

## 23.5%-efficient heterojunction silicon solar cell using Molybdenum Oxide as hole-selective contact

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Keywords: solar cell, silicon, heterojunction, molybdenum oxide, carrier-selective contacts, dopant free, thickness

### Abstract

Interest in silicon heterojunction solar cells is growing due to their manufacturing simplicity and record efficiencies. A significant limitation of these devices stems from parasitic absorption in the amorphous silicon layers. This can be mitigated replacing the traditional (p) and (n) doped amorphous silicon selective layers by other materials. While promising results have been achieved using molybdenum oxide ( $\text{MoO}_x$ ) as front-side hole-selective layer, charge transport mechanisms in that contact stack have remained elusive and device efficiencies below predictions. By carefully analyzing the influence of the  $\text{MoO}_x$  and intrinsic a-Si:H thicknesses on current-voltage properties, we discuss transport and performance-loss mechanisms. In particular, we find that thinning down the  $\text{MoO}_x$  and (i) a-Si:H layers (down to 4 nm and 6 nm respectively) mitigates parasitic optical sub-bandgap  $\text{MoO}_x$  absorption and drastically enhances charge transport, while still providing excellent passivation and selectivity. High-resolution transmission microscopy reveals that such thin  $\text{MoO}_x$  layer remains continuous and close to a  $\text{MoO}_3$  stoichiometry in spite of the reactive sputtering and annealing steps involved in the electrode deposition. Based on these insights, a screen-printed device reaching a certified efficiency of 23.5% and a fill factor of 81.8% is demonstrated, bridging the gap with traditional Si-based contacts and demonstrating that dopant-free selective contacts can rival traditional approaches.

### Introduction

The interest in silicon heterojunction (SHJ) is growing in the field of photovoltaics due to the simplicity of their structure and processing, as well as the high efficiencies that can be achieved [1], [2]. In fact, the record efficiency for a silicon single junction is currently held by a SHJ combined with an interdigitated back contacted (IBC) design, reaching 26.7% [3], [4]. Most of the SHJs use stacks of intrinsic and doped hydrogenated amorphous silicon (a-Si:H) as passivating and carrier-selective layers [5]. These stacks provide good surface passivation and selectivity thanks to the high hydrogen content of these layers and dopability of amorphous silicon, respectively [1], [6]–[8].

However, the excellent passivation performance of this contact architecture comes often at the expense of carrier transport in comparison with silicon homojunctions which translate into fill factor losses. This effect stems from the lower conductivity of doped a-Si:H compared to crystalline silicon, band offsets between these two phases [9][10], and the formation of an opposing Schottky contact between a-Si:H and the transparent conductive oxide (TCO) [11]–[13]. In addition, the use of a-Si:H (bandgap of  $\sim 1.7$  eV) on the light-incoming side of the device leads to severe short-wavelength (below 500 nm [1], [14]) parasitic light absorption, part light absorbed in the intrinsic a-Si:H ((i)a-Si:H) layers and all light absorbed in the doped a-Si:H layer being lost [14], [15]. This typically causes a short circuit current density ( $J_{sc}$ ) loss of up to  $2.1 \text{ mAcm}^{-2}$  compared to the one of a cell with an ideal cell front-side [15].

One investigated strategy to overcome this parasitic absorption is to replace the intrinsic and p-type (i/p)a-Si:H layers by more transparent carrier selective contacts. Silicon-based materials (in their nanocrystalline phase and/or alloyed with carbon and oxygen) [16]–[21], and transition metal oxides (TMOs) [22]–[26] are the most widely followed approaches, alongside a few other organic and inorganic materials [27], [28].

Most efficient devices using non-Si contacts have been achieved with molybdenum oxide ( $\text{MoO}_x$ ) as hole-selective contact [29]–[31]. This  $\sim 3.0$ -eV-bandgap material substitutes p-type a-Si:H on the light-incoming side of the solar cell to reduce parasitic absorption at short wavelengths [23], [26], [29], [32]–[34]. The high workfunction of  $\text{MoO}_x$  (5.7 eV to 6.9 eV [22], [26], [32], [35], [36]) enables this n-type semiconductor to ensure hole selectivity. Yet it does not provide sufficient surface passivation when deposited directly on c-Si [37]. An (i)a-Si:H layer is thus usually inserted as sketched in Fig 1a.

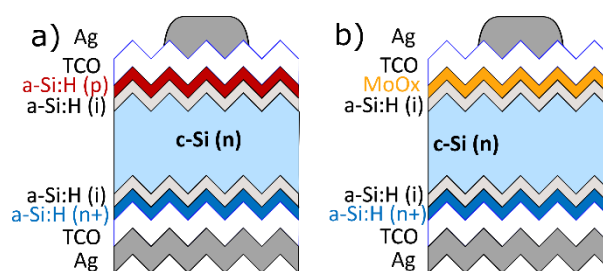


Fig 1: a) standard SHJ and b)  $\text{MoO}_x$ -based cell

As for standard SHJ devices (see Fig. 1b), a transparent electrode is required to provide lateral transport of charges on the positive-electrode side. A remarkable 22.5%-efficient device (with a fill factor, FF, of 80%) was demonstrated in 2015 using a combination of (i)a-Si:H, MoO<sub>x</sub> and hydrogen-doped indium oxide and a copper-plated grid [29]. Efficiency and FF were on par with those of standard SHJ state-of-the-art values at the time [38].

However, there are still challenges when contacting c-Si with a (i)a-Si:H/MoO<sub>x</sub>/TCO stack. First, a strong degradation of hole extraction, characterized by a FF drop, occurs upon annealing above 130 °C, a temperature required to cure industrial screen-printing pastes. Such degradation was attributed to the formation of an interlayer between MoO<sub>x</sub> and TCO [29], the formation of SiO<sub>x</sub> at the MoO<sub>x</sub>/a-Si:H interface [39], or/and the reduction of MoO<sub>x</sub> by H effusing from adjacent layers leading to a decrease of its workfunction [40]. The latter could be successfully mitigated by reducing the H-content of the a-Si:H layer prior to MoO<sub>x</sub> deposition, improving efficiency by 2% absolute yet without reaching similar performances as best-in-class devices [40].

Second, sub-bandgap absorption in the MoO<sub>x</sub>-based contact stack hampers its transparency and thus its advantage over standard (p)a-Si:H [29], [39]. Reducing the MoO<sub>x</sub> thickness is therefore a promising strategy, especially since TMOs have been reported to better protect (i)a-Si:H layers from damage induced by TCO sputtering compared to (p)a-Si:H [39], [41]–[44]. However, since electrical transport in the MoO<sub>x</sub> layer is carried by electrons through defect states, the MoO<sub>x</sub>/(i)a-Si:H interface is expected to be more recombining than the (p)a-Si:H/(i)a-Si:H one. This could affect passivation, and call for a thicker (i)a-Si:H layer than when using a (p)a-Si:H contact.

Following these considerations, we vary here the thicknesses of the (i)a-Si:H and MoO<sub>x</sub> layers in SHJ devices and discuss their influence on the optical and electrical performance of the cell. Furthermore, a low temperature curing paste is employed to avoid degradation and to assess the potential of this contact strategy using state-of-the-art SHJ processing [40][45].

### Experimental details

Fig. 2 details the processing sequence followed here. We used nine 180-μm-thick n-type textured float zone wafers with a resistivity of 1.7-2.3 Ω cm. A stack composed of (i)a-Si:H/(n)a-Si:H/(n)μc-Si:H was deposited at the back side by plasma enhanced chemical vapor deposition (PECVD) at 200 °C on all wafers. Three different thicknesses<sup>1</sup> (6 nm, 8 nm and 10 nm) of (i)a-Si:H were then deposited on the front side of the wafers (three co-processed wafers per thickness). Annealing in air at

250 °C, prior to MoO<sub>x</sub> deposition, was then performed in an oven (“pre-annealing” step [40]) to reduce the hydrogen content in the front-side (i)a-Si:H layer. After removal of the surface oxide in an aqueous solution of 5% diluted hydrofluoric acid (HF), a 4-nm-, 8-nm- or 9-nm-thick MoO<sub>x</sub> layer was thermally evaporated on the front side, each time on three wafers with a different (i)a-Si:H thickness. MoO<sub>x</sub> evaporation was done in a vacuum chamber (base pressure before deposition ~ 4\*10<sup>-6</sup> mbar) from stoichiometric MoO<sub>3</sub> powder at a deposition rate of about 0.03 nm/s. Finally, indium tin oxide (ITO) was sputtered on the front (70 nm) through a mask to form five 2 x 2 cm<sup>2</sup> devices per wafer. ITO (150 nm) and Ag (100 nm) were then sputtered over the full back side area. For these steps, only wafers with the same MoO<sub>x</sub> thickness (thus three wafers each time) were co-processed. A silver grid was finally screen-printed on the front side using a low-temperature paste, cured at 130 °C.

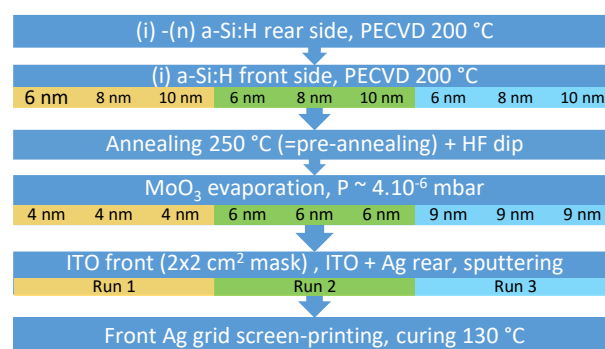


Figure 2: Process flow.

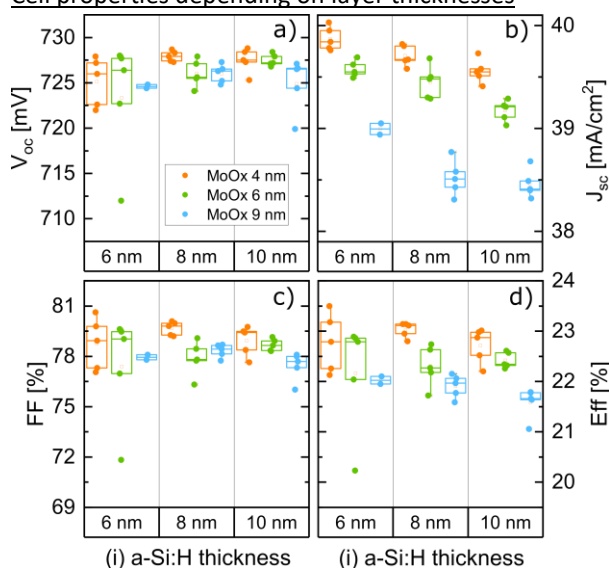
Standard SHJ solar cells were processed as references, using the same rear side stack, a 6-nm-thick (i)a-Si:H layer on the front side capped with an 8-nm-thick PECVD (p)a-Si:H layer instead of evaporated MoO<sub>x</sub>, omitting the pre-annealing step. Standard screen-printing Ag paste cured at 210 °C was used. The structure and chemistry of the contact was assessed by transmission electron microscopy (TEM). For that purpose, a layer stack of (i)a-Si:H/MoO<sub>x</sub>/ITO was deposited on a <111>-oriented mechanically polished (n)c-Si wafer. After annealing at 130 °C for 30 minutes, mimicking curing of the low temperature paste, a thicker MoO<sub>x</sub> layer (of 20 nm) was deposited on top of ITO to serve as a freshly evaporated MoO<sub>x</sub> reference. The contact stack was finalized by sputtering a Ag layer. A thin lamella was prepared using the conventional focused ion beam lift-out technique with a final thinning voltage of 2 kV in a Zeiss Nvision 40. Scanning TEM (STEM) high-angle annular dark-field (HAADF) images and energy-dispersive X-ray spectroscopy maps were recorded in a FEI Titan Themis microscope operated at 200 kV with a beam current of 200 pA. Electron energy-loss spectra (EELS) were

<sup>1</sup> All thicknesses are given on textured surfaces, calculated from measurements on flat Si or glass divided by a ratio of 1.5 accounting for the pyramids geometry.

acquired at 80 kV and at cryogenic temperature (-165 °C) to assess the oxidation states of the thin (4 nm) and thick (20 nm) MoO<sub>x</sub> layers without introducing electron beam artifacts [46]. A convergence semi-angle of 30.5 mrad was employed, while the collection semi-angle was 47 mrad. A dispersion of 0.1 eV/channel was employed for the dual EELS scans. For comparison purposes, M3 edges were aligned and rescaled to a value of 1 after removing the conventional power law background (using data within the ranges 380-390 eV and 445-450 eV to fit the background). Experimental data was compared to the reference MoO<sub>2</sub> and MoO<sub>3</sub> EELS spectra of [46] to assess the oxidation state of MoO<sub>x</sub> as a function of its thickness and processing history.

## Results and discussion

### Cell properties depending on layer thicknesses



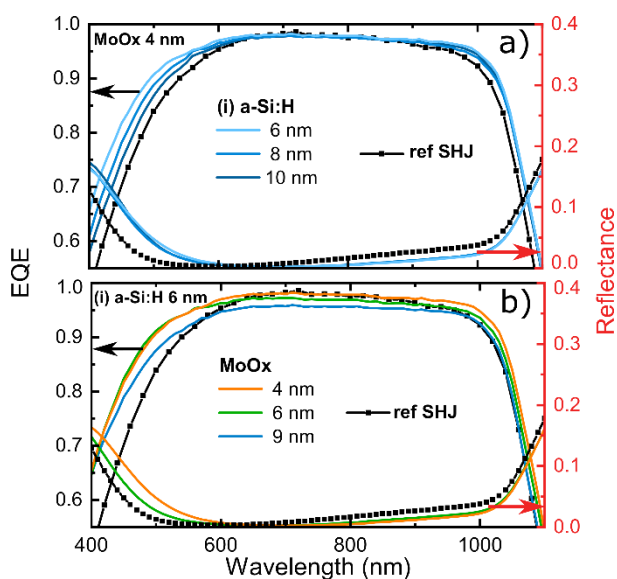
**Figure 3:** a) Voc, b) Jsc, c) FF, d) Efficiency of MoO<sub>x</sub>-based solar cells with different thicknesses for the front (i)a-Si:H and MoO<sub>x</sub> layers. Each dot represents one of five cells on a wafer. One wafer was prepared per condition.

**Figure 3** shows the parameters extracted from J-V measurements of the devices with different (i)a-Si:H and MoO<sub>x</sub> thicknesses. The open circuit voltage (Voc) values in Fig. 3a are similar for all MoO<sub>x</sub> and (i)a-Si:H thicknesses. Yet, statistical analysis indicates a slight Voc gain upon thickening the (i)a-Si:H layer of  $0.61 \pm 0.27$  mV/nm (p-value, a statistical criteria which estimates the analysis significance [47], of 0.03; here, the significance is set at 0.05, meaning that all statistical analysis with a p-value < 0.05 is considered significant) and a Voc loss of  $-0.41 \pm 0.25$  mV/nm when changing the MoO<sub>x</sub> thickness (p-value of 0.11, thus less significant), with no correlation between the two thicknesses. The thinnest MoO<sub>x</sub> thus globally yields a Voc ~2 mV higher than for thicker layers. Part of this gain (0.6 mV) can be attributed to the photogeneration difference, as estimated using a one-diode model with short-circuit current density (Jsc) shown in Fig. 3b as the

photogenerated current. Photoluminescence imaging (Fig. S1) and high-intensity Voc measurements (Fig. S2) [8] suggest that the thinnest MoO<sub>x</sub> provides better performance both in terms of passivation and selectivity than thicker films (discussed in Supporting Information). This demonstrates that the 4 nm MoO<sub>x</sub> film is sufficiently thick to protect the passivation provided by the (i)a-Si:H layer from sputtering damage and maintains sufficient selectivity to extract efficiently holes.

Fig. 3b shows that Jsc decreases when thickening either the MoO<sub>x</sub> or (i)a-Si:H layer. The optical loss induced by MoO<sub>x</sub> ( $0.25 \pm 0.01$  mA cm<sup>-2</sup> per nm of MoO<sub>x</sub> thickness, irrespective of the (i)a-Si:H thickness in the range tested) is higher than that induced by (i)a-Si:H ( $0.10 \pm 0.01$  mA cm<sup>-2</sup> per nm of a-Si:H, irrespective of the MoO<sub>x</sub> thickness, and as reported in Ref. [15]).

To identify more accurately the origin of these losses, external quantum efficiency (EQE) and reflectance measurements are shown in Fig. 4 and Table 1, highlighting gains in Jsc compared to the reference (p)a-Si:H SHJ cell. Reflectance is similar for all MoO<sub>x</sub> devices, with the exception of a slight variation in the 400-600 nm range, which may originate from batch-to-batch ITO variations. From Fig. 4a, the (i)a-Si:H thickness influences absorption mostly for wavelengths below 600 nm, and has no effect on reflection. On the other hand, Fig. 4b shows that the use of MoO<sub>x</sub> influences absorption over the whole wavelength range. Noticeably, the EQE signal is reduced from 600 nm to 1300 nm when increasing the MoO<sub>x</sub> thickness. This is due to sub-bandgap parasitic absorption in this range, as previously observed in Refs. [29], [39]. As this effect occurs in the visible range, it may also explain why MoO<sub>x</sub> sub-bandgap absorption impacts more strongly Jsc than (i)a-Si:H UV-blue absorption, as the photon flux is lower in the UV. In our study, a Jsc loss of  $0.4$  mA cm<sup>-2</sup> in the 600-900 nm range is recorded for the thickest MoO<sub>x</sub>, but interestingly no loss is observed for the thinnest MoO<sub>x</sub>. In the 320-600 nm range, gains in Jsc vary from  $0.5$  to  $0.8$  mA cm<sup>-2</sup> depending on the MoO<sub>x</sub> thickness (see Table 1). Although larger Jsc gains compared to standard SHJ cells were reported in literature in this range ( $1.9$  mA cm<sup>-2</sup> in [33],  $1.3$  mA m<sup>-2</sup> in [23], and  $0.9$  mA cm<sup>-2</sup> in [29]), these gains were mitigated by a strong loss at longer wavelengths, reducing the overall Jsc gain. In Ref. [33], the replacement of ITO by more transparent IO:H presumably compensated losses associated with sub-bandgap absorption in the MoO<sub>x</sub> layer. Overall, cells featuring the thinnest MoO<sub>x</sub> film investigated in this study exhibit a Jsc gain of  $1.3$  mA cm<sup>-2</sup>. These results demonstrate that an adequate device architecture enables to fully benefit from the improved transparency of MoO<sub>x</sub> compared to (p)a-Si:H.



**Figure 4:** EQE and reflectance of a) devices with a MoO<sub>x</sub> thickness of 4 nm and a variable (i)a-Si:H one; b) devices with a (i)a-Si:H thickness fixed at 6 nm and a variable MoO<sub>x</sub> one.

Samples with thinnest (i)a-Si:H (6 nm)	Jsc gain or loss (mA/cm <sup>2</sup> )			
	320 to 600 nm	600 to 900 nm	900 to 1180 nm	Total
MoO <sub>x</sub> 4 nm	0.84	0.03	0.46	1.33
MoO <sub>x</sub> 6 nm	0.83	-0.16	0.27	0.94
MoO <sub>x</sub> 9 nm	0.53	-0.43	0.05	0.15

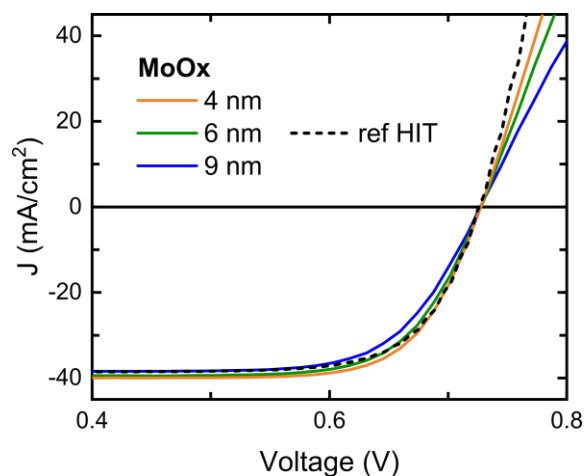
**Table 1:** Jsc gain for MoO<sub>x</sub>-based cells compared to the reference SHJ cell over various wavelength ranges calculated from EQE curves.

Fig. 3c shows that the thinnest layers investigated here are sufficient to ensure efficient charge extraction and passivation as a FF of up to 81% is achieved for the thinnest combination of MoO<sub>x</sub> and (i)a-Si:H layers. Statistical analysis indicates a stronger influence of the MoO<sub>x</sub> thickness, with a FF loss of  $-0.3 \pm 0.1 \%$ /nm (p-value of 0.03) and no significant influence from the (i)a-Si:H thickness (p-value of 0.44). In the case of a standard SHJ with (p)a-Si:H, the (i)a-Si:H thickness affects significantly the FF [15], suggesting that the MoO<sub>x</sub> layer imposes stronger selectivity than the (p)a-Si:H one. Indeed, if MoO<sub>x</sub> is more selective (thanks to its higher workfunction compared to (p)a-Si:H [23]), the hole population in (i)a-Si:H would be higher, which should lower the FF dependency on (i)a-Si:H thickness.

For standard SHJ solar cells, it is reported that an increase in (p)a-Si:H thickness leads to a higher FF [13], [15], [48], which might be due to the screening effect provided by the (p)a-Si:H layer thickness (“screening length”) towards the reverse Schottky diode induced at the interface ITO/(p)a-Si:H [13]. The same trend for FF was observed by increasing the p-type layer doping [49]. Increasing the MoO<sub>x</sub> thickness, however, leads to a drop in FF. This behavior might thus not be due to Schottky

diode screening. Instead, an increased series resistance (R<sub>s</sub>) due to transport in the MoO<sub>x</sub> bulk seems more likely to limit cell performance. Indeed, on glass samples with 8-nm sub-stoichiometric MoO<sub>x</sub>, a resistivity of  $4.5 \cdot 10^4$  Ohm cm is measured after evaporation, while 300 Ohm cm is measured after annealing at 180°C, literature values span from 200 Ohm cm [50] to  $5 \cdot 10^4$  Ohm cm [51]). Indeed, the R<sub>s</sub> of the hole contact stack appears to follow the same variation as FF. These effects are further discussed in the supporting information (see Fig. S3).

Figure 5 shows the J-V plots of cells with different MoO<sub>x</sub> thicknesses. We observe a non-linear characteristic, usually referred to as “s-shape,” which is typical for solar cells with passivating contacts. This effect arises when the extraction rate of majority carriers at the contact becomes limited, or when the leakage rate of these carriers at the opposite contact is not negligible [52]. Although thinning down the MoO<sub>x</sub> layer mitigates this effect, it is still present in the thinnest layer as evidenced by the lower current at high forward voltages compared to the reference SHJ. The impact on cell performance is however small as the reference SHJ and the thinnest MoO<sub>x</sub> cells (orange) have a similar performance in the fourth quadrant. The higher Jsc of the cells with a thin MoO<sub>x</sub> leads to a higher efficiency compared to the reference SHJ cell.



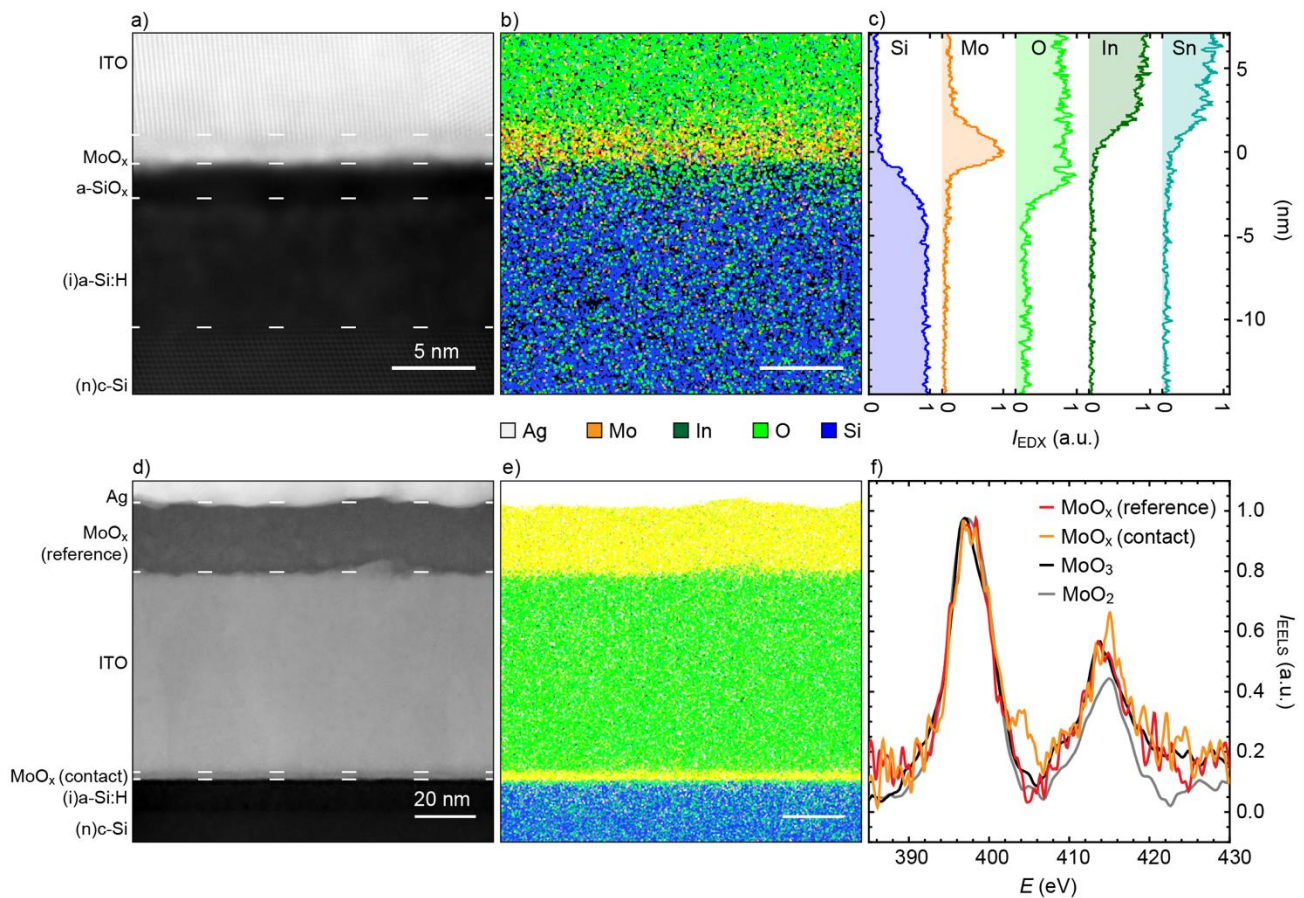
**Figure 5:** J-V of the three different cells featuring different MoO<sub>x</sub> thicknesses, with 6 nm of (i)a-Si:H for all devices. The dashed black line represents the reference SHJ cell ((p) a-Si:H of 8 nm, (i) a-Si:H of 6 nm).

#### Microstructure and chemistry of the contact

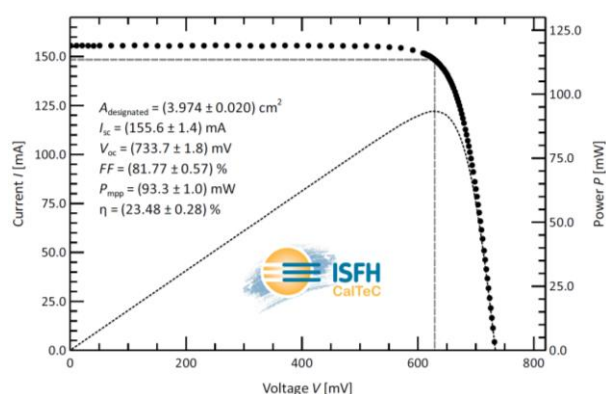
The microstructural and chemical properties of MoO<sub>x</sub> were investigated by STEM HAADF imaging and EDX, as shown in Fig. 6a-c. The EDX line profile shown in Fig. 6c highlights the presence of a thin amorphous SiO<sub>x</sub> layer at the (i)a-Si:H/MoO<sub>x</sub> interface, as reported in [39]. The Mo M3-M2 EELS fine structure of the thin MoO<sub>x</sub> contact layer is comparable to the one of the thicker reference MoO<sub>x</sub> layer (~20-nm-thick) that was not subject to any annealing or plasma exposure (Fig. 6d-f). These MoO<sub>x</sub> EELS spectra indicate that the layers have a

stoichiometry closer to  $\text{MoO}_3$  than  $\text{MoO}_2$ , even for a layer as thin as 4 nm and sandwiched between a-Si:H and ITO and subjected to annealing at 130 °C. Either the  $\text{MoO}_x$  stoichiometry is indeed unchanged by its surroundings, or the

higher affinity of Si to Mo does result in a  $\text{MoO}_x$  reduction after deposition (leading to the observed interfacial  $\text{SiO}_x$  layer), which is cancelled by the subsequent plasma exposure during ITO deposition which could induce its re-oxidation.



**Figure 6:** a) STEM HAADF image of the (i)a-Si:H/ $\text{MoO}_x$ /ITO contact deposited on a <111>-oriented (n)c-Si wafer, b) corresponding EDX map and c) background-subtracted EDX line profiles of the Si K, O K, In L, Sn L edges. d) STEM HAADF image of the contact coated with an as-deposited  $\text{MoO}_x$  layer/Ag stack, e) corresponding EDX map and f) EELS Mo M3-M2 edges of the thin and thick  $\text{MoO}_x$  layers compared to reference spectra.



**Fig. 8:** certified J-V and power plots of the best  $\text{MoO}_x$ -based cell processed in the lab (certification for ISFH)

### Optimized $\text{MoO}_x$ contact design for record performance

The highest efficiency of this series was obtained for a 4-nm-thick  $\text{MoO}_x$  layer and a 6-nm-thick (i)a-Si:H film. Given that

results with the thinnest (i)a-Si:H are quite spread, we processed another 4-nm  $\text{MoO}_x$  cells with thicker (i)a-Si:H layer of 8 nm, and a certified efficiency of 23.5% was obtained (Fig. 8), with a remarkable FF close to 82%. This record efficiency for such solar cell was obtained without any masking step or photolithography and uses a low-temperature Ag screen-printed metallization causing ~3% shadowing [45], [53] [54].

### **Conclusion**

In summary, we studied here the influence of the thicknesses of the  $\text{MoO}_x$  and (i)a-Si:H films used as hole contacts in SHJs. The thinnest layers studied here (~4 nm and ~6 nm for  $\text{MoO}_x$  and (i)a-Si:H, respectively) resulted in best device performances, highlighting that thin such contact stacks enable simultaneously high selectivity, passivation, and good charge carrier transport. Thicker layers were found to lead to higher series resistance or charge carrier selectivity losses, without any strong correlation between the two. The device employing the thinnest  $\text{MoO}_x$  showed a higher optical

response (higher EQE) than the reference (p)a-Si:H-based SHJ over the whole wavelength range (320–1180 nm). This gain in photocurrent of 1.3 mA/cm<sup>2</sup> resulted from a mitigation in parasitic absorption losses. High resolution transmission electron microscopy imaging and electron energy-loss spectroscopy revealed that the 4-nm-thick evaporated MoO<sub>x</sub> layer is continuous and has a stoichiometry close to MoO<sub>3</sub>, even after sputtering the overlying ITO layer and annealing the contact stack. Overall, the high J<sub>sc</sub> allowed by this contact stack enabled the demonstration of a 23.5%-efficient device, highlighting that MoO<sub>x</sub> is an excellent hole contact for c-Si heterojunctions. By reaching a similar efficiency to that of best SHJs cells produced in our laboratory [55], this study demonstrates that MoO<sub>x</sub> can rival traditional contacting schemes in spite of its lower level of optimization. It demonstrates that carrier-selective contacts relying on non-Si materials are relevant for high-efficiency silicon photovoltaics. As a perspective, reducing further the thickness of the MoO<sub>x</sub> layer and investigating the industrial feasibility and long term stability of such devices will be necessary steps before large-scale deployment of this technology can be envisaged

#### Acknowledgments

The authors would like to thank C. Allebé and P. Wyss for the wafer cleaning and texturization, V. Paratte for processing passivation layers and electron contact on c-Si wafer, N. Badel for the Ag screen-printing metallization, and Cédric Bucher and Aymeric Schafflützel for technical support.

This project is funded by the Swiss national Ambizione Energy Grant ICONS.

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