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Native Oxide Barrier Layer for Selective Electroplated Metallization of Silicon

**Heterojunction Solar Cells** 

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The metallization of silicon heterojunction (SHJ) solar cells by electroplating of highly

conductive copper onto a multifunctional patterned metal layer stack is demonstrated.

The approach features several advantages: low temperature processing, high metal

conductivity of plated copper, no organic making and low material costs (almost Ag-

free). A PVD layer stack of copper and aluminum is deposited onto the cell

subsequently to TCO deposition. The aluminum layer is patterned with a printed

etchant and its native oxide on the remaining areas inhibits plating. The full area

aluminum layer while electroplating supports plating current distribution and allows

homogeneous plating height distributions over the cell. The NOBLE (native oxide

barrier layer for selective electroplating) approach allows reaching a first encouraging

SHJ solar cell efficiency of 20.2% with low contact resistivity.

1. Introduction

Electroplated copper contacts are particularly attractive for crystalline silicon

heterojunction (SHJ) solar cells which require a low-temperature metallization.

Current trends in the silicon photovoltaic field point towards passivated contacts architectures for high efficiency solar cells.<sup>[1,2]</sup> Indeed, an outstanding efficiency of 25.1% was reached by Adachi *et al.* with a bifacial SHJ solar cell contacted by electroplating.<sup>[3]</sup>

The symmetrical SHJ solar cell architecture is created by first depositing a thin intrinsic amorphous hydrogenated silicon layer – i.e. a-Si:H(i) onto the cleaned and textured c-Si wafer, followed by the doped a-Si:H (n or p) which act as carrier selective junction. Both ultra-thin a-Si layers are quite sensitive to process temperatures above 200 °C.<sup>[4]</sup> Typically, a transparent conductive oxide (TCO) is sputtered onto the a-Si:H layers to contact the Si, promote the lateral conductivity in the solar cell and to serve as antireflective coating. Other passivated contact solar cell architectures based on poly-Si, such as tunnel oxide passivating contacts (TOPCon),<sup>[5]</sup> or polycrystalline silicon on oxide (POLO),<sup>[6]</sup> are more recently in the focus of development. In both cases the poly-Si layers may also be covered by TCOs. All mentioned cell architectures then need to be contacted by metallization approaches, where metal-paste printing is the most common ("Reference" – Figure 1) and metal plating is a promising alternative.

Metal pastes printed on SHJ solar cells are designed for low-temperature application, due to the thermal sensitivity of the amorphous layers. As the cells cannot be heated to above 200 °C, the conductivity of the metal paste is relatively low.<sup>[7,8]</sup> This problem has been partly reduced by application of the smart wire technology, which can directly interconnect printed contact fingers with small solder-coated wires that form the module interconnect.<sup>[9]</sup> However, a metallization using busbars, especially on

bifacial solar cell design, leads to a higher metal consumption (mostly Ag) due to the print requirements (wide fingers) on both surfaces.<sup>[10,11]</sup>

On the other hand, a plated metallization is intrinsically a low temperature process. Plated approaches using highly conductive and less expensive copper are currently investigated. [12–20] The Cu-based contacts can be electroplated simultaneously on both-sides of the solar cells and TCOs like indium-tin-oxide (ITO) are good barrier to metals diffusion into silicon. [21] This manufacturing of plated bifacial SHJ solar cells intends to increase the contacts conductivity and reduces drastically the precious Ag consumption. Several plated routes are reported to metallize standard bifacial 6 inches SHJ solar cells as depicted in Figure 1.

The "Dielectric masking route" starts by printing of a silver paste on TCO, dried and covered afterwards by a  $SiO_x$  layer prepared by a plasma-enhanced chemical vapor deposition (PECVD) – in a second vacuum tool. The following curing of the printed seed in grid-positions cracks through the  $SiO_x$  layer. The  $SiO_x$  on TCO acts as insulating mask to plate copper only onto the printed seed. Moreover it is an interesting approach for increased reliability of the solar cells. [22]

The classical "Resist masking route" (Figure 1) includes a full area metal-seed (Cu) which can be deposited subsequently to TCO deposition in the same tool (without vacuum interruption). The thin Cu layer promotes a homogeneous current distribution for simultaneous fast bifacial plating and allows reaching very low contact resistivities on TCO. [23,24] An organic resist-mask is then inkjet-printed (> 10  $\mu$ m thick) for grid-patterning before plating into the mask openings, resulting in fingers below 20  $\mu$ m wide. After copper and silver plating, the resist mask is stripped (organic contamination of water waste) and the metal seed-layer etched-back in non-contact

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positions without affecting the underlying TCO layer. This sequence has produced bifacial 6" SHJ solar cells with up to 24.1% efficiency. [25,26]

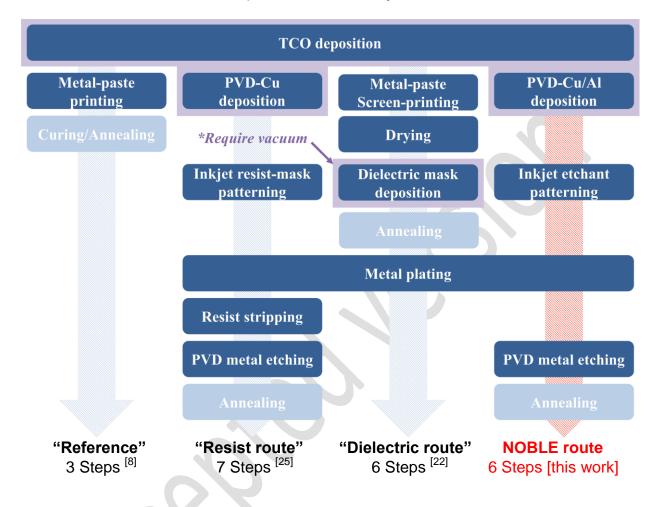


Figure 1. Overview of the low-temperature routes to metallize bifacial SHJ solar cells covered by TCOs

Our work demonstrates an alternative shorter plating approach (one step less, still almost Ag-free). Our novel native oxide barrier layer for selective electroplating i.e. "NOBLE route", (Figure 1) aims at improving the process flow, with fewer steps and by avoiding the use of any costly organic masks. In this concept, the native oxide of a thin aluminum (Al) layer enables performing selective Cu plating onto a patterned

metal-seed. An etchant is inkjet-printed to structure the contact positions in the Al

layer. This grid-patterning method is even cheaper than the deposition of a reactive

silver ink onto the aluminum. [15,17] Only a small area – contact-grid area < 10% of the

cell, has to be printed in contrary to the "Resist route" (masking > 90% of the cell

area). In all other positions, the thin Al layer effectively suppresses parasitic plating

issue.

2. Approach

Our NOBLE process is based on the selectivity of the plating and etching behaviours

of different materials. The material combinations of the metallization are chosen to

enable:

- selective etching of the top PVD metal layer against the underlying PVD metal-

seed in a fast and robust process,

- selective plating on the bottom PVD metal-seed against the top PVD metal

covered by a native oxide,

- selective etching of the full PVD metal layers against the top plated layer of the

contact and against the TCOs.

The NOBLE metallization route depicted in Figure 2, allows fast and homogeneous

electrodeposition of copper (Cu) onto a multifunctional patterned metal layer. The

metal stack consists of a metal-seed (low contact resistivities to TCO, susceptible to

plating) and an Al layer at the surface, both deposited by physical vapor deposition

(PVD) onto the TCO - Figure 2 (1). The additional cost for the PVD metal layer

depositions is relatively low since it can be realized in the same tool as the TCO

deposition, without vacuum interruption. The sputtered metals are reported to

achieve good adhesion and low contact resistivity on TCOs. [23,24] In addition, they promote a quick and homogeneous current distribution along the bifacial solar cell

during plating. Indeed, the sheet resistance of the TCOs is too high to accomplish a

homogeneous Cu deposition on both-sides 6" surface with acceptable process speed.

The native oxide Al<sub>2</sub>O<sub>3</sub> (alumina) growing in ambient atmosphere on the Al surface is

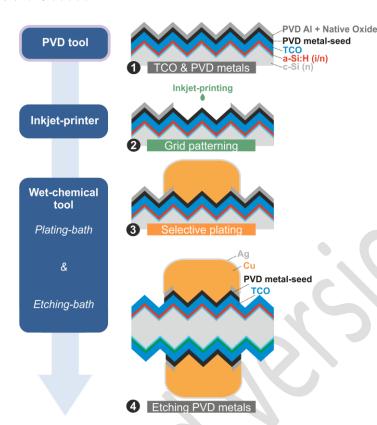
convenient for the selective electroplating described below.

After vacuum deposition of the PVD layer stack, the grid-patterning is achieved by printing a low concentrated alkaline ink or paste to etch the Al-Al<sub>2</sub>O<sub>3</sub> selectively – Figure 2 (2). In the present work, inkjet printing was employed. This selective etching removes the Al layer only in grid positions and does not impact the underlying metal-seed (on top of the TCO). This patterning step allows to plate Cu selectively onto the metal-seed – Figure 2 (3). At this state, the less noble metal Al and the Al<sub>2</sub>O<sub>3</sub> are acting as barrier to inhibit Cu plating in the non-grid positions as described by

Hatt et al.[17] A thin silver capping (~ 200 nm) follows to protect the Cu-contacts

against oxidation.

The remaining PVD layers (Al and metal-seed) in non-grid positions are then etched-back by chemical processing without damaging the underlying TCO – Figure 2 (4). On the contact flank, only few nanometers of Cu are not protected by Ag in comparison to the "Resist route" which lets several micrometers un-capped.



**Figure 2.** Sequence and tools used for the low-temperature NOBLE metallization on TCO for bifacial silicon heterojunction solar cells.

### 3. Experimental details

## 3.1. Metal etching

The investigations on metal etching were realized on planar glass (soda-lime glass, 49x49 mm², 1 mm thick). Different metals: Al, Cu, Ag, NiV (93:7) and Ti were sputtered on the glass after a plasma pre-treatment to increase the adhesion of the coatings. Electron-gun evaporation was also used to deposit Al onto glass to observe the impact of the PVD technique on the etching rate for this metal. Around 500 nm were deposited except for the thicker sputtered Al coating (~ 900 nm). These layer thicknesses are far greater than needed on solar cells, but facilitates evaluation of the etching rate. After a variation of etchant volume, 500 mL of etchants solution

were used to etch four samples of each metal to avoid the saturation of the solution by the metal ions, which might result in a reduction of the etching kinetics. Four etching times were chosen depending on the etching kinetics for each solution and metal. The reactions were realized under agitation at room temperature (22 °C for all) and at 35 and 50 °C for some etchants. Samples were thoroughly rinsed immediately after the desired etching duration was reached. The metal thicknesses were measured (five different spots per sample) before and after etching by a confocal microscope (LEXT — Olympus OLS4000), profilometer (Dektak — STYLUS PROFILER) or X-ray fluorescence tool (FISCHERSCOPE® X-RAY XDV- $\mu$ ). The thicknesses etched in dependence of the time were plotted. The etching rates were then determined by linear fits presenting all R<sup>2</sup>  $\geq$  0.9. An etching rate of zero was considered if the impact on the metal was not significant after one hour. Indeed, one hour is quite sure-estimated for etching steps which are typically performed in few seconds along the NOBLE metallization.

# 3.2. Cell fabrication

Commercial bifacial textured SHJ solar cells precursors covered on both sides by indium-tin-oxide (ITO) were covered in the same sputtering tool (as the glass) with thin Cu/Al or Ag/Al layers: 50/100 and 20/100 nm, respectively. A diluted aqueous solution of sodium hydroxide (NaOH) at 1 wt % was prepared to pattern the Al layer. A PIXDRO LP50 printer from Meyer Burger Technology AG was used to print the grid positions with NaOH<sub>aq</sub>, thus etching completely the Al. For our first solar cell the non-optimized patterning (pitch of 2 mm between the fingers, one busbar centered up to 0.2 mm wide) has been realized only on the front side. An Ag-paste was used to

metallize the rear-side of the solar cell. The wafers were cleaned in deionized water before immersion in a slightly acidic copper sulfate-based electrolyte for electroplating. The selective Cu plating onto the PVD metal-seed, composed by Cu or Ag, was realized by a forward / reverse pulsed current. The plating selectivity is favored on large Cu regions while parasitic Cu-seeds are dissolved during the short reverse current pulse. A clamp on the wafer edge allows to apply a medium current density of 6 A dm<sup>-2</sup>, forward and reverse current pulse times of 15 ms and 1 ms respectively and a high anodic to cathodic current ratio of 4.5. A thin immersion silver capping was then deposited onto the plated Cu. With the contacts metallized, the PVD layers in non-grid positions could be etched-back by a short dip of ~ 5 s in an aqueous phosphoric-nitric acid mixture at 50 °C. A short anneal in air was finally realized at 200 °C for 15 min to recover the sputtering damages of the a-Si:H layers. The contact metallization was characterized by the confocal microscope after each processing step. The 1-sun current voltage (I-V) parameters of the solar cell were measured under standard testing conditions (STC: AM1.5g, 100 mW cm<sup>-2</sup>) using a sun-simulator. The solar cells were then diced and fingers were polished by an ionbeam before being observed in cross-section by a Schottky emission scanning electron microscope (SEM) SU-70 from *Hitachi*. The transfer length method (TLM) was also applied on the diced cell (1 cm length) to assess the sheet resistances and contact resistivities of the metals on the electron or hole selective contacts of the commercial SHJ precursor. Then, tape tests were realized by glueing a Kapton® adhesive band on the contacts metallized and removing them by hand.

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### 4. Results and discussions

## 4.1. Wet metal etching

The etching selectivity of different metals was studied to enable the selective etching steps mentioned above. There are many solutions that may etch metals deposited by PVD. The investigation was focused on PVD AI, Ti, NiV, Cu and Ag which are promising for either low contact resistance, or plating selectivity. The etch rates in nm s<sup>-1</sup> are presented in Figure 3 for metals sputtered on planar glass. The etching rates might be slightly different on the TCOs or on the textured surface of the SHJ solar cells. However, general trends are expected (and were confirmed) to be similar. As already discussed earlier, the metal stack could also have an impact on the etching rate, as corrosion elements occur between base and noble metals. The sputtered AI is hard to etch due to its self-passivation but can still be quickly dissolved (> 1-3 nm s<sup>-1</sup>) by a solution of sodium hydroxide NaOH, even at low concentration as follows:

$$2 \text{ Al} + 2 \text{ NaOH} + 6 \text{ H}_2\text{O} \rightarrow 2 \text{ Na[Al(OH)}_4] + 3 \text{ H}_2$$
 (1)

As can be seen, NaOH does not etch Ag or Cu (at least during one hour), thus being a selective etchant for the removal of Al from these contact layers. Comparing the metal deposition technique, it was observed that the sputtered Al is more easily dissolved than our thermal evaporated Al. The strong acidic blend composed of phosphoric and nitric acid H<sub>3</sub>PO<sub>4</sub>/HNO<sub>3</sub> removes Al quite slowly (< 1 nm s<sup>-1</sup>), but is a very fast etchant for Cu and Ag (> 10 nm s<sup>-1</sup>) even at room-temperature. Copper is also selectively removed in iron chloride FeCl<sub>3</sub> or ammonium persulfate (NH<sub>4</sub>)<sub>2</sub>S<sub>2</sub>O<sub>8</sub>. Another important etching selectivity was demonstrated for Ag, which is quickly etched in solutions of ammonia and hydrogen peroxide NH<sub>3</sub>/H<sub>2</sub>O<sub>2</sub> at even lower

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concentration than the one effective for Al, Cu and NiV. Sputtered Ti and NiV are generally quite stable against chemical attack due to their native oxides, but Ti is still significantly and selectively impacted by NH<sub>3</sub>/H<sub>2</sub>O<sub>2</sub>.

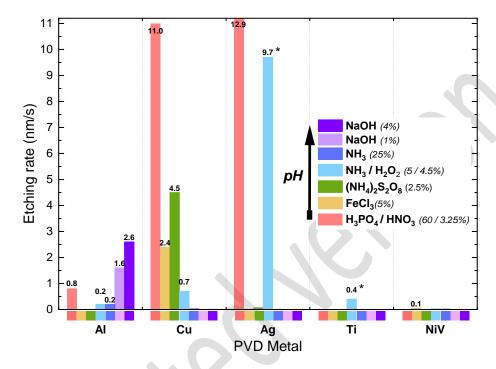


Figure 3. Etching rates of thin (≤ 1µm) sputtered metal layers removed from planar glass at room temperature in different solutions (\*lower NH<sub>3</sub>/H<sub>2</sub>O<sub>2</sub> concentration).

In summary, Al can be selectively removed versus Cu or Ag with dilute alkaline solutions and at high etching rates. On the other hand, Cu can be etched with FeCl<sub>3</sub> or (NH<sub>4</sub>)<sub>2</sub>S<sub>2</sub>O<sub>8</sub>, while Ag and Al will remain unaffected. These selective metal etching possibilities lead to simple chemical processes for metal patterning in the photovoltaics or microelectronics field. The NOBLE processing route discussed below presents an application of this principle.

## 4.2. NOBLE processing

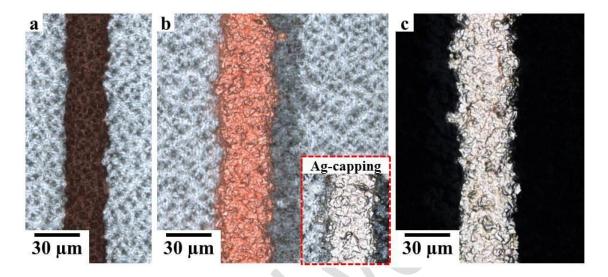
Both sides of the textured SHJ precursor with ITO are entirely covered by thin layer stacks of either PVD-Cu/AI (50nm/100nm) or Ag/AI (20nm/100nm). The selective etching of AI against the metals below (Cu or Ag) enables to pattern the grid-positions easily. In the present work, inkjet-printing of NaOH<sub>aq</sub> was employed to structure lines in the AI-AI<sub>2</sub>O<sub>3</sub> layer. Narrow lines in the PVD AI of around 25 µm wide without spreading or interruption were created. The underlying PVD Cu-seed can be observed in line positions as demonstrated in Figure 4 (a). A slight effect of the NaOH<sub>aq</sub> on the PVD Cu-seed could occur but may only result in surface roughening which could even increase the adhesion of the plated contacts as presented for silicon by Büchler *et al.*<sup>[27]</sup>

A further development might allow to reduce the line width since the volume of the droplets jetted are only up to 2-5 pL and a high contact angle between the etchant and the AI / AI<sub>2</sub>O<sub>3</sub> surface seems benificial. Increasing the temperature catalyzes the reactions and the kinetic is favorable to AI vertical etching, enabling complete removal of the AI layer without horizontal spreading. Moreover, the AI etching rate might increase due to local corrosion elements which may take place at the interface AI / nobler metal.<sup>[28]</sup> Typical seed materials are Aq, Cu (both used in this work).

The selective Cu electrodeposition is then performed onto the Cu-seed as observed in Figure 4 (b). The native oxide – covering the Al surface, acts as inhibitor for Cu deposition outside of the designated contact area. The Cu grows isotropically onto the metal-seed which widens the contact a bit. The Cu deposition is fine-granular which gives high contact conductivity near the value of the Cu-bulk. The forward / reverse pulsed current avoids parasitic plating as already described in a previous

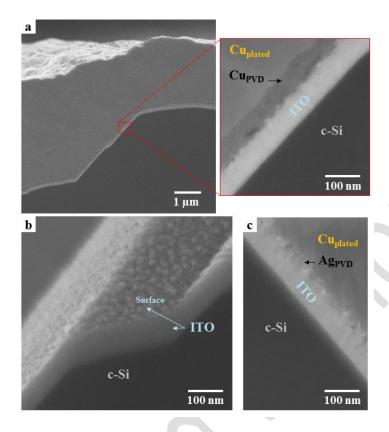
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study where the acidity, composition of the electrolyte and the plating settings were optimized.<sup>[17]</sup> A thin silver capping is also electroplated to prevent Cu from any oxidation.



**Figure 4.** Microscopic pictures of a finger on the ITO from a SHJ solar cells along the NOBLE metallization after: (a) inkjet-printing of NaOH<sub>aq</sub>, (b) Cu-Ag plating and (c) etching-back PVD layers in non-contacted positions.

After plating, the PVD layers are selectively etched-back (Figure 3) in non-grid positions. In this case both layers were etched by a short dip in a solution of  $H_3PO_4/HNO_3$  at 50 °C. Narrow contacts – i.e. fingers, composed of  $Cu_{PVD}/Cu_{plated}$  /  $Ag_{plated}$  (50 nm / 1 to 10  $\mu$ m / 200 nm) were created on ITO as presented in Figure 4 (c). The ITO thickness and surface were observed by SEM in cross-section which confirms that ITO is not significantly damaged through the etch-back step (Figure 5 (b)).



**Figure 5.** SEM pictures in cross-section of the SHJ solar cell covered by ITO after NOBLE metallization: (a) finger plated on Cu-seed, (b) area on a pyramid flank in non-finger position and (c) finger plated on Ag-seed.

## 4.3. Contact characterization

Fingers after NOBLE metallization on the SHJ precursors are characterized in cross-section by SEM as illustrated in Figure 5 (a) and (c). The metal stack of sputtered Cu (~ 50 nm) or Ag (~ 20 nm) and plated Cu-Ag are well adhering on the ITO of the SHJ solar cells even with a non-optimized metal sputtering realized after vacuum interruption (samples were prepared on commercial precursors). No voids or uncontacted area could be noticed as it is regularly the case for printed fingers, [7] which would increase the resistivity at the interface. Otherwise, the ITO thicknesses

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below the finger and in the non-contact positions (Figure 5 (b)) are similar (~ 80 nm) i.e. ITO was not attacked in the etching step.

The adhesion of the plated contacts metallized on ITO already easily passed tape tests and further peel-off tests will be realized on full size 6" solar cells. Furthermore, the adhesion peel forces of PVD metal stacks on TCOs, are typically high, as has been shown on SHJ solar cells using a resist mask and plating (measured adhesion above 4 N mm<sup>-1</sup>).[26]

**Table 1.** Sheet resistances (R<sub>sh</sub>) of ITO and contact resistivities (ρ<sub>contact</sub>) of different stacks measured by TLM after NOBLE metallization on commercial SHJ solar cells.

Metal stack	$R_{sh}$ [ $\Omega sq^{-1}$ ]		
c-Si (n) / a-Si:H(i/n) / ITO / Cu <sub>PVD</sub> / Cu-Ag <sub>plated</sub>	82.7 ± 0.2	4.0 ± 0.4	
c-Si (n) / a-Si:H(i/n) / ITO / Ag <sub>PVD</sub> / Cu-Ag <sub>plated</sub>	80.6 ± 0.2	$3.5 \pm 0.8$	
c-Si (n) / a-Si:H(i/p) / ITO / Cu <sub>PVD</sub> / Cu-Ag <sub>plated</sub>	169.7 ± 0.5	2.8 ± 0.5 Not influenced	
c-Si (n) / a-Si:H(i/p) / ITO / Ag <sub>PVD</sub> / Cu-Ag <sub>plated</sub>	120.3 ± 0.4	1.8 ± 0.3  by the bulk	

TLM measurements were realized to measure the ITO sheet resistance R<sub>sh</sub> and the contact resistivity  $\rho_c$  of the fingers composed by two different metal stacks directly on both sides of the textured SHJ solar cells as presented in

Table 1. The contact resistivity between metal and TCO, which is the fraction that can be influenced by the selected metallization process, can be extracted from the measurement on the a-Si:H(p) side, as this is isolated from the bulk on the used ntype material. The values obtained on this side are approximatively 2-3 m $\Omega$  cm<sup>2</sup>. On the a-Si:H(n) side, the values are slightly higher. However, the PVD metal-seed composed by Cu or Ag do not seem to impact significantly the pc. In term of cost reduction, a PVD Cu-seed is preferred to get closer to the Ag-free metallization. Furthermore, the  $\rho_c$  values are in accordance with the literature even if Lee et al. reported values between 0.4-0.8 mΩ cm<sup>2</sup> for an optimized stack ITO/Cualloy<sub>PVD</sub>/Cu<sub>plated</sub>. [23,24] Moreover, our contact stack after NOBLE metallization is similar to the one used by CSEM to reach a very high efficiency up to 24.1% on bifacial plated SHJ solar cell.[25]

#### 4.4. Solar cell properties

As reference, screen printed metallization allowed to achieve 21.5% as best efficiency on large-area solar cells with this precursor batch. With the above presented NOBLE metallization a first lab-scale SHJ solar cell - almost silver-free, was produced. The contacts are composed by ITO / Cu<sub>PVD</sub> / Cu<sub>plated</sub> / Ag<sub>plated</sub>. The efficiency (n) reached 20.2% and an encouraging fill-factor (FF) of 78.0% was obtained. This is despite the fact that the pseudo fill factor (pFF) of this cell is already limited to only 80.5%, due to the small cell size (see below).

Table 2 presents the SHJ solar cell properties under 1-sun illumination.

**Table 2.** SHJ solar cells properties after NOBLE metallization

Area	V <sub>oc</sub>	pFF	FF	J <sub>sc</sub>	η
[cm²]	[mV]	[%]	[%]	[mAcm <sup>-2</sup> ]	[%]
6.25	718	80.5	78.0	36.1	20.2

The open-circuit voltage V<sub>oc</sub> and pseudo fill-factor pFF limitations result from the nonoptimized metal sputtering process on ITO and from the small cell size - i.e. edge impact already studied for homo-junction cells by Rauer et al. [29] The fill-factor is impacted by the non-optimal grid design – i.e. the pitch between the fingers, and the non-optimized rear-side metallization (described earlier). The short-circuit current  $J_{sc}$  might be reduced by the lab-scale etch-back procedure of the PVD layers. Optimizations of the process to exploit the full potential of the solar cell in combination with the apparently well performing metallization are ongoing.

### 5. Conclusion

An alternative approach for low temperature metallization of solar cells with TCO layer (e.g., SHJ solar cells), which is currently in early development stage, was demonstrated. It takes advantage of the selectivity of etchants and plating processes towards different metals.

A thin stack of two PVD metal layers were sputtered onto the cell right after the TCO in the same tool without breaking the vacuum. Thus, sputtering costs are kept low, especially for these thin layers and materials such as Al and Cu. These metal layers allow to plate the patterned contacts on both sides of the cell at the same time quickly and homogeneously. The selective etching of metals was investigated in a study of etching rates for Al, Cu, Ag, Ti and Ni in different acidic and alkaline solutions. This selectivity enabled grid patterning of thin PVD-Al layer by inkjet-printing of an alkaline solution. Copper could then be plated in the printed areas — without parasitic deposition, onto the underlying PVD metal-seed due to the presence of native oxide on the Al surface. After electroplating, the thin PVD layers were removed by etching in non-grid positions selectively. Contact resistivities below 3 m $\Omega$  cm $^2$  were achieved on stack systems of ITO-Cu<sub>PVD</sub>-Cu<sub>plated</sub>-Ag<sub>plated</sub> and the improvement of the sputtering might allow reaching 0.4 m $\Omega$  cm $^2$  as reported in the literature. A promising efficiency

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of 20.2% with a FF of 78.0% was presented on a commercial SHJ solar cell

precursor.

This approach saves processing steps and consumables as compared to the

electroplating into openings in an organic mask. An upscaling of the process onto

standard 6 inches SHJ and carrier selective junction (CSJ) solar cells is currently

ongoing to reach the full potential of the passivated contacts solar cells with plating.

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