

24.18% efficiency TOPCon solar cells enabled by super hydrophilic carbon-doped polysilicon films combined with plated metal fingers



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ABSTRACT

Electroplating technology which has the potential of reducing silver consumption and lowering fabrication costs has been widely used in the photovoltaic (PV) devices. However, the poor wettability of the polysilicon film limits the application of the electroplating technology on tunnel oxide passivated contact (TOPCon) solar cells (SCs). In this work, we propose a carbon (C)-doped polysilicon with highly polar C-Si bonds to improve the surface wettability of polysilicon film. By reducing the hydrogen bubble retention during plating, a uniform, dense, and void-free Ni seed layer can be obtained. To maintain the good conductivity of polysilicon, a double-layer polysilicon structure consisting of a C-doped and a C-free polysilicon films was constructed for TOPCon device fabrication. The new design shows an excellent passivation quality with an implied open-circuit voltage (iV_{oc}) of 745 mV for the lifetime sample and a good contact performance with a low contact resistivity (ρ_c) of 2–3 $m\Omega \text{ cm}^2$ between plated grids and polysilicon films. Finally, the proof-of-concept TOPCon devices with the double-layer polysilicon structure and plated metal electrodes deliver a remarkable power conversion efficiency (PCE) of 24.18%. Therefore, the C-doped polysilicon combined with the electroplating technology demonstrated in this work shows great potential to obtain high-efficiency and low-cost TOPCon SCs in the PV industry.

1. Introduction

High-efficiency and low-cost crystalline silicon (c-Si) solar cells (SCs) have always been the main subject of the photovoltaic (PV) industry. Currently, the high-efficient c-Si technologies include silicon heterojunction (HJT) [1,2], tunneling oxide passivation contact (TOPCon) [3–6], and integrated-back contact (IBC) technologies [7,8]. Among them, TOPCon SC is widely recognized as one of the most promising c-Si technologies for the next-generation PV industry due to its high compatibility with the traditional passivated emitter and rear contact (PERC) SCs in production lines and relatively low capital cost to upgrade the production lines [5]. However, the front- and rear-sided

fingers/electrodes of TOPCon SCs use the high-conductivity silver materials, yielding a large silver consumption and thus an increased material cost [9]. Ample evidence has shown that the supply of silver is a critical factor limiting the upcoming terawatt (TW)-scaled PV market in the next decade [9]. Therefore, developing metallization schemes with low silver consumption is particularly crucial to reduce material costs and accelerate the market expansion of TOPCon SCs.

As a promising alternative metallization technology for the PV industry, copper plating [10–14] has been successfully introduced into the TOPCon devices with the same level of device performance as that of the commonly-used screen-printed silver pastes [15,16]. According to the annual report of International Technology Roadmap for Photovoltaics

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(ITRPV), the copper plating technology could reduce the silver consumption of TOPCon electrodes from 20 mg/W of the screen-printed method to 0.6 mg/W in 2022 [17]. From an industrialization perspective, plating the metal electrodes directly on the polysilicon films is more desirable to fabricate the TOPCon devices as it could avoid using the costly vacuum physical vapor deposition (PVD) to deposit the metal seed layers. However, it is still challenging to adopt the copper plating technology in mass production, which is mainly due to the poor adhesion between plated grids and polysilicon films, leading to reduced device performance and increased risk of peeling and cracking of the plated grids during the subsequent transfer and packaging process [18]. To achieve high-efficiency TOPCon SCs, the rear-side surface of the c-Si substrate is typically textured by a smooth alkali-polished structure to balance the optical and electrical performance with high short-circuit current (J_{sc}) and open-circuit voltage (V_{oc}) simultaneously [19,20]. In addition, the poor wettability of the polysilicon layer is one of the key factors limiting the contact performance of electroplated electrodes, which is due to that the poor wettability makes it difficult for the plating solution to diffuse completely and allows air bubbles to be easily trapped, thus forming voids and making poor electrical properties and adhesion of the plated fingers [21–23].

In this work, we propose innovatively a double-layer polysilicon structure consisting of a thick carbon (C)-free polysilicon and a thin C-doped polysilicon as the electron transport layer for the TOPCon SCs, in which the C-free polysilicon has high conductivity for carrier transport and the C-doped polysilicon has good wettability for plating the Ni seed layer. The experimental results confirm that the C-doped polysilicon shows good wettability with a super hydrophilic contact angle of about 0°. As a result, the uniform, dense, and void-free Ni seed layer was obtained, suggesting good physical contact between polysilicon films and metal electrodes. Moreover, the TOPCon lifetime sample with such a double-layer polysilicon design shows excellent passivation with the highest implied open-circuit voltage (iV_{oc}) of 745 mV and good contact performance with a contact resistivity (ρ_c) of 2–3 mΩ cm². Finally, the proof-of-concept TOPCon SCs with the double-layer polysilicon structure and Ni/Cu/Sn-plated electrodes achieve a high-power conversion efficiency (PCE) of 24.18%, much higher than the C-free devices.

2. Experimental details

2.1. Preparation and characterization of the C-doped polysilicon

The <100> oriented and 170 μm thick *n*-type Czochralski (CZ) silicon wafers with a resistivity of 0.8–2.1 Ω cm were used as the substrates with the purpose of characterization. Both the front and rear surfaces of c-Si wafers were polished by TMAH. An ultrathin SiO_x layer was grown in a concentrated nitric-sulfuric (CNS) acid bath [24] at 60 °C for 10 min after standard RCA cleaning and HF dipping. Subsequently, a 5 nm thick C-doped amorphous silicon (n^+) layer was deposited on both sides of the c-Si substrate using a 13.56 MHz PECVD system with silane (SiH₄), phosphorane (PH₃), methane (CH₄), and hydrogen (H₂) as the reaction gases. The carbon content was controlled by the gas flow ratio (R) of CH₄/(CH₄+SiH₄). The samples were then annealed at 840 °C for 30 min in a quartz tube furnace in a moisture/nitrogen atmosphere [25]. The chemical composition of the C-doped polysilicon was characterized by XPS (AXIS ULTRA DLD). The surface wettability of the C-doped polysilicon samples was characterized by measuring the water contact angle using a contact angle tester (OCA-20).

2.2. Preparation of the double-layer polysilicon structure and TOPCon device

N-type phosphorus-doped CZ crystalline silicon wafers with a thickness of 205 ± 20 μm and a resistivity of 1–7 Ω cm were used for the preparation of solar cells. The front surface was textured by KOH solution to construct the random pyramid structure. Then the boron emitter

was prepared through the high-temperature diffusion of the BBr₃ source, and then the rear surface was polished with alkali to remove the surrounding deposition zone of boron diffusion. After RCA cleaning and HF dipping, a thin silicon oxide film with a thickness of 1.5 nm was grown on the rear side of the c-Si wafer by CNS acid at 60 °C for 10 min [24]. A 25–35 nm phosphorus-doped a-Si:H layer followed by a 5 nm C-doped a-Si:H layer with an R -value of 0.75 was deposited successively using the PECVD system. Subsequently, the samples were annealed at 800°C–860 °C for 30 min for amorphous silicon crystallization and dopant diffusion to form the TOPCon structure. The front side boron diffusion emitters were covered by AlO_x/SiN_x stack by using atomic layer deposition (ALD) and PECVD, respectively. After defining the front electrode pattern by photolithography, the Ti/Pd/Ag seed layer was deposited by electron-beam evaporation and then it was thickened by plating. The pattern of the Ni/Cu/Sn electrode with a line width of 15 μm on the rear side of the devices was defined using photolithography. Then, the Ni layer served as a seed layer, and a barrier layer for Cu was directly plated on the polysilicon film at a current density of 30 mA/cm² for 2 min. The Cu layer was subsequently deposited at 20 mA/cm² for 10 min. The Sn layer was electroless deposited in a 40 °C Sn electroless plating solution for 15 min to protect Cu from being oxidized. Ni and Cu plating solutions and Sn electroless plating solutions were provided by Atotech. Finally, a magnesium fluoride (MgF₂) passivation and anti-reflection layer was deposited on the front side by thermal evaporation.

The sheet resistance (R_{sheet}) of the polysilicon films was measured by a four-point probe meter (Polytec, 4 Dimensions). The ρ_c between polysilicon and plated grids was measured using a semiconductor parameter analyzer (Keithley 4200-SCS) combined with the transmission length method (TLM) [26]. The iV_{oc} , single-sided saturation current density ($J_{0,s}$), and effective minority carrier lifetime (τ_{eff}) were extracted using a quasi-steady-state photoconductance (QSSPC, Sinton WCT-120). The photovoltaic performance of TOPCon SC was measured by a solar simulator (Enlitech, SS-F5-3 A) under AM1.5 (100 mW/cm²) irradiation at 25 °C, and the external quantum efficiency (EQE) and reflectance (Ref.) spectra of the devices were measured by an EQE system (Enlittech, QE-R3011).

3. Results and discussion

The wettability of C-doped polysilicon films at the different R values was first investigated by measuring the contact angles of the distilled water droplets. For the C-free sample (*i.e.*, $R = 0$), the distilled water droplets were difficult to spread, suggesting a high surface tension in the C-free polysilicon film. As a result, a large contact angle of 84 ± 3° was obtained as shown in Fig. 1(a). With the incorporation of C atoms, the wettability of the polysilicon films was significantly improved with the corresponding water contact angles decreasing as shown in Fig. 1(b)–(d). Notably, the polysilicon film with a relatively higher C content (*i.e.*, $R = 0.75$) exhibited super hydrophilicity with the water droplets completely spread on the surface. The wetting behavior of Ni electrolyte on the C-doped polysilicon was also measured, as shown in Fig. 1(e)–(h). As a result, the contact angles of the Ni electrolyte exhibit the same trend as the water, *i.e.*, the contact angles of the Ni electrolyte are negatively correlated with the amount of C doping in the film, suggesting that C doping could effectively promote the diffusion of Ni electrolyte on the polysilicon. Therefore, it can be concluded from these observations that the introduction of C atoms into the polysilicon can effectively improve surface wettability.

To uncover the improved wettability of the C-doped sample, we performed X-ray photoelectron spectroscopy (XPS) measurements to examine the chemical states of the C-doped samples. The XPS spectra of C 1s are shown in Fig. 2, which can be split into three typical peaks corresponding to the C–Si (282.8 eV), C=C (284.8 eV), and C–O–C (286.5 eV) bonds [27]. Compared to the C-free sample in Fig. 2(a), the intensity of C=C and C–O–C bonds of the C-doped sample in Fig. 2(b)

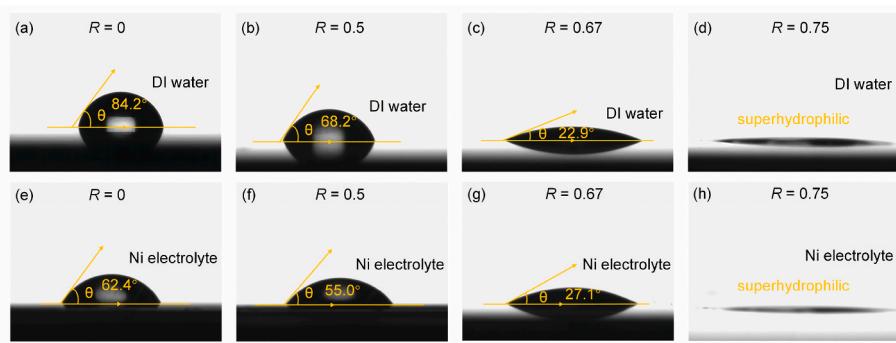


Fig. 1. Images of (a)–(d) a distilled water droplet and (e)–(h) a Ni electrolyte droplet for the samples without C-doping and with C-doping at an R -value of 0.5, 0.67, and 0.75. All samples were annealed at 840 °C.

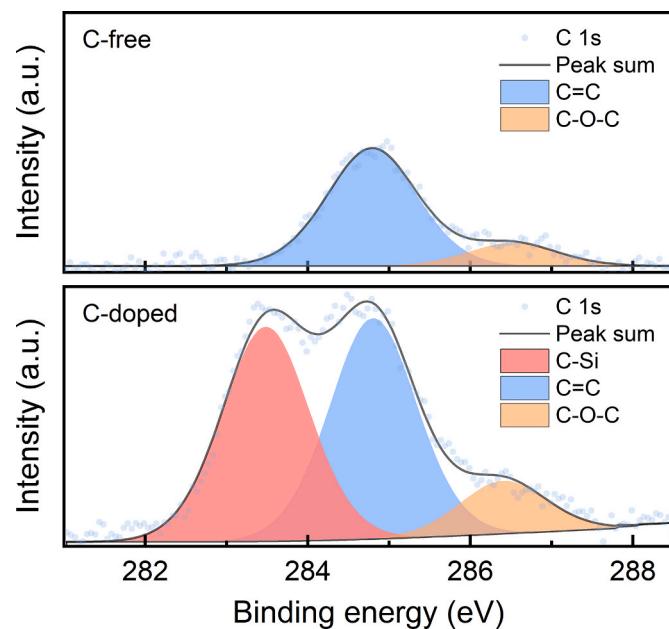


Fig. 2. XPS spectra of C 1s for the (a) C-free and (b) C-doped ($R = 0.67$) samples at 840 °C annealing.

slightly increased. This indicates that only a small part of C=C or C–O–C bonds were introduced by C doping, and most of the C=C or C–O–C bonds originate from the contaminated carbon introduced from the testing environment. However, an additional XPS peak at 282.8 eV that can be assigned to C–Si bonds appears for the C-doped sample, suggesting that C tends to bond with polysilicon to form C–Si bonds. Compared to the nonpolar covalent Si–Si bonds, the C–Si bonds are highly polar, which may be the main reason for the better wettability of the C-doped polysilicon film [28].

The good wettability of the film could ensure the adequate spreading of the plating solution and reduce the surface energy which is conducive to the escape of hydrogen during the plating process to obtain a more uniform and dense seed layer. In this section, the physical morphology of Ni seed layers plated on the C-free and C-doped polysilicon films was characterized by optical and laser confocal microscopy, as shown in Fig. 3(c)–3(f). For the Ni seed layer formed on C-free polysilicon film, a considerable number of voids can be observed, which can be attributed to the non-uniform distribution of the plating solution and the trapping of hydrogen bubbles on the film during the plating process in Fig. 3(a) [21,23]. In contrast, the C-doped sample with better wettability allows the plating solution to spread more evenly and leads to a reduced surface tension, thus making it less susceptible to the retention of hydrogen bubbles on the surface as shown in Fig. 3(b). As a result, a uniform,

dense, and void-free Ni seed layer is formed on the C-doped polysilicon film as can be seen from Fig. 3(d) and (f).

Our previous studies have shown that the presence of C severely degrades the conductivity of the polysilicon films [27], which will in turn affect the uniformity of the current distribution during plating. Herein, we propose a double-layer polysilicon structure as the electron transport layer of the TOPCon SCs, i.e., a C-free polysilicon layer covered by a thin C-doped polysilicon film, which could ensure good wettability and conductivity simultaneously. The following section will systematically investigate the effect of deposition conditions and annealing temperature on the passivation and contact properties of such a structure.

Here, it is worth noting that the introduction of C atoms into polysilicon not only could improve surface wettability but also promote the quality of the polysilicon film. Previous works have confirmed that the presence of C could improve the physical contact of polysilicon/SiO_x/c-Si structure by suppressing the blistering of polysilicon film during high-temperature annealing, maintaining high-quality H passivation by restraining H escape, regulating the diffusion profile of P dopant, and tailoring the energy alignment, which are all beneficial to promote the device performance [27,29–34]. Therefore, using the C-doped polysilicon in this study will also benefit from the abovementioned advantages, and thus achieve the high-efficiency TOPCon SCs. In addition, the improved surface wettability of polysilicon demonstrated in this study also provides an effective way to address the film deposition for other polysilicon-based devices such as perovskite/TOPCon tandem SCs [35, 36].

The passivation quality of lifetime samples with the double-layer polysilicon structure under the different annealing temperatures was comprehensively evaluated, where the structure of the double-layer polysilicon lifetime samples is shown in Fig. 4(a). The passivation quality of the samples with double-layer structure shows the same change trend as that of the C-free polysilicon samples under the various annealing temperatures, which, however, is almost independent of R values as shown in Fig. 4(b)–4(d). This means that the passivation quality of such a structure has a high tolerance with the C content. Meanwhile, we can find that the lifetime samples with the different R values at 840 °C annealing show high-quality passivation with iV_{oc} values > 740 mV, $J_{0,s} < 4$ fA/cm², and the $\tau_{eff} > 3$ ms at the injection level of 10^{15} cm⁻³. In particular, the best passivation with an iV_{oc} of 745 mV, a $J_{0,s}$ of 3 fA/cm², and a τ_{eff} of 4.6 ms is obtained under $R = 0.75$ and 840 °C annealing. These results demonstrate that the double-layer polysilicon structure can achieve excellent passivation quality.

To further examine the electrical properties of the plated Ni/Cu/Sn grid and polysilicon film contact, the ρ_c was evaluated by the TLM. The dark I – V curves of the C-doped and double-layer samples with the different R values are plotted in Fig. 5(a) and (b), respectively. Obviously, all curves exhibit distinct Ohmic behavior, and the variation in the slope of the dark I – V curves of the double-layer structure is insensitive to the R values compared with the C-doped sample. It is worth

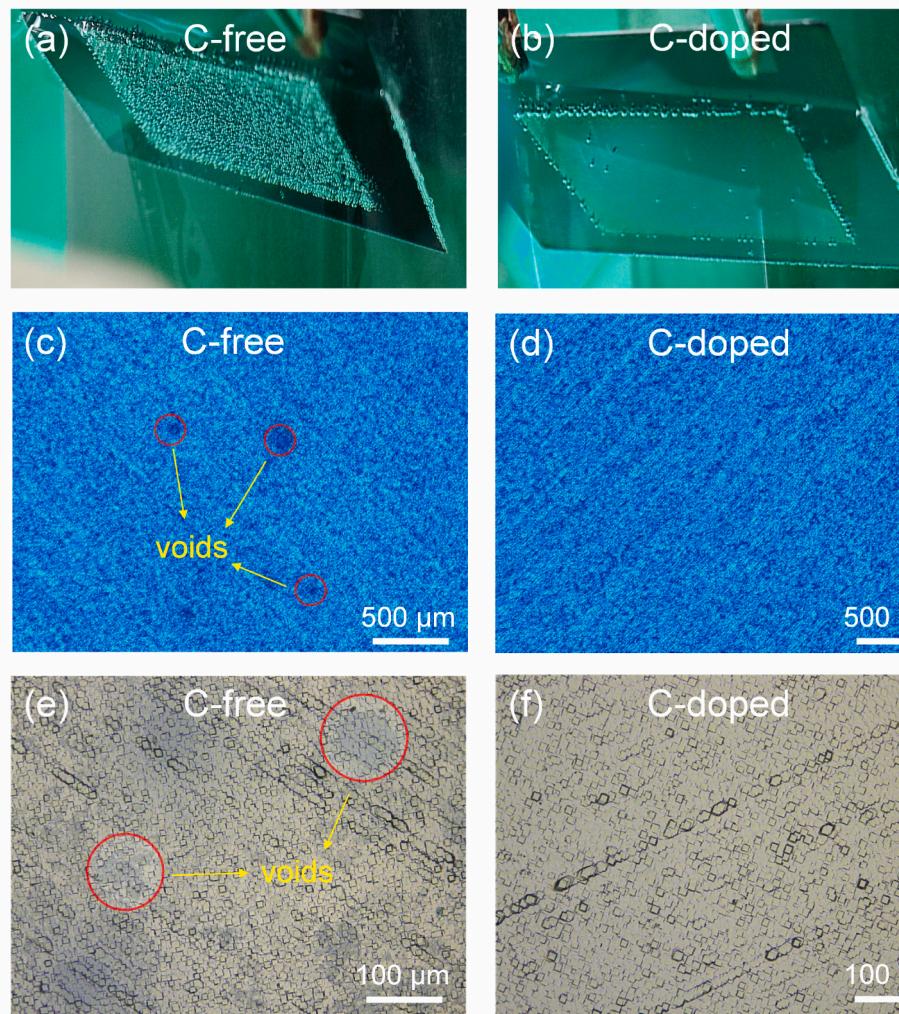


Fig. 3. (a)/(b) Photographs of the electroplating process, (c)/(d) optical microscopy photographs, and (e)/(f) images of the laser confocal microscopes of the plated Ni seed layer for the C-free/C-doped polysilicon samples. The Ni seed layer was electroplated at a current density of 15 mA/cm^2 for 2 min.

mentioning that although the C-doped samples with higher R values could achieve excellent passivation [27], the high resistivity of the C-doped film makes it difficult to be plated. By comparing the ρ_c of the C-doped and double-layer samples, as shown in Fig. 5(c), we can obtain the following conclusions. For the C-doped samples, the ρ_c shows an upward trend as the R increases from 0 to 0.67. This is due to that the presence of C suppresses the activation of P in the polysilicon film [27, 37], leading to a rise in the sheet resistance as shown in Fig. 5(d). In contrast, the double-layer samples show a relatively lower ρ_c , which can be maintained at $2\text{--}3 \text{ m}\Omega \text{ cm}^2$ even though the R -value is high enough (such as $R = 0.75$). It is worth mentioning that sufficiently low ρ_c can be obtained based on the C-free samples as shown in Fig. 5(c), although there may be voids between the plated electrode and the C-free polysilicon due to the poor surface wettability. We speculate that this may be due to the fact that the TLM pattern used in this work with a width of 1 mm is much higher than that of the voids with a size of tens of microns as shown in Fig. 3(c) and (e), these voids thus have less effect of the measured contact current. While grids for rear electrodes with a line width of about $15 \mu\text{m}$ are compared to the size of voids, which may have a negative effect on the device performance. In summary, such a double-layer polysilicon structure shows not only better wettability but also low ρ_c .

Finally, the proof-of-concept *n*-type TOPCon SCs featuring the double-layer polysilicon structure and plated fingers were fabricated. The structure diagram is shown in Fig. 6(a) and the electrical parameters

of TOPCon SCs are shown in Table 1.

It can be seen from the *J-V* curves and the corresponding photovoltaic parameters in Fig. 6(b) that the device performance featuring the double-layer structure is significantly improved compared to that of the C-free device. Typically, the TOPCon SC with the double-layer structure delivers a remarkable PCE of 24.18%, with an open-circuit voltage (V_{oc}) of 707.3 mV, a short circuit current density (J_{sc}) of 42.03 mA/cm^2 , and a fill factor (FF) of 81.34%. In contrast, the C-free device shows a relatively poor PCE of 23.61%, a V_{oc} of 698.8 mV, a J_{sc} of 41.80 mA/cm^2 , and an FF of 80.85%. Here, we find that although the samples based on C-free polysilicon and double-layer structure exhibit the same level of passivation, as shown in Fig. 4, the final device based on the double-layer structure shows better performance. Metallization is well known to reduce the passivation performance of the device due to factors such as metal diffusion, while the polysilicon film for the device in this work is only 30 nm, which makes the V_{oc} more susceptible to metallization. It has been proven that C-doped polysilicon exhibits higher atomic density [37], which may have a better blocking effect on Cu atoms, thereby resulting in a small decrease in passivation performance. Besides, the improved FF may be attributed to the void-free fingers due to the better wettability of C-doped polysilicon. It is worth noting that we obtained a notably higher V_{oc} (*i.e.*, 707.3 mV) by using the plating process to prepare the metal grids, which is much higher than our previous work ($<700 \text{ mV}$) [27,38]. Here, we speculate that this may be due to less metallization damage by plating compared to thermal evaporation or

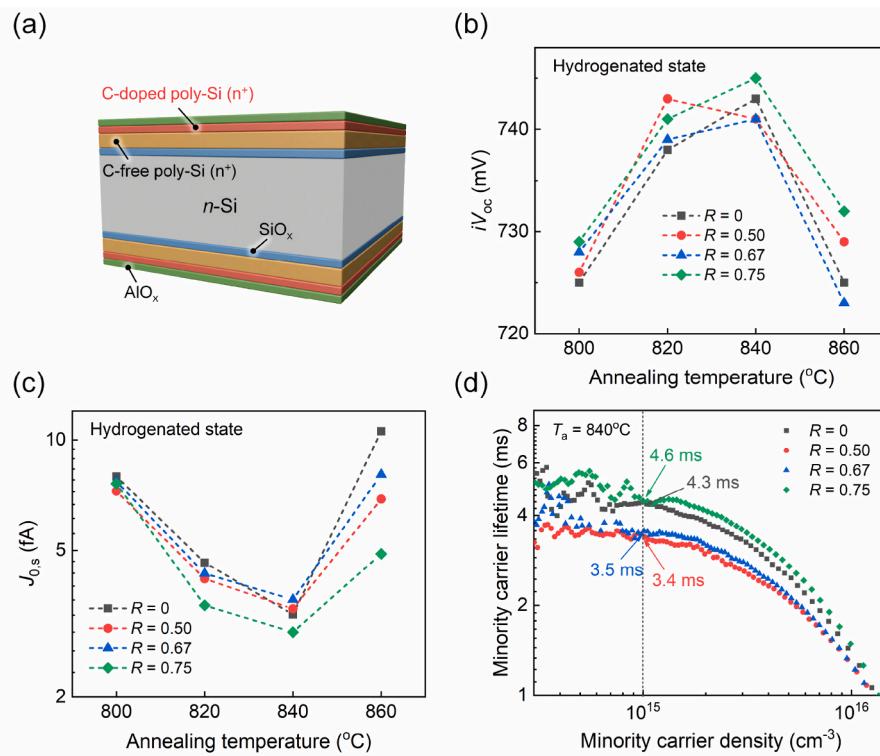


Fig. 4. (a) Structure of the lifetime samples with double-layer polysilicon films, and the corresponding (b) iV_{oc} and (c) $J_{0,s}$ of the samples annealed under the different temperatures and R values. (d) Effective lifetime spectra of the 840°C-annealed samples with different R values.

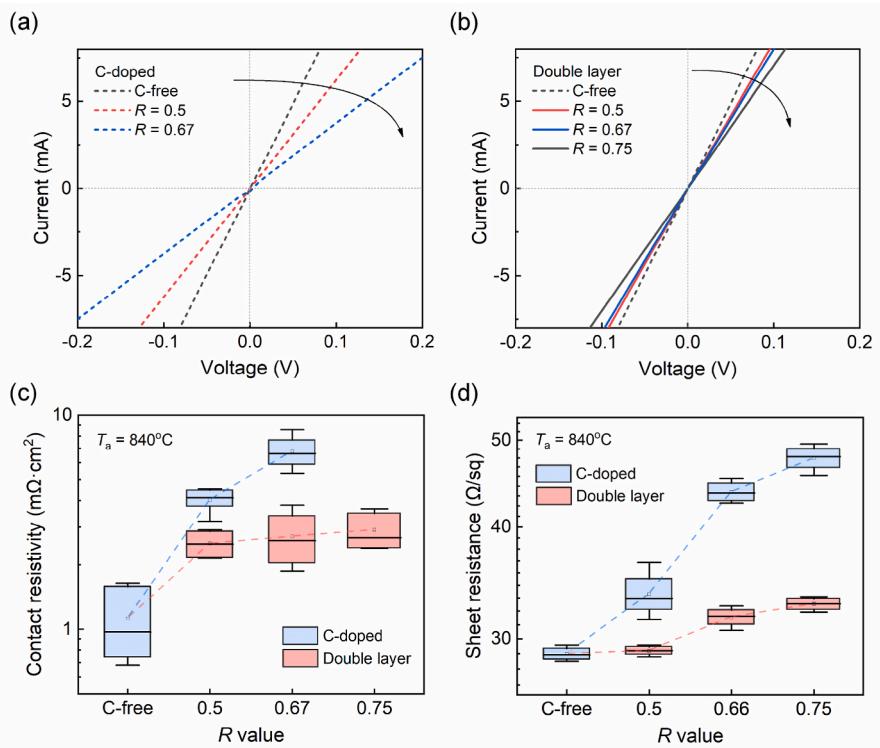


Fig. 5. Dark current-voltage (I - V) curves for the (a) C-doped and (b) double-layer samples with different R values. (c) The ρ_c between the plated Ni/Cu/Sn grid and polysilicon film measured by the TLM, and (d) the corresponding R_{sheet} of the C-doped and double-layer samples with the different R values. All samples were annealed at 840 °C.

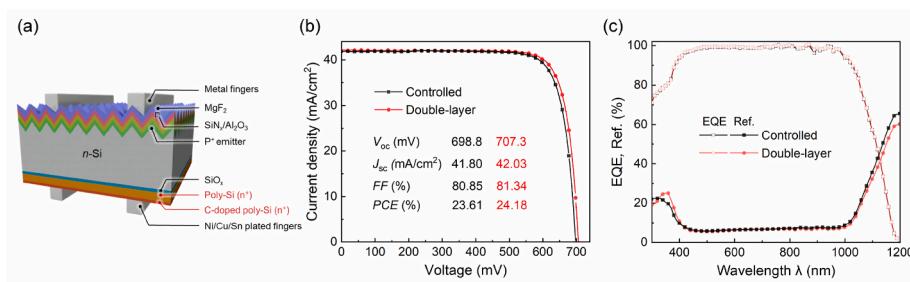


Fig. 6. (a) Schematic diagram of the TOPCon solar cell featuring a double-layer polysilicon structure. (b) Current-voltage (J - V) characteristic curves, and (c) the corresponding EQE and Ref. spectra of the TOPCon SCs.

Table 1

Photovoltaic parameters of the TOPCon SCs featuring the C-free polysilicon and the double-layer polysilicon.

| Samples | V_{oc} (mV) | J_{sc} (mA/cm ²) | FF (%) | η (%) |
|--------------|---------------|--------------------------------|--------------|--------------|
| Controlled | 694.7 ± 4.1 | 41.58 ± 0.22 | 80.50 ± 0.35 | 23.25 ± 0.36 |
| Double-layer | 703.8 ± 3.5 | 41.83 ± 0.2 | 81.18 ± 0.16 | 23.90 ± 0.28 |

electron-beam evaporation. The EQE and Ref. spectra of the devices featuring the double-layer structure and C-free polysilicon are shown in Fig. 6(c), which is consistent with the J - V curves in Fig. 6(b). In a word, the double-layer structure combined with the plated fingers can effectively improve the performance of TOPCon SCs.

4. Conclusion

In this work, we proposed a double-layer polysilicon structure (*i.e.*, C-doped/C-free polysilicon) for TOPCon SCs with the purpose of improving the wettability of the polysilicon films for plating the Ni seed layer while maintaining the contact performance of the plated grids with the polysilicon. Compared with the C-free polysilicon, the C-doped polysilicon shows better wettability, making it easier for hydrogen to release during electroplating. Therefore, a uniform, dense, and void-free Ni seed layer was obtained. By optimizing the deposition conditions and the annealing temperature, samples with the double-layer polysilicon structure show high passivation with a remarkable iV_{oc} of 745 mV. Benefiting from the double-layer polysilicon structure, a low ρ_c between the plated Ni/Cu/Sn grids and the double-layer structure with a value of 2–3 mΩ cm² was achieved. Finally, the proof-of-concept TOPCon devices featuring such a double-layer structure were fabricated with a remarkable efficiency of 24.18%, demonstrating that the double-layer polysilicon structure combined with the plated metal fingers has considerable prospects for application in the manufacturing of high-efficiency TOPCon SCs.

CRediT authorship contribution statement

Haojiang Du: Writing – review & editing, Writing – original draft, Methodology, Investigation, Data curation, Conceptualization. **Taiqiang Wang:** Methodology, Data curation. **Wei Liu:** Investigation, Data curation. **Yali Ou:** Investigation, Data curation. **Mengchao Xing:** Investigation, Data curation. **Weiguang Yang:** Methodology, Investigation. **Jiang Sheng:** Supervision, Conceptualization. **Mingdun Liao:** Supervision, Conceptualization. **Zhijie Gu:** Investigation. **Baojie Yan:** Visualization, Validation, Resources, Conceptualization. **Zhenhai Yang:** Writing – review & editing, Visualization, Validation, Supervision, Resources, Project administration, Funding acquisition. **Yuheng Zeng:** Writing – review & editing, Visualization, Validation, Supervision, Resources, Project administration, Funding acquisition. **Jichun Ye:** Writing – review & editing, Supervision, Resources, Project administration.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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