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Gradient doped nickel oxide hole selective heterocontact and ultrathin passivation for silicon photovoltaics with efficiencies beyond 20%

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ABSTRACT

A prominent feature of photovoltaic devices is their ability to drive photo-generated charges to pathways of asymmetrical conductivity, guiding them to the corresponding electrodes. Under this guideline, the crystalline silicon (c-Si) solar cells dominate the photovoltaic (PV) market for decades and several optoelectronic losses and technological limitations are gradually emerging by now. To circumvent these issues, here, we replaced the doped-silicon emitter layer with a gradient copper-doped nickel oxide (Gd-NiO_x), and incorporated a silicon oxide passivation (SOP) layer at the Gd-NiO_x/c-Si interface, to construct the novel c-Si PV devices. Interestingly, it was observed that the Gd-NiO_x hole selective layer can strengthen the built-in field by minimizing the front electrode/hole-selective layer barrier width and the SOP can reduce interface recombination simultaneously, thereby yielding a remarkable 20.3% efficiency for the proof-of-concept device. We believe the design proposed in this may be of interest for applications in PVs and other optoelectronic devices.

1. Introduction

Solar photovoltaics (PV) has gradually developed from a potential energy technology, which was heavily subsidized by governments, into a green industry with a certain market competitiveness. [1–4] Crystalline silicon (c-Si) solar cells are the main products in PV market, accounting for >90 % of the market-share [5]. As a new member of the c-Si PV family, transition metal oxides (TMOs)/c-Si heterojunction solar cells are considered promising because of their unique optical and electrical properties [6–9]. The charge selective layer of TMOs has a wider optical band gap (>3.0 eV) and less doping concentration than conventional diffusion-doped c-Si solar cells, yielding low optical parasitic absorption loss. Due to this advantage, the theoretical power conversion efficiency (PCE) of TMOs/c-Si solar cells is higher than that of diffusion-doped c-Si solar cells [10]. Correspondingly, different kinds of TMOs materials with high work function (WF) have been studied, such as Cu₂O, MoO₃, V₂O₅, and WO₃, etc. [11–20]. The WF difference between c-Si and TMOs

induces a decent built-in electric field to separate photogenerated charges and effectively extract the charges to an external circuit. Quite recently, a p-type nickel oxide (NiO_x) with various dopants (such as Cu, Li, Ag, and Co, etc.) and high WF has been used as hole selective layer (HSL) [21–24]. The doped NiO_x possesses wide bandgap and high electrical conductivity ($\sim 10^{-3}$ – 10^{-1} S cm⁻¹), it therefore can make a large quasi-Fermi level splitting with the device and enable a high potential-PCE.

However, at present, research on NiO_x/c-Si solar cells (NSSCs) is still in its infancy, and the latest reported record PCE is only 15.2 % [25]. According to the physical model of the device and the theoretical analysis of carrier behavior, the main PCE loss can be traced to an insufficient built-in field within the device and to defects-induced interface recombination losses. The built-in field of NSSCs is greatly dominated by the dopant profile of the NiO_x HSL, and the interfacial defects are mainly originated from the inherent dangling bonds on c-Si surface and the lattice mismatch of NiO_x/c-Si interface. Therefore, an

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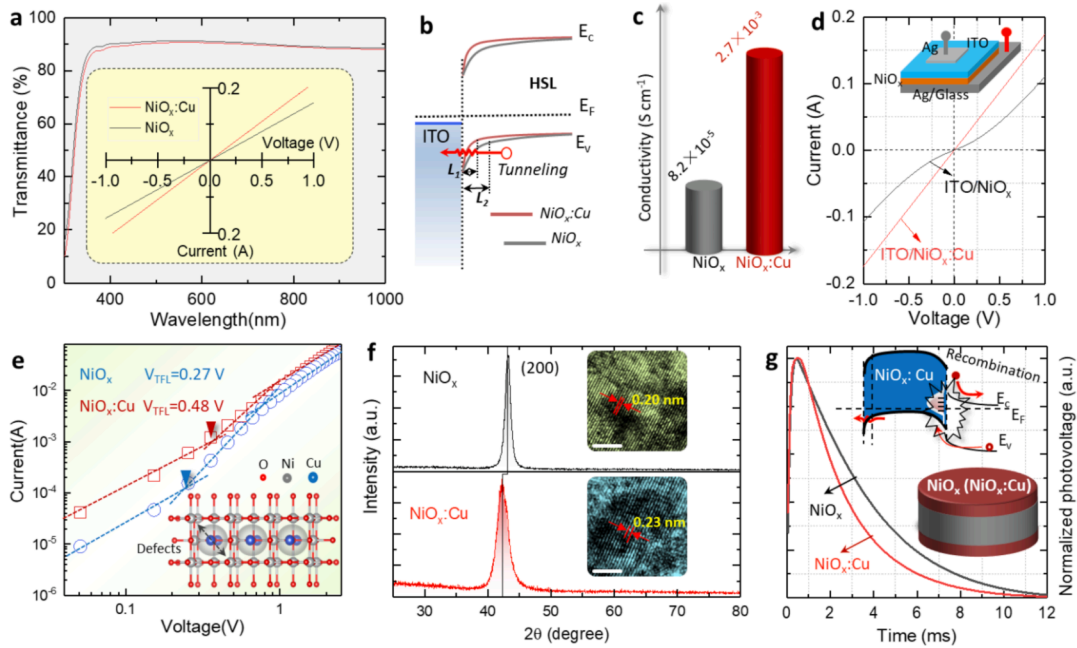


Fig. 1. Key properties of the 50 nm NiO_x and 50 nm $\text{NiO}_x\text{:Cu}$ films. (a) The optical transmittance and the conductivity of the films. (b) Schematic diagram of the ITO/HSL potential barrier impacts on the charge tunneling. The L_1 and L_2 are the distinct barrier widths within the NiO_x and $\text{NiO}_x\text{:Cu}$ HSLs, respectively. (c) The ITO/HSL interface charge transportation behavior obtained from I to V measurement. Inset: the device setup for measuring. (d) The SCLC measurement for NiO_x and $\text{NiO}_x\text{:Cu}$ films. The inset shows the Cu-doping induced lattice expanding, which creates the point defects within the films. (e) The XRD patterns and corresponding HRTEM images of the films. (f) The photovoltage decay curves of the $\text{NiO}_x/\text{c-Si}/\text{NiO}_x$ and $\text{NiO}_x\text{:Cu}/\text{c-Si}/\text{NiO}_x\text{:Cu}$ devices.

accurate design of the doping profile of the NiO_x HSL and effective passivation of the $\text{NiO}_x/\text{c-Si}$ heterointerface are likely to be decisive factors for the development of NSSCs.

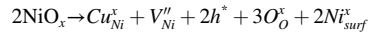
We propose two strategies to address the aforementioned issues: First, we prepared a gradient Cu-doped NiO_x (Gd-NiO_x) HSL using magnetron sputtering, because it possesses higher hole conductivity than the intrinsic NiO_x thin film and can also overcome the negative barriers (indium thin oxide (ITO)/p- NiO_x) and substantially reinforce the built-in field of NSSCs. Second, an ultrathin silicon oxide passivation (SOP), achieved by *in situ* growing an ultrathin silicon oxide (SiO_x) on a c-Si surface, has been implemented through gas-assisted wet-chemical oxidation (GWCO). SOP was previously shown to be effective for eliminating interface defects and pushing the *PCE* of the tunneling oxide passivation contact (TOPcon) c-Si up to 26.0 % [26]. Moreover, by using the SOP strategy, we also achieved high efficiency silicon nanowire heterojunction solar cells, demonstrating the effectiveness for interface passivation [27].

In this work, the synergy of combining Gd-NiO_x and SOP effectively improves the interfacial electric field, passivates the interface defects, enhances the photogenerated charge transfer, and subsequently increases the record *PCE* of the NSSCs to 20.3 %. Through a comprehensive characterization, we also report comprehensive insights into the microstructure and the optoelectronic properties of the Gd-NiO_x HSL. In addition, we also demonstrate the delicate $\text{NiO}_x/\text{c-Si}$ interfacial charge behavior (recombination, transfer, and tunneling transport, etc.), which is promising for further improving advanced Si-based photovoltaic devices.

2. Results and discussion

We first give a glimpse of the basic properties of the 50 nm intrinsic NiO_x and ~ 5 at% Cu-doped NiO_x ($\text{NiO}_x\text{:Cu}$). Fig. S1 shows the roughness of the $\text{NiO}_x\text{:Cu}$ film is slightly larger than that of the dopant-free NiO_x film. X-ray photoelectron spectroscopy (XPS) was adopted to study the surface composition and chemical states of the films surface. Fig. S2 shows the XPS patterns of the Ni 2p core levels of the films. Four

distinct Ni-O composites including Ni, NiO , Ni(OH) , and Ni_2O_3 , can be extracted by deconvolution using the Lorentzian-Gaussian function, which indicates the non-stoichiometric nature of the films [24,28]. However, the Ni(OH)_2 peak at the binding energy of 855 eV rises as the Cu doping, implying defective nickel oxides with hydroxyl groups absorbed onto the films. Fig. 1a shows the optical transmittance and the dark current-voltage (I - V) curves of the films. With Cu doping, the optical transmittance slightly decreases yet the conductivity apparently improves, which may stem from the microstructure changes. We know the ionic radius of Cu is slightly larger than that of Ni, which implies that the direct relevance between Cu doping and charge concentration is linked to the generation of Ni vacancy (V_{Ni}''): [29,30].



The V_{Ni}'' and excess holes ($2h^+$) enhance the p-type doping for the $\text{NiO}_x\text{:Cu}$ films, which is beneficial for improving the conductivity of the films. In addition, according to Schottky's theory [31,32], in silicon heterojunction solar cells, the Fermi-level difference between the HSL and ITO will create a reversed potential barrier in the HSL (non-ohmic contact) against the built-in potential of the device and hinder the carrier transportation (Fig. 1b). We have shown that doping the HSL is an effective way to solve this problem by narrowing this barrier so that making the holes pass through the barrier by a tunneling effect [33]. Fig. 1c shows the conductivity of the NiO_x and $\text{NiO}_x\text{:Cu}$ films obtained by four-point probe measurement. The conductivity the $\text{NiO}_x\text{:Cu}$ ($2.7 \times 10^{-3} \text{ S cm}^{-1}$) is much higher than that of the NiO_x ($8.2 \times 10^{-5} \text{ S cm}^{-1}$). The contact properties between HSL and ITO electrodes are recorded by monitoring the J - V character with an Ag/ITO/ NiO_x (or $\text{NiO}_x\text{:Cu}$)/Ag/Glass structure (Fig. 1d). For the intrinsic NiO_x device, a rectification characteristic can be observed, indicating a non-ohmic contact between ITO and NiO_x . In contrast, when the HSL was substituted by $\text{NiO}_x\text{:Cu}$, a liner J - V character was observed, which indicates that Cu doping offsets the negative barrier and forms a superior ohmic contact between ITO and $\text{NiO}_x\text{:Cu}$.

However, doping within guest materials usually decreases the

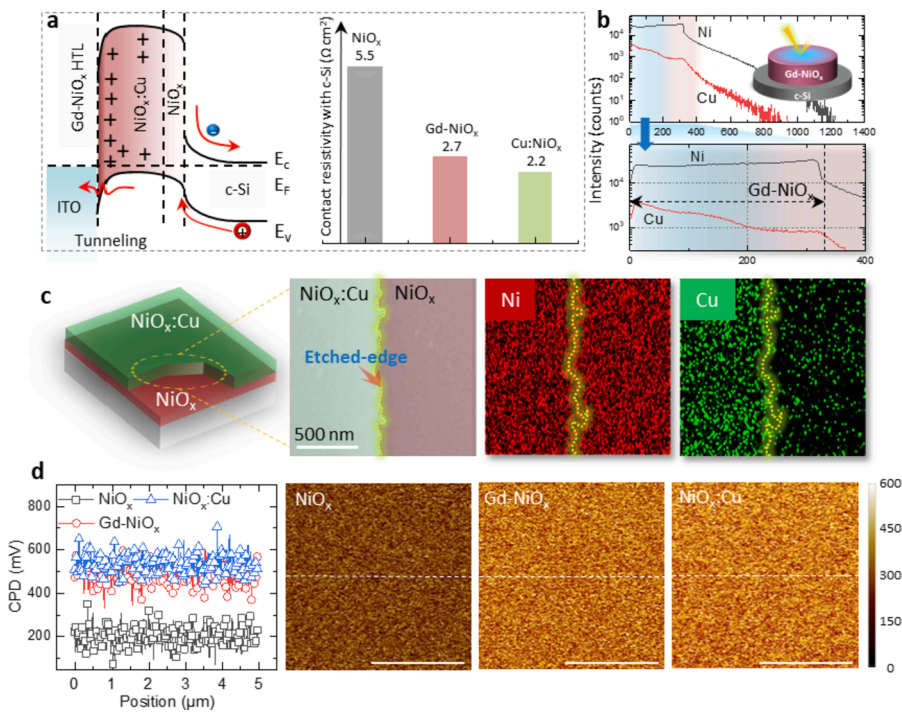


Fig. 2. Realization of the upgraded Gd-NiO_x HSL. (a) The schematic diagram of the energy levels of the ITO/Gd-NiO_x/c-Si structure. The contact resistivity between different kinds of NiO_x and c-Si. (b) The SIMS curves of the Ni and Cu within in the Gd-NiO_x films. (c) The SEM of the post-etched edge, and the EDS maps of the Ni and Cu. The dash lines were used to mark the edge. (d) The CPD values and the SKPM of the NiO_x, Gd-NiO_x, and NiO_x:Cu films, respectively. The scale bar represents 2.5 μm.

crystallization of the host materials by distorting the original lattice and forming extra traps within the host materials [34]. We examined the trap density (N_t) of the films by using space-charge limit current (SCLC) with a hole-only structure of Glass/ITO/PEDOT:PSS/NiO_x (or NiO_x:Cu)/PEDOT:PSS/Ag (Fig. S3). The corresponding J - V curves and the trap-filling limit voltage (V_{TFL}) were displayed in Fig. 1e. The yielded N_t for NiO_x and NiO_x:Cu films are $5.29 \times 10^{14} \text{ cm}^{-3}$ and $3.15 \times 10^{15} \text{ cm}^{-3}$, respectively, which shows that Cu doping increases trap states in the NiO_x film. Fig. 1f shows the X-ray diffraction (XRD) patterns of the NiO_x and NiO_x:Cu films. The peak located at $\sim 42.6^\circ$ is assigned to the cubic (200) crystalline planes. After doping with Cu, the full width at half maximum (FWHM) increases from 1.07° (NiO_x) to 2.09° (NiO_x:Cu) since the divalent Cu with a large atomic radius breaks the original stoichiometric ratio and expands the lattice network of the NiO_x films [35]. The slight low-angle-shift of the (200) peak after Cu doping also confirms this interpretation, which indicates that the lattice constant expands according to Bragg's formula [36]. Also, high-resolution transmission microscopy (HRTEM) (inset of Fig. 1f) shows that the lattice constant of (220) crystal plane increases from 0.20 to 0.23 nm after Cu doping. Therefore, the design of a homogeneous Cu doping NiO_x HSL may be a double-edged sword for NSSCs: On one hand, it improves the conductivity and narrows the negative ITO/HSL barrier, on the other hand, it also interrupts the matrix lattice of the NiO_x and elevates the trap density, which may cause an increment of the traps-assisted recombination at the NiO_x/c-Si interface. Fig. 1g shows the surface photovoltage decay of the c-Si (n -type FZ wafer, (100), $1\text{--}3 \text{ } \Omega \text{ cm}$, $200 \pm 10 \text{ } \mu\text{m}$ thickness) covered by NiO_x and NiO_x:Cu. The measurement setup is NiO_x/c-Si/NiO_x and NiO_x:Cu/c-Si/NiO_x:Cu. Usually, the p -type layer covering the n -type c-Si can selectively separate electrons and holes and lead to a rise in surface photovoltage. In this case, the high conduction band offset may also repel the electron from the surface and reduce the charge recombination [37–39]. However, in this case, we found that the NiO_x:Cu device has faster photovoltage decay than the NiO_x device, implying that the trap states within NiO_x:Cu will trigger significant interface recombination [40].

Therefore, to fully leverage the advantages of the NSSCs, the interface recombination and the negative ITO/HSL barrier must be overcome simultaneously without offsetting each other. The gradient doping

process can change the longitudinal doping concentration of the thin film by controlling a certain variable in the film deposition process, realizing a doping concentration from heavy to light. Herein, a gradient-doped NiO_x HSL (Gd-NiO_x), which consist of 15 nm NiO_x:Cu and 5 nm NiO_x, was proposed to synergistically regulate the energy level structure of ITO/HSL and suppress the recombination at the HSL/c-Si heterointerface (Fig. 2a). The contact resistivity between different types of NiO_x (20 nm) and n -type c-Si was measured by a transfer length method (by TLM-SCAN⁺, PV-tools GmbH). The contact resistivity of NiO_x/c-Si, Gd-NiO_x/c-Si, and NiO_x:Cu/c-Si are $5.5 \text{ } \Omega \text{ cm}^2$, $2.7 \text{ } \Omega \text{ cm}^2$, and $2.2 \text{ } \Omega \text{ cm}^2$, respectively, which shows that doping the NiO_x can effectively reduce the contact resistivity by narrowing the inversion region at the heterointerface. Fig. 2b shows the element depth profile of the Gd-NiO_x films recorded by secondary-ion mass spectrometry (SIMS). The recorded profile shows that Ni is homogeneously distributed in the whole Gd-NiO_x films while the distribution of Cu is gradually reduced along the depth direction, indicating a gradient Cu concentration within the Gd-NiO_x films. To further verify the gradient structure, we prepared thick ($\sim 50 \text{ nm}$) Gd-NiO_x films with identical NiO_x:Cu/NiO_x ratio and partially etched the surface NiO_x:Cu to expose the bottom NiO_x films. The energy-dispersive spectra (EDS) of the etched-edge show the Ni is homogeneously distributed in the films, yet the Cu is remarkably reduced in the bulk of the Gd-NiO_x film (Fig. 2c), indicating a gradient distributed Cu element.

Apparently, the different Cu doping profile will inevitably affect the built-in field of the device. Fig. 2d exhibits the contact potential difference (CPD) and the scanning Kelvin probe microscopy (SKPM) of the different NiO_x films grown on n -type c-Si substrates under light irradiation. The intrinsic NiO_x film generates an average CPD of 247 mV, which is much lower than that of the Gd-NiO_x (522 mV) and NiO_x:Cu (565 mV), respectively. Under light excitation, since the photoexcited holes can be extracted from the c-Si substrate to NiO_x HSL, the CPD enhancement of NiO_x:Cu and Gd-NiO_x films, which is caused by an increase of hole concentration on the surface, indicates a stronger built-in field than the intrinsic NiO_x film [41]. This result is consistent with the aforementioned expectation.

With these results in hand, we prepared the NSSCs with a structure of Ag grid/ITO/Gd-NiO_x/ n -c-Si/ n^+ -c-Si/Al doped zinc oxides (AZO)/Ag

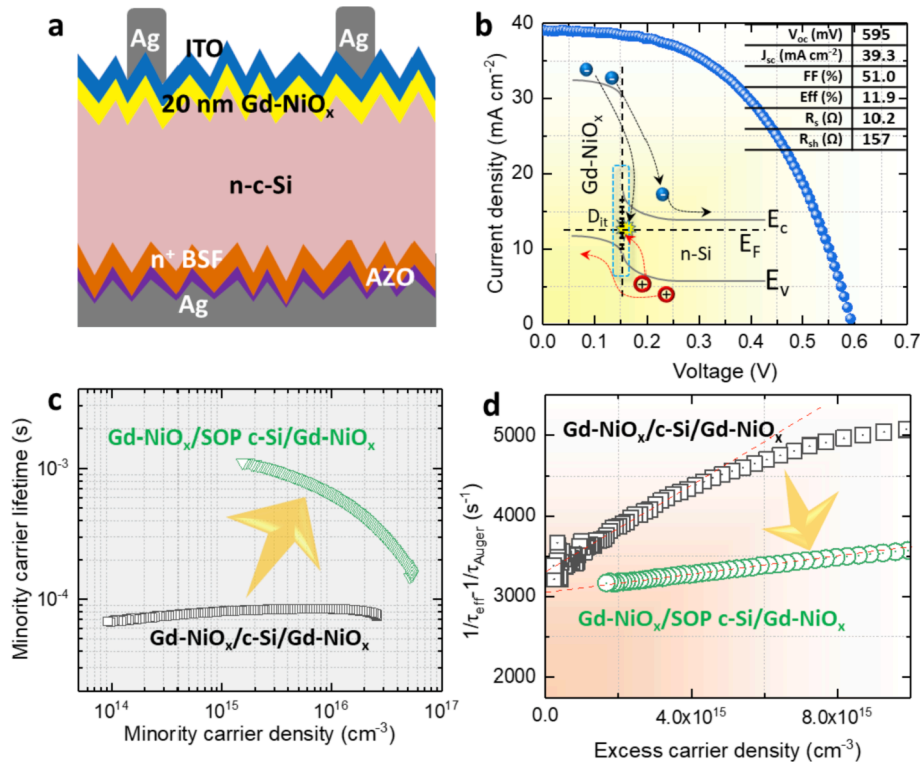


Fig. 3. The photovoltaic performance of the pristine NSSCs. (a) Schematic diagram of the NSSCs structure. (b) The illuminated J - V curves of the device and output parameters. The inset shows the electrical losses caused by interface recombination. The excess carrier density vs (c) the minority carrier lifetime and (d) the $1/\tau_{\text{eff}}/1/\tau_{\text{Auger}}$ of the device with- and without-SOP treatment, respectively.

electrode (Fig. 3a-b). A PCE of 11.9 % (device area: 4 cm^2) was achieved with an open circuit voltage (V_{oc}) of 595 mV, a fill factor (FF) of 51 %, and a short circuit current density (J_{sc}) of 39.3 mA cm^{-2} . The low V_{oc} and FF suggest a significant recombination-induced electrical loss in the devices. However, the V_{oc} and FF are still higher than that of the device

with dopant-free NiO_x HSL (Fig. S4), which may be ascribed to the lower resistivity of the Gd-NiO_x HSL and a better Ohmic-contact at the ITO/HSL interface.

Compared with $\text{NiO}_x\text{:Cu}$ HSL, although the Gd-NiO_x HSL could reduce the HSL/c-Si hetero-interface trap states by suppressing the

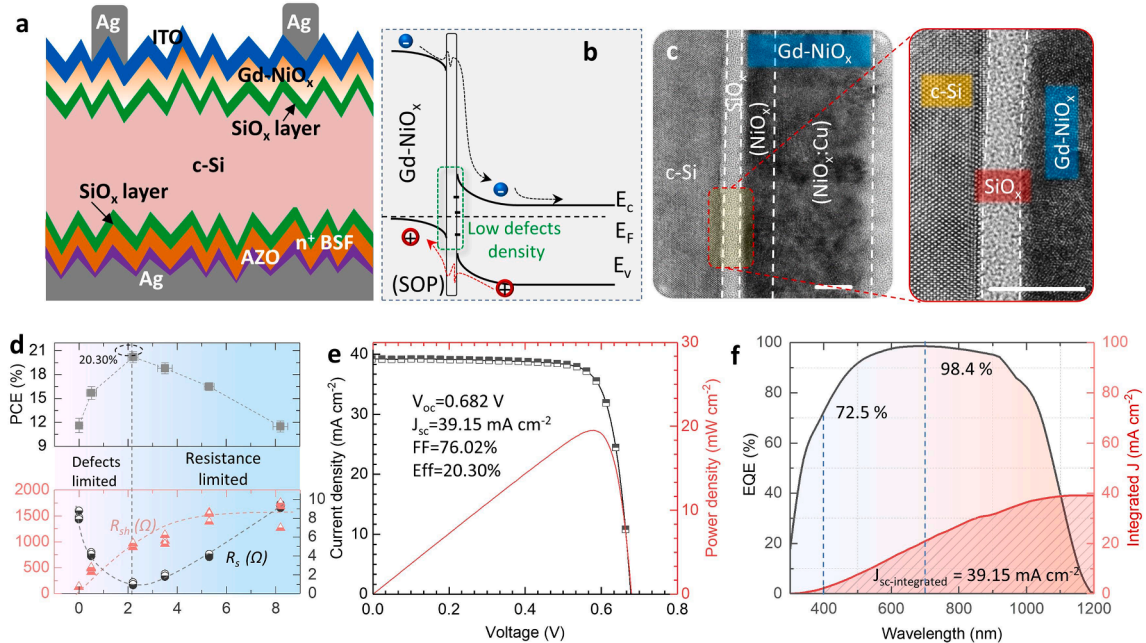


Fig. 4. The upgraded NSSCs. (a) Schematic diagram of the upgraded NSSCs structure. (b) The energy levels of the SOP-based NSSCs. (c) The cross-sectional HRTEM images of the c-Si/ SiO_x /Gd- NiO_x profile. The scalebar is 5 nm for each figure. (d) The PCE variation as a function of the SiO_x thickness. (e) The illuminated J - V , output power density, and corresponding photovoltaic parameters of the NSSCs. (f) The EQE spectra of the NSSCs. The spectrum response at 400 and 700 nm are 72.5 % and 98.4 %, respectively, and the integrated current density is 39.15 mA cm^{-2} .

native Cu_{Ni}^x interstitial defects, the inherent dangling bond on the c-Si surface and the ions bombardment for c-Si during the Gd-NiO_x deposition inevitably produce extra defects (Fig. S5). Further passivating these defects by inserting a media layer, for example hydrogenated amorphous silicon (a-Si:H) or silicon oxide (SiO_x), into the HSL/c-Si interface has been shown to be an effective approach to minimize the electrical losses [42,43]. Previously, we investigated both a-Si:H and SiO_x passivation for TMOs/c-Si solar cells, and found that a SOP can effectively terminate the surface traps by using a simple and low-cost process [27,33,42].

Therefore, we introduced the SOP in NSSCs by the GWCO process and optimized the technique for achieving high photovoltaic performance (the detailed illustration is depicted in Fig. S6 and Supplementary Note 3). As shown in Fig. 3c, we made bifacial passivated devices with and without-SOP to examine the interface recombination by the quasi-steady-state photoconductance (QSSPC) technique (WCT-120, Sinton). The device with SOP shows a minority carrier lifetime of 1.37 ms compared with the control devices of 0.08 ms (at excess carrier density $\sim 2 \times 10^{15} \text{ cm}^{-3}$), indicating an effective surface passivation. We also qualitatively study the defects density of the devices before and after SOP treatment by deducing the QSSPC results (Fig. 3d). The SOP device shows a smaller slope of the linear fit minority carrier density *versus* the $1/\tau_{\text{eff}} - 1/\tau_{\text{Auger}}$ curve than the non-passivated device (the τ_{Auger} and τ_{eff} are the Auger lifetime and effective lifetime, respectively), indicating a lower defect density at the passivated device (Supplementary Note 5) [44]. The decrease of interface defects reduces recombination and improves the electrical properties of the devices. The implied V_{oc} of the devices, which are the theoretical maximum V_{oc} values at one-sun illumination, are correspondingly enhanced from 624 mV to 714 mV (Fig. S7).

Subsequently, as shown in Fig. 4a, we upgraded NSSCs with Gd-NiO_x HSL and SOP treatment to construct a positive built-in field and suppress the Gd-NiO_x/c-Si interface recombination loss simultaneously. The Gd-NiO_x/c-Si bandgap alignment was depicted in Fig. 4b, according to data extracted from Fig. S8 (ultraviolet photoelectron spectroscopy, absorption spectra). It shows a narrow energy gap ($\sim 0.63 \text{ eV}$) between the valence band of Gd-NiO_x and the conduction band of c-Si. Therefore, if there is no SiO_x passivation layer, the narrow energy gap and the large amounts of traps at Gd-NiO_x/c-Si interface will cause a major trap-assisted tunneling recombination [45], which means the electrons and holes may tunnel through the energy gap and recombine at the trap centers. Fortunately, the aforementioned SOP could effective suppress these electrical losses by terminating the interface traps and isolating the valence band of Gd-NiO_x and the conduction band of c-Si. We performed cross-sectional HRTEM to reveal the interface micro-structure and a hierarchical profile (c-Si/ $\sim 2 \text{ nm}$ SiO_x/ $\sim 20 \text{ nm}$ Gd-NiO_x) can be observed (Fig. 4c). The boundary between NiO_x and NiO_x:Cu is not very clear, but one can see the $\sim 5 \text{ nm}$ NiO_x grown on SiO_x is denser than the NiO_x:Cu, which may be related to the Cu doping induced microstructure changes (as mentioned in Fig. 1). However, based on our experience, the SiO_x thickness is a very sensitive factor to determine device performance because both the passivation effects and the carrier tunneling transportation are directly related to the SiO_x thickness. A typical variation of the PCE, series resistance (R_s), and shunting resistance (R_{sh}) as a function of the SiO_x thickness are shown in Fig. 4d and Table S1 (the thicknesses in this case are inferred by the HRTEM test. A $\sim \pm 0.2 \text{ nm}$ error was added on these data). With the thickness increasing from 0 to 8 nm: the PCEs firstly increases to the top value (at $\sim 2 \text{ nm}$) and then exhibit a sharply decrease; the R_s first decreases to the lowest value (at $\sim 2 \text{ nm}$) and increase, while the value of R_{sh} continues to increase. Clearly, the phenomena infer that device performance is transformed from defects-limited region ($< 2 \text{ nm}$) to resistance-limited region ($> 2 \text{ nm}$) due to the trade-off between passivation effects and carrier transport. It is worth noting that in the case of present TOPCon cells (i.e., polySi/SiO_x), thicknesses $\geq 2 \text{ nm}$ would need a high temperature step to break up the oxide and allow conduction through pinholes. We here also found the 2

nm thickness is slightly larger than our previously optimized thickness for the MoO_x/c-Si solar cells ($\sim 1.5 \text{ nm}$) [27]. We speculate that this abnormal thickness may be attributed to the fact that Gd-NiO_x HSL in this study was prepared by magnetron sputtering, which has a stronger ions bombardment process for the substrates compared to previously used thermal evaporation. Moreover, the Gd-NiO_x has higher conductivity than MoO_x, thereby the output voltage losses may be lower than that of the MoO_x/c-Si devices under a similar output current density. Therefore, the optimal SiO_x thickness may be increased to meet the passivation requirement without sacrificing the charge transporting efficiency.

Finally, a high PCE of 20.3 % was achieved for NSSCs (Fig. 4e). The external quantum efficiency (EQE) has also been measured and an integrated current density of 39.15 mA cm^{-2} was obtained. The NSSCs exhibit high EQE values in the short and medium wavelength region (72.5 % at 400 nm; 98.4 % at 700 nm) due to the wider bandgap of the Gd-NiO_x and the excellent interface passivation, which minimizes the optical parasitic absorption and the electrical recombination losses.

3. Conclusion

In summary, we reported the conceptual design of advanced NSSCs, featuring a gradient doped HSL and tunneling oxides passivation contact. The Gd-NiO_x HSL developed in this work reduces the negative ITO/HSL barrier and suppresses the defects density at the Gd-NiO_x/c-Si hetero-interface simultaneously, thereby strengthening the built-in field for charge extraction and reducing the interface recombination losses, respectively. Moreover, the SOP realized by GWCO further eliminates the c-Si surface defects, which improves the electrical properties of the Gd-NiO_x/c-Si interface. Ultimately, we achieved a PCE of 20.3 % for these NSSCs. By further optimizing the back-surface-field and the architecture of the NSSCs, we believe this kind of device could be the next trend in the silicon-based PV market.

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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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.cej.2022.138060>.

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