NANO-IMPRINT TECHNIQUE FOR BACK REFLECTOR IN HIGH EFFICIENCY N-I-P THIN FILM SILICON SOLAR CELLS

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ABSTRACT: In this contribution, the replication of nano-textures enhancing the short circuit current of thin film silicon solar cells onto low cost substrates (glass or PolyEthylene Naphtalate (PEN)) is studied. Optical and morphological analysis is performed to asses the quality of these replicas. Single and tandem a-Si:H solar cells are deposited on top of the master and replica structures to verify their suitability to be used as substrates for solar cells in substrate (n-i-p) configuration. We find similar stabilized efficiencies (> 8 %) for tandem cells on a master and a PEN replica of a different texture. The use of a soft stamp to make these replicas is smoothing the original features of the masters. Replications with higher fidelity of fine features are obtained with solid stamps. Such replicas do not exhibit discernible differences when compared to their master textures by atomic force microscopy measurements. Moreover a comparison of single junction amorphous cells made on a replica and on its corresponding master shows no difference in term of light trapping capability of the texture. Keywords: Thin film Solar Cells, Replication Process, Back Reflector

1 INTRODUCTION

In substrate configuration, thin film silicon solar cells open the possibility to use plastic foils as substrate because the deposition can be realized by roll to roll processing [1, 2, 3]. This advantage could lead to a reduction of the cost of the final module and provides unique lightweight unbreakable products. To further increase the efficiency of the thin film silicon solar cells, the texturing of the substrate, when performed properly, produces light trapping i.e. an increase of the optical path of the light within the device. This approach permits to decrease the thickness of the absorbing layer, providing better charge collection and reduced light induce degradation while keeping the short circuit current and thus the efficiency as high as possible. In substrate configuration, the texturing is often realized on the back reflector itself by growing rough metal layer at high temperature such as silver (hot Ag) [4]. For research, textured substrates are often obtained by metalizing textured transparent conductive oxides such as Asahi type SnO₂:F or boron doped ZnO grown by low-pressure chemical vapor deposition (LP-CVD-ZnO) [5, 6]. However, these processes are not normally suited for plastic substrates (temperature for hot Ag and Asahi type substrate and easy stress fracturing for LP-CVD-ZnO on flexible substrate). Possible workaround are the use of a sacrificial high temperature substrate (Heliantos process) [7], plasma texturing of the PET substrate [8], or roll to roll embossing processes (e.g. substrates made by the company OVD Kinegram A.G. that were used in a previous work [10]).

In this work we present some of our latest results for the texturing of low cost plastic substrates by UV-Nano-Imprinting Lithography technique (UV-NIL) which is a room temperature process operating under reasonable pressure (~1bar) and which can be easily up-scalable [10]. We present here two types of replication processes, a first one using soft stamps [11] and a second one using solid stamp that improve the replica quality [12]. We use optical and morphological analysis to quantify this

quality, and we present amorphous single junction and tandem solar cells on replicas on either glass or PolyEthylene Naphtalate (PEN). The suitability of the process to produce high quality textured substrates for solar cells is evidenced by equally high yields of operating devices on master and replica. The replication process using solid stamp shows a complete replication of the original texture under atomic force microscopy measurements (AFM) and, when inserted into single junction amorphous device, a light trapping behavior similar to the one of the master is observed.

2 EXPERIMENTAL

A UV-NIL system made "in-house" is used for the transfer of the nano-metric structure on PEN or glass substrates. The replication of the original surface textures are made from stamps that contain the inverse of the original texture; a UV curable lacquer is spin coated on a substrate and transferred into an evacuated chamber together with the stamp. Then, pressure and UV light are applied to cure the UV sensitive lacquer. The original positive texture is thus replicated onto the cured lacquer after its separation from the stamp. The stamps were made by exposing the original texture either to polydimethylsiloxane (PDMS), or to the UV-lacquer itself. The PDMS is an easily deformable soft stamp, UV replica is a solid stamp made of the lacquer.

In this work, we present the replication of two types of structure which are known to lead to high efficiency amorphous solar cells: Type A structure is made of silver sputtered under high temperature conditions (~400°C) and Type B is a texture formed naturally during growth of ZnO by LP-CVD. Because in solar cells the structures are covered by a thin layer of silver to make both the contact and the back reflector, we cover the master of Type B and all replicas with a thin layer of sputtered silver (~120nm) before characterization. AFM measurements are used to extract the root mean square roughness (σ_{rms}) and the correlation length of the texture

(L), diffuse reflectance is obtained using a spectrophotometer coupled with an integrating sphere and the angle resolved scattering properties is measured using a laser at 543 nm. We then incorporate the structures into devices (single and tandem junction amorphous solar cells) to see whether the produced substrates are fully compatible with the complete solar cell deposition process using plasma enhanced chemical vapor deposition at a frequency of 70 MHz and a temperature around 190 °C (see e.g. [13]).

3 RESULTS AND DISCUSSION

In the first part, the results obtained with the soft PDMS stamp are shown. As an important modification of the original texture is observed using this stamp, we present in the second part of this section the newly developed replication process using a solid stamp made of a direct replica of the master.

3.1 Soft stamp

The quality of the replica is first presented using morphological and optical measurements. Then both masters and replicas of Type A and B textures made on glass are used as substrates for single junction solar cells. As the Type A master and the Type B replica show similar diffuse light behavior and similar efficiency for the single junction solar cells grown on them, Type A master made on glass and Type B replica made on a flexible PEN substrate are compared when used in tandem amorphous/amorphous cells. It shows the capability to obtain high efficiency substrate-configuration solar cells on flexible substrate using the method of UV-Nano-Imprinting.

3.1.1 Morphological and optical analysis

Figure 1 shows the AFM images made on the masters and the replicas obtained on glass for both types of structures. It is observed that the principal overall shape is reproduced in the replicas, but a blurring of the features occurs. Table 1 presents the $\sigma_{\rm rms}$ and the L values for both master and replica of Type A and B extracted from the AFM image of figure 1. We attribute the increase of the correlation length for both types of structures in the replica compared to the master to an unfaithful replication of the smaller features present in the original textures. On the contrary we see that the $\sigma_{\rm rms}$ of Type A texture is unchanged while it decreases for Type B texture. It shows that, the bigger the original features are, the more losses we obtain after replication of the structure.

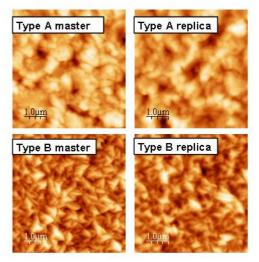
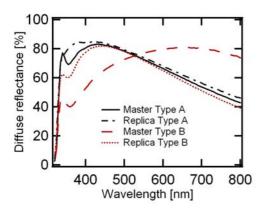


Figure 1: AFM images of masters and replica obtain on glass using soft stamps for Type A and B textures

Table I: Comparison of morphological parameters extracted from the AFM images of master and replica made with soft stamps

	Type A		Type B	
	Master	Replica	Master	Replica
$\sigma_{rms} [nm]$	46	47	67	51
L [nm]	228	280	167	202

The distribution between specular and diffuse light, as well as the angle resolved scattering properties are the most commonly studied optical properties of textured solar cell substrates. Indeed, these characteristics give some insight on the light trapping capability of the texture for the solar cells. Figure 2 presents the diffuse reflectance measurements on top and the angle resolved scattering at 543 nm on the bottom for the various master and replica surfaces. The replica of Type A reproduces quite well the optical behavior of its master, but at 360 nm the absence of the surface plasmon absorption peak in the replica's diffuse reflectance could indicate that the excitation of the surface plasmon is due to the smaller features of the textures. Concerning the Type B structure, it is quite clear from the diffuse reflectance that the replica is much smoother than the original texture, and the replica does not reproduce the massive absorption loss of the plasmon resonance. However the angular dependence of the scattering is still quite close to the one produced by its mater texture.



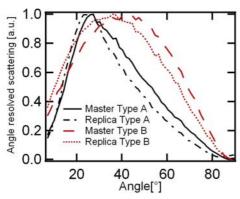


Figure 2: Optical analysis of master and replica using soft stamp. Top) diffuse reflectance, Bottom) angle resolved scattering properties at 543nm

3.1.2 Single junction amorphous solar cells

The replicas are tested as substrate into solar cell devices to assess the suitability of this process towards efficient cells in substrate configuration. Figure 3 and table II present the important parameters (External Quantum Efficiency (EQE), short circuit current density (J_{sc}), open circuit voltage (V_{oc}), fill factor (FF) and efficiency (η)) of single junction amorphous solar cells grown on masters and replicas of both types of structures (200 nm of <i> absorber layer, 65 nm of ITO front contact layer).

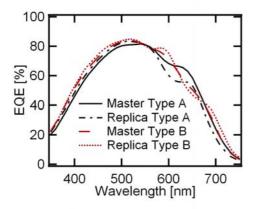


Figure 3: External quantum efficiency of cells on master of Type A and B and their corresponding replica using a soft PDMS stamp

It can be seen in figure 3 and in table II that the

replication of Type A suffers of a small loss in light trapping properties (J_{sc} decrease) which decrease the efficiency of the cells compared to the one grown on the master. For type B structure the imperfect replication of the original texture is in fact beneficial in term of V_{oc} and FF due to the smoothing of the original texture. We conclude that the absence of losses in the J_{sc} for the replica compared to the master is due to parasitic absorption in the silver that covers the master texture. The overall efficiency of the cell grown on the replica of Type B texture is therefore bigger than the one of the cell grown on the original texture.

Table II: Initial parameters of the single junction amorphous cells grown on masters and replicas structure for Type A and B

	Type A		Type B	
	Master	Replica	Master	Replica
V _{oc} [V]	0.884	0.881	0.847	0.876
FF [%]	72.7	70.1	64.8	70.4
J_{sc} [mA/cm ²]	13.41	12.85	13.51	13.84
η [%]	8.6	7.9	7.4	8.5

3.1.3 Tandem amorphous/amorphous solar cells

From the two previous subsections we extract the following: Type A master and Type B replica are similar substrates in term of morphology, diffuse reflectance and efficiency of the single junction cell grown on top. To prove the ability of this process to obtain high efficiency cells with good light trapping properties on a flexible substrate we use Type A master and Type B replica made on PEN as substrates for tandem amorphous/amorphous cells.

The tandem cells are made of the same 300 nm thick bottom cells. As the replica of Type B textures shows diffusion into higher angles compare to Type A master, the thickness of the top cell on the replica substrate is decreased to obtain current matching between the two sub-cells. The thicknesses of the top amorphous cells are thus 70 nm for Type A master substrate and 60 nm for Type B replica on PEN substrate.

To evaluate statistically the cells quality, V_{oc} and FF are measured under low illumination (0.4% of AM1.5). Cells with a Voc below 1.2 V or a FF below 30% at low illumination are judged as non functional. With these criteria, from the 20 cells on each substrate, 90% were judged functional on the PEN substrate and 85% on the glass substrate. It proves that the replica on PEN does not introduce additional problems compared to a standard glass substrate during the process of the cell deposition.

Table III shows the parameters of the cells on each substrate. The $V_{\rm oc}$ and FF are taken as a mean of all the working cells and the $J_{\rm sc}$ and the efficiency are shown for the best cells. From these values it is clear that the mean values on all the working cells of $V_{\rm oc}$ and FF are similar for both stabilized and initial state. Similarly the stabilized current and efficiency (>8%) achieved for the

best cell on each substrate are comparable even though the top cell grown on Type B replica has a slightly lower thickness compared to the cells grown on Type A master.

Table III: Initial and stabilized (in bold) values of the tandem cells. $V_{\rm oc}$ and FF are taken as a mean on all the working cells while the $J_{\rm sc}$ and efficiencies are presented for the best cell on each substrate

•	Type A master on glass		Type B replica on PEN	
	Initial	Stab.	Initial	Stab.
V _{oc} [V]	1.73	1.68	1.70	1.68
FF [%]	71.7	59.6	72.8	62.3
J _{sc} [mA/cm ²]	7.54	7.30	7.62	7.31
η [%]	9.8	8.3	9.5	8.1

3.2 Solid stamps for Type B structure

The previous section has shown that the process of replication can be used to obtain substrates with good light trapping properties, imposing no additional difficulties for the growth of the cells. The smoothing of the features during the replication process can be beneficial for the efficiency of the cells. Because this cannot be the case in general, we aim towards a better quality of the replication, for example in cases when the original texture is already optimized for the cell efficiency. Therefore, we developed a replication process using solid stamps. The solid stamp is made of a direct replica of the master which gives the inverse of the structure. As Type B was not perfectly replicated previously we concentrate on this structure in this last section.

3.2.1 Morphological analysis

We use AFM to qualify the quality of the replica. The analysis of these images is shown in table IV and figure 4.

Table IV: Comparison of morphological parameters extracted from the AFM images of master and replica made with solid stamp

	Type B	
	Master	Replica
σ _{rms} [nm]	70	69
L [nm]	165	159

Table IV shows that the σ_{rms} and the L values for both master and replica of Type B are similar. Figure 4 presents the histograms of the height distribution and the local inclination angle for the master and the replicas using both soft and solid stamps. The improvement of the

quality of the replica using the solid stamp can be observed clearly and it is difficult to see a difference between the master and the replica made with the solid stamp. For further illustration, figure 5 shows scanning electron microscopy images of both master and replica using solid stamp, again showing no discernible differences.

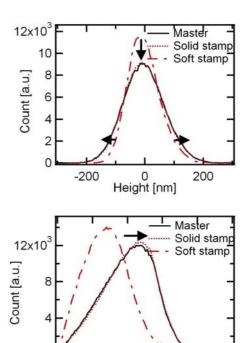


Figure 4: Height (top) and angle (bottom) histograms for the master and the replicas using both soft and solid stamps

40

60

Angle [°]

80

100

0

20

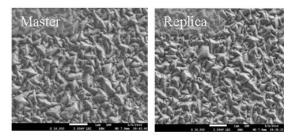


Figure 5: SEM images of Type B master and its replica using solid stamps. The blank rod at the bottom represents $1\mu m$

3.2.2 Single junction amorphous solar cells

As the replica and the master are similar using solid stamps, we co-deposited a single junction amorphous cell of 200 nm of <i> layer (65 nm of ITO layer as front contact) on both substrates. The corresponding EQEs are shown in figure 6. The red responses (>550nm) of both cells indicates all but equal light trapping properties of the master and the replica.

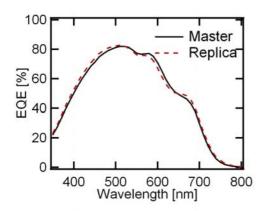


Figure 6: External quantum efficiency of two codeposited cells on master of type B and its corresponding replica using solid stamp

4 CONCLUSION

We have established a replication process using a UV curable lacquer that offers new possibilities of textures which were previously not obtainable directly on flexible low cost plastic substrates (e.g. Hot Ag / LP-CVD-ZnO). We demonstrate comparable stabilized efficiencies (>8%) using tandem amorphous cells grown on a master structure obtained on glass and a replica obtained on PEN. The use of a soft stamp permits to smooth the master texture when the latter is strongly textured while the use of a solid stamp permits to obtain replica showing no noticeable differences with the original texture that is known for its excellent light trapping properties when used as a substrate in thin film silicon solar cells.

5 ACKNOWLEDGEMENTS

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6 REFERENCES

- S. Guha and J. Yang, Journal of Non-Crystalline Solids, 352 (2006) 1917.
- [2] Y. Ichikawa, T. Yoshida, T. Hama, H. Sakai, and K. Harashima, Solar Energy Materials and Solar Cells, 66 (2001) 107.
- [3] E. A. G. Hamers, M. N. Van den Donker, B. Stannowski, R. Schlatmann, and G. J. Jongerden, Plasma Processes and Polymer, 4 (2007) 275.
- [4] A. Banerjee and S. Guha, Journal of Applied Physics, 69 (1991) 1030.
- [5] H. Iida, N. Shiba, T. Mishuku, H. Karasawa, A. Ito, M. Yamanaka, and Y. Hayashi, Electron Device Letters, IEEE, 4 (1983) 157.
- [6] S. Faÿ, J. Steinhauser, N. Oliveira, E. Vallat-Sauvain, and C. Ballif, Thin Solid Films, 515 (2007) 8558

- [7] J.K. Rath, Y. Liu, A. Borreman, E.A.G Hamers, R. Schlatmann, G.J. Jongerden, and R.E.I. Schropp, Journal of Non-Crystalline Solids, 354 (2008) 2381.
- [8] V. Terrazzoni-Daudrix, J. Guillet, F. Freitas, A. Shah, C. Ballif, P. Winkler, M. Ferreloc, S. Benagli, X. Niquille, D. Fischer, and R. Morf, Progress in Photovoltaic, 14 (2006), 485.
- [9] J. Bailat, V. Terrazzoni-Daudrix, J. Guillet, F. Freitas, X. Niquille, C. Ballif, T. Scharf, R. Morf, A. Hansen, D. Fischer, Y. Ziegler, A. Closset, Proc. 20 EPVSEC, Barcelona, (2005)
- [10] S. H. Ahn, and L. J. Guo, ACS Nano, 3 (2009), 2304.
- [11] K. Söderström, J. Escarré, O. Cubero, F.-J. Haug, S. Perregaux, and C. Ballif, Progress in Photovoltaic, DOI: 10.1002/pip.1003.
- [12] J. Escarré, K. Söderström, C. Battaglia, F.-J. Haug and C. Ballif, (Unpublished).
- [13] T. Söderström, F.-J. Haug, V. Terrazzoni-Daudrix, X. Niquille, M. Python, and C. Ballif, Journal of Applied Physics, 104 (2008) 104505.