H/c-Si interface did not occur the epitaxial growth, the bulk quality of the a-Si:H film had obvious influence on the passivation.

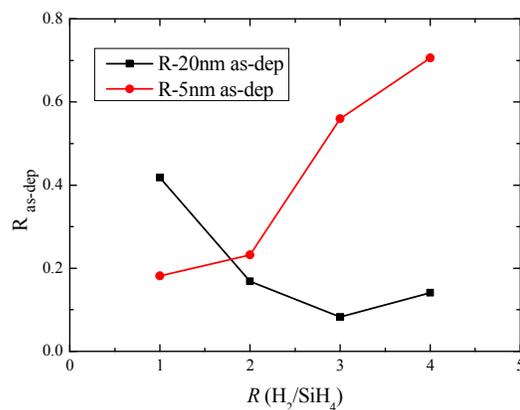


Fig. 3 The a-Si:H films' microstructure factors (R_{as-dep}) with thickness of 20nm and 5nm deposited on the c-Si substrates as functions of R , (lines are guides to the eye).

Table 1: The lifetimes and the corresponding microstructure factors of the as-deposited i-a-Si:H films with thickness of 5nm and 20nm deposited on the c-Si substrates as functions of R .

Hydrogen dilution ratio (R)	20nm-i-a-Si:H layer		5nm-i-a-Si:H layer	
	Lifetime (μs)	Microstructure factor (R)	Lifetime (μs)	Microstructure factor (R)
1	303.17	0.42	608.01	0.18
2	1330.94	0.17	1021.52	0.23
3	604.18	0.08	793.49	0.56

4	433.4	0.14	455.47	0.71
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Sample number	1 st step <i>i</i> -layer		2 nd step <i>i</i> -layer		As-deposited		annealed	
	<i>R</i>	Thick-ness (nm)	<i>R</i>	Thick-ness (nm)	Life-time (μs)	V_{oc-imp} (V)	Life-time (μs)	V_{oc-imp} (V)
1	1	5	3	15	1473.6	0.714	2119.7	0.722
Ref.1	1	20			303.2	0.669	406.5	0.678
Ref.2			3	20	604.2	0.691	774.7	0.697
2	2	2	3	3	1770	0.714	2548.6	0.724
Ref.3	2	5			1021.5	0.700	1032	0.704
Ref.4			3	5	793.5	0.690	734.6	0.690

Table 2: The parameters of the two-step *i*-layer growth process and the relevant results of the samples measured by Sinton Consulting WCT-120.

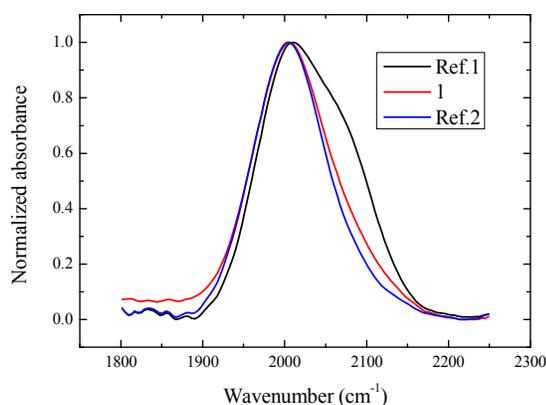


Fig. 4 Normalized FT-IR absorption spectra for post-annealing a-Si:H films of sample 1, ref.1 and ref.2.

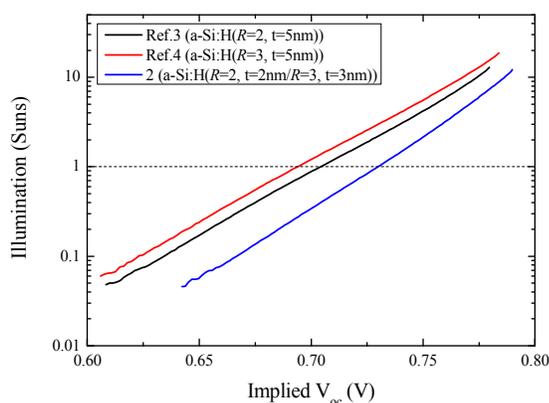


Fig. 5 The implied V_{oc} after post-annealing process for sample 2, Ref.3 and Ref.4 by QSSPC measurements.

Based on the results and conclusion presented above, we proposed a two-step *i*-layers growth process: the purpose of the first step *i*-layer was to make the a-Si:H/c-Si interface abrupt, no epitaxy phase growth, and the second step *i*-layer used the optimum a-Si:H bulk growth process. Table 2 shows the parameters of the two-step *i*-layer growth process and the relevant results of the samples measured by Sinton Consulting WCT-120. Firstly, we chose the 20nm thick a-Si:H film to use the two-step *i*-layers growth process. The R ($R=[H_2/SiH_4]$) of the first step *i*-layer with 5nm thick we adopted was 1, that was because the Fig. 2 showed the lifetime increased after annealing when the R of the 5nm thick a-S:H film was 1, so we can confirm the a-Si:H/c-Si interface was abrupt and did not exist epitaxial growth in such case.²⁰ Meanwhile, the R of the second step 15nm thick *i*-layer we adopted was 3 because the Fig. 1 showed the bulk quality of the a-Si:H film when $R=3$ was the best. As a result, the carrier lifetime was 1473.6μs ($V_{oc-implied}=0.714V$), and after annealing, it was up to 2119.7μs ($V_{oc-implied}=0.722V$). However, the lifetime of reference 1 and 2 which used the single step 20nm thick as-deposited *i*-layer was only 303.2μs ($V_{oc-implied}=0.669V$) and 604.2μs ($V_{oc-implied}=0.691V$), respectively. Even after annealing, they were only up to 406.5μs ($V_{oc-implied}=0.678V$) and 774.7μs ($V_{oc-implied}=0.697V$), respectively. Fig. 4 describes the normalized FT-IR absorption spectra for post-annealing a-Si:H films of sample 1, ref.1 and ref.2. A peak at 2000 cm^{-1} originated from a-Si:H stretching mode in compact material, and the observed peak at 2100 cm^{-1} implied that a-Si:H stretching mode comes from the internal surface of voids in a-Si:H film.²³ This suggested that the samples with a low H_2 -diluted intrinsic layer may have porous structures with many Si dangling bonds inside the a-Si:H film, resulting in high absorbance at 2100 cm^{-1} . So the ref.1 had the poorest film quality, and ref.2 had the best one. However, by using the two-step *i*-layers, sample 1 had

nearly the same film quality as the ref.2 without risking the epitaxy growth at the a-Si:H/c-Si interface. Similarly, with only 5nm thick a-Si:H two-step *i*-layers, the carrier lifetime which was up to 1770 μ s was obtained on double polished Cz-Si wafers, and after annealing, that increased to 2548.6 μ s. They were all superior to those using the single step *i*-layer(ref.3, ref.4). Fig. 5 shows the implied V_{oc} after post-annealing process for sample 2, ref.3 and ref.4 by QSSPC measurements. The implied V_{oc} of sample 2 showed 724mV, which was much higher than 704mV of Ref.3 and 690mV of Ref.4. Thus, these results were accordance with our conclusion present above. In addition, we have also found that when the two-step *i*-layers growth process was used, the passivation quality of thinner film(5nm) was better than that of thicker film(20nm): 1770 μ s Vs 1473.6 μ s at the as-deposited state, 2548.6 μ s Vs 2119.7 μ s after annealing. That demonstrated once again that the a-Si:H/c-Si interface had more dominating influence on the passivation than the bulk of the a-Si:H film. In sum, it was expected that the two-step *i*-layers growth process would provide outstanding surface passivation for the SHJ solar cells, especially when combining with the post-annealing process.

Conclusions

In this work, we investigated the passivation quality of different thickness a-Si:H films deposited on the c-Si substrates with different H₂ dilution and the bulk quality of them deposited both on the glasses and on the c-Si substrates. The results demonstrated that the a-Si:H/c-Si interface had more dominating influence on the passivation than the bulk of the a-Si:H films. But when the a-Si:H/c-Si interface did not occur the epitaxial growth, the bulk quality of the a-Si:H film had obvious influence on the passivation. So we proposed a two-step *i*-layers growth process, with only 5nm thick a-Si:H two-step *i*-layers, the carrier lifetime which was up to 1770 μ s ($V_{oc-implied}=0.714V$) was obtained on polished Cz-Si wafers, and after annealing, that increased to 2548.6 μ s ($V_{oc-implied}=0.724V$). They were all better than those using the single step *i*-layer.

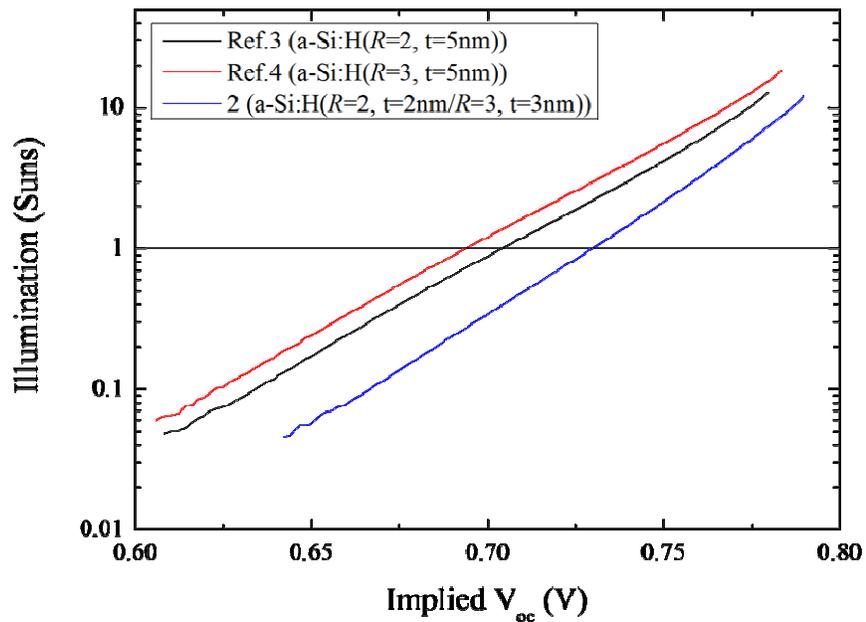
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Notes and references

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The implied V_{oc} of the sample with two-step i-layers was much higher than that using traditional one step i-layer.