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Translucent, Color-neutral and Efficient Perovskite Thin Film Solar Modules[†]

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Thin film perovskite photovoltaic devices combine high power conversion efficiencies with low weight, large area high speed production capabilities and high versatility in form factor. With this paper, an additional feature is added to the device property portfolio: optical transparency. Results on translucent perovskite modules with selectively tuned levels of transparency are reported by adopting the industrial established fabrication process of partial area ablation to high efficiency opaque devices. After process completion, the final devices consist of opaque areas, that absorb the light, and transparent areas, that permit neutral light transmission. We compare the effectiveness of use of two active area removal techniques: mechanical scribing and laser ablation and determine the factors that limit the maximum transparency and power conversion efficiency. Moreover, we show how the choice of the ablation method and design of active area removal pattern affect and limit the maximum light utilization efficiency ratio that can be achieved with this fabrication process. Resulting 4 cm² translucent modules cover a range of visible light transparency from 7-37 % with corresponding power conversion efficiency of 5-13 %.

World energy, and especially electricity, sector is rapidly changing. Year 2016 marked the first year that new photovoltaic installations surpassed any other electricity generation technology¹. Being modular technology photovoltaic can fulfill various electricity demands through distributed generation. In parallel, urban areas are growing and currently account for two thirds of total global energy supply². Currently only 20 % of the energy used in building and transport sectors of urban areas is supplied by renewable technology. This represents a significant potential for modular distributed electricity technology such as photovoltaics. Transparent photovoltaics create additional options for integration of electricity generation in these sectors. In comparison to silicon based photovoltaics, thin film PV offer many degrees of freedom in terms of size, flexibility and color. Perovskite thin film PV is an emerging technology currently known for unexpectedly fast improvements in power conversion efficiency on the lab scale devices. This study shows implementation of an existing industrial fabrication process on optimized perovskite thin

film module technology to provide variable visible light transparency solutions. Limitations of this process are defined in order to help improve future development of highly efficient and transparent perovskite based photovoltaics.

1 Introduction

Photovoltaic technology plays a major role in converting the electricity supply from conventional fossil and nuclear fuels towards renewable energy. With constant increase in energy consumption in highly populated urban areas³, integration of transparent photovoltaic (TPV) systems on different levels of scale from large buildings to small low power electronic devices enables distributed electricity generation in close proximity to the consumer⁴⁻⁷. The final application of stable TPV will depend not only on the power conversion efficiency and transparency that the system is able to achieve, but also on design (color and shape), size and flexibility of the panel.

Regardless of materials used, TPVs can be sorted into two groups based on the range of the light spectrum they are utilizing for the power generation: TPV that harvest solar radiation beyond the visible spectrum and TPV that utilize a fraction of the incident visible light.⁷ In this paper, we are focusing on the latter group. This group includes commercially available TPV based on amorphous Si and thin film PV (TFPV) with organic and perovskite photoactive layers.⁶⁻⁸ Such TPV devices can be realized by two different methods: by applying transparent layers, referred to as

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semi-transparent PV^{9–11} or by partial area ablation of opaque layers, referred to as translucent PV^{12,13}. For semi-transparent PV, the increase in band-gap or a decrease in the active layer thickness raises the transparency but reduces the power conversion efficiency (PCE). On the other hand, for translucent PV the thickness remains optimal but a part of the active area is traded for transparency.^{6,8,14} The advantages of translucent PV over semi-transparent PV include color-neutrality, ease of transparency variation and direct transferability of opaque PV technology efficiency improvements.

Translucent PV technology has been demonstrated for Si-based and thin-film PV technology.^{6–8} The first generation, Si-based PV, is less desired for TPV applications due to efficiency decrease with partial shading and rigidity in transparency design.¹⁵ TFPV offers large area, light weight, low-cost options for translucent PV with a high variability in the design.^{8,16} However, existing translucent PV technology struggles with limited efficiencies and red color light transparency, which limits the range of applications.⁶ Perovskite TFPV offers the possibility to combine high power conversion efficiencies^{17,18} with color-neutral light transmission. Number of studies have demonstrated TPV using small area perovskite solar cells.⁷ The best reported semi-transparent perovskite solar cell of 0.06 cm² area has a power conversion efficiency of 7.7 % and an average visible transmittance (AVT)^{7,19} of 39.0 %²⁰, while a matching translucent perovskite solar cell of area 0.09 cm² achieves a PCE of 4.0 % with an AVT of 50.0 %.¹³ However if potential of use of this application is to be assessed, upscaling of processing is crucial. While translucent perovskite modules have not been reported until now, a semi-transparent perovskite module with a power conversion efficiency of 12.0 % and limited AVT of 2.4 % for an aperture area of 4.0 cm² has been demonstrated.²¹

In this work, we present translucent perovskite modules of 4 cm² aperture area with PCEs of 5.4 - 13.1 % and corresponding AVTs from 37-7 %, respectively. We compare the efficacy of using laser ablation or mechanical patterning in the processing of translucent modules by varying the active area removal from 10-50 % of the aperture area. Based on the electrical and optical characterization of processed translucent devices we determine that the patterning method, the pattern design and the perovskite stack are the three crucial factors affecting the performance of translucent perovskite modules.

2 Experimental

2.1 Translucent device fabrication

Perovskite based solar cells and modules used in this study are planar n-i-p devices†. Cells have active areas of 0.54 cm², while modules exhibit an aperture area of 4 cm². Since, modules consist of monolithically interconnected cells, their fabrication is an alternation of patterning and deposition steps†. Initial processing results in opaque modules with an optimized design consisting of four cells with 1 cm² aperture area each. The module interconnections are fabricated using 355 nm nanosecond laser ablation (Trumpf Trumark 6330) for the initial patterning step P1 and mechanical patterning for P2 and P3 (Fig. 1d).²² Resulting opaque

modules exhibit geometrical fill factors (GFF) of 92-94 % giving the fraction of the aperture area that is actively contributing to the photogeneration of electricity.

The opaque modules are converted to translucent by applying an additional patterning step P4 in a stripe design (Fig. 1).⁶ The P4 lines are perpendicularly oriented to the interconnection lines and are formed by the removal of 4 layers: the bottom contact, hole transport layer, the perovskite photoactive layer and electron transport layer. The resulting translucent modules are an alternation of opaque and transparent areas with a very high aspect ratio (Fig. 1c). In the following, we define the ratio of transparent area to aperture area of the device as the transparent area ratio. Opaque areas, each of width W, now perform as modules that are electrically connected in parallel. In this study, P4 is patterned using either picosecond laser ablation (355 nm wavelength, Coherent Talisker Ultra) or mechanical scribing.²²

In order to study the differences in use of laser or mechanical patterning for the preparation of translucent modules, the opaque area width W, and the pitch length have been varied for targeted removals of 10 %, 25 % or 50 % of the module aperture area. Moreover, the effect of opaque area width W on translucent device performance is further analyzed using translucent cells with targeted active area removal from 20-60 % of the active area.

2.2 Characterization

Samples were electrically characterized using a four-probe setup with a Keithley 2602A Source-Measure unit under 1 Sun illumination. The electrical module and cell performance is determined by J-V scans and maximum power point measurements†. The values are reported for devices measured in the opaque configuration before the P4 patterning and after patterning in the translucent condition (η_{MPP} in Tab. 1).

Translucent devices were furthermore characterized using optical microscopy, scanning electron microscopy (SEM) and spectrophotometry. Transparent area ratio is calculated based on optical microscopy of translucent devices imaged from ITO, front contact, and gold, back contact, sides. SEM is used to study cross-sections of translucent devices and analyze damage edge effect and re-deposition for laser and mechanical patterning technique, while spectrophotometers are used to measure specular and total transmittance of translucent devices.

3 Results and Discussion

3.1 Device performance

Translucent device performance is optimal if the decrease in PCE is reflected only in J_{SC} , proportional to active area removal by P4 patterning. The decrease in J_{SC} of translucent modules presented here corresponds to the increase in transparent area ratio (Tab. 1 and inset Fig. 2a). Moreover, based on J-V curves of translucent modules with 10 %† and 25 % transparent area ratio, these devices exhibit no significant additional resistance losses due to processing. Exemplary J-V curves of modules in opaque and translucent configurations for 25 % transparent area ratio are given in Fig. 2a. Additional J-V data for other transparent area ratios are provided in the supporting information (Fig. S1).

Fig. 1 a) Image of 25 % laser patterned 4 cm² translucent module with a metal shadow mask covering the non-active edge area emphasizing the aperture area; b) Optical microscope close-up image of the translucent module with apparent removal design pattern with the blue line showing the area of cross section shown in d); c) An illustration of translucent module showing transparent and opaque areas perpendicular to the interconnections of the cells in a module. Generated charges are extracted along the long axis of the cells formed by the separated opaque areas.

Table 1 Optical and electrical parameters for 4 cm² opaque and translucent modules. For each device the data before and after the ablation process are given for reference. J-V parameters for reverse scan of translucent modules are given, as well as PCE at 5 minute maximum power point voltage for reference and translucent modules.

RM*	Configuration	TAR [%]	J_{SC} [mA/cm ²]	V_{OC} [V]	FF [%]	η [%]	η_{MPP} [%]	LT [%]	AVT [%]
O	Reference	0.0	19.2	0.950	77	13.7	13.1	0.0	0.0
	Reference**	0.0	20.5	0.963	76	14.9	14.5	0.0	0.0
M	Opaque	0.0	19.2	0.950	77	13.8	13.1	0.0	0.0
	Translucent	9.8	18.4	0.963	74	13.1	12.8	7.6	7.6
M	Opaque	0.0	19.3	0.950	77	13.9	13.4	0.0	0.0
	Translucent	25.0	14.8	0.950	73	10.3	10.1	18.0	18.2
M	Opaque	0.0	19.5	0.925	78	14.1	13.2	0.0	0.0
	Translucent	51.1	8.6	0.938	72	5.8	5.1	37.2	36.8
L	Opaque	0.0	19.4	0.950	77	13.9	13.0	0.0	0.0
	Translucent	8.9	18.6	0.950	72	12.8	12.5	7.3	7.2
L	Opaque	0.0	19.3	0.938	71	12.5	11.1	0.0	0.0
	Translucent	23.3	14.9	0.938	70	9.8	9.6	17.2	16.9
L	Opaque	0.0	19.6	0.938	71	12.8	11.6	0.0	0.0
	Translucent	42.9	9.3	0.950	60	5.4	4.9	33.0	32.4

*RM - removal method: O-opaque, M-mechanical patterning, L-laser patterning.

**During air storage, the reference opaque module shows an increase in performance over time (13.1 % to 14.5 %)

Fig. 2 a) I-V curves of two modules before (opaque) and after (translucent) partial area removal. The modules have been either laser (red) or mechanically (blue) patterned with a targeted transparent area ratio of 25 %. Inset shows the change of the current density for translucent modules with increase in transparent area ratio. b) Normalized opaque area PCE for modules with different transparent area ratios. The errors reflect the uncertainty in the determination of transparent area.

In the presented translucent concept the opaque area is only a fraction of the aperture area. Therefore we also represent the generated power relative to the opaque area by the opaque area power conversion efficiency (OAPCE) allowing for a comparison of the same device in opaque and translucent configuration. By normalizing the OAPCE with the initial PCE of the same device before the P4 patterning we can compare different modules with a variation in transparent area ratio (Fig. 2b). Up to a transparent area ratio of 30 %, the modules show the same OAPCE, confirming that no additional losses are introduced by the processing.

However, modules with a transparent area ratio above 40 % show a decrease of 15 % - 25 % in normalized OAPCE depending on the patterning method used. In the I-V scans these translucent modules show an increased series resistance, in comparison to translucent modules with transparent area ratio below 40 % (Fig. S1†). Since decrease in performance is mainly reflected in a lower FF, we analyze the P4 pattern with microscopy.

Material removal quality and pattern profiles of P4 show significant difference for the two patterning methods in SEM images (Fig. 3). In contrast to laser patterning where layer removal up to ITO is complete in most of the P4 pattern except for the P4 edges, mechanical patterning leaves material residue along the whole width of P4. Moreover, significant difference in mechanical and laser profiles is seen at the edge of opaque area where mechanical patterning shows irregular lateral damages due to chipping, while laser patterning results in delamination of top contact and Spiro-OMeTAD. This effect is caused by thermal effects of laser processing of the stack where Spiro-OMeTAD and gold have a low adhesion to the underlying perovskite layer.²³

Even though these two patterning methods exhibit significant differences, both show a structural breakdown of the perovskite stack close to the edge on the order of 10-20 μm for each edge of the opaque area (damage edge). Since such damage is seen with all samples, it is expected that as the opaque area width W decreases, the ratio of non-active to unaltered opaque area will increase and the performance of modules will be greatly affected. The proposed hypothesis is tested by comparing the OAPCE of two sets of translucent cells where the targeted transparent area ratio was varied from 20-60 %. One set of samples had fixed W of 180 μm , while for the other W decreased with increasing transparent area ratio from 180 to 20 μm (Fig. 4a). Results show significant difference in performance between the two sets of cells. With increase in transparent area ratio, performance of the normalized OAPCE remains constant up to 40 % transparent area ratio for the set of cells with constant W of 180 μm . For the cells with decreasing W the OAPCE is dropping drastically for increasing transparent area ratios. With increasing transparent area ratios over 50 % a substantial material re-deposition from the increased total ablated area is occurring which causes additional efficiency and transparency declines (Fig. 4b).

To quantify the effects of an uneven patterning damage edge on the PCE of a translucent module, we apply a model that assumes a gradual change of the PCE at the damage edge (Fig. 5a). We calculate the PCE in dependence of the opaque area width W with $PCE(W) = \int_0^W PCE(x)dx$. In Fig. 5b the normalized OAPCE

Fig. 3 Scanning electron microscope images of cross sections of P4 of translucent modules with a) mechanically or b) laser patterned P4. Each image gives zoomed in profile of the P4 damage edge.

Fig. 4 a) Comparison of opaque area PCE of set of translucent cells with fixed wide opaque area width, W of 180 μm (black) and cells with decreasing opaque area width from 180-20 μm (gray). Opaque area PCE is normalized to reference opaque cell PCE. b) SEM image of cross section of laser patterned P4 with damage edge for high transparent area ratio translucent cell with opaque area width, W of 90 μm , showing apparent redeposition of removed material covering the sample.

of translucent cells and modules is compared for the two patterning methods. The measurements are presented together with the simulation of the normalized OAPCE for damage edge widths of 5 μm to 80 μm . In the presented data, most of the devices show a PCE dependence on the width W that corresponds to damage edge widths of 20–40 μm .

The presented analysis allows for a precise determination of the edge damage of the P4 pattern. The ongoing process optimization of the patterning method, the patterning design and the perovskite stack is targeting a reduction of the edge damage and its influence of the PCE. The results of the analysis are also relevant for opaque modules since the P4 ablation is identical to the P3 ablation process. In conventional opaque modules, the P3 damage edge is reducing the geometrical fill factor of a module by increasing the dead area of the interconnection pattern.²²

3.2 Transparency

The combined analysis of total and specular light transmittance of the translucent modules reveals scattering due to the higher specular light transmission (Fig. 6). The light transmission is gradually increasing in the range from 300–800 nm with a step at around 780 nm that corresponds to the band-edge of the absorbing perovskite layer. This increase is primarily seen in the total transmission and is more pronounced with mechanically patterned modules. Such partial light absorption in the translucent areas is due to incomplete material removal (as seen in Fig. 3), re-deposition of ablated material and edge effects of the opaque area which corresponds to the SEM observations.

Since, translucent modules consist of opaque and transparent areas, light is transmitted through transparent areas consisting of glass/ITO layers. To analyze the efficacy of the transparent area to transmit light in the visible region, we define the transparent area transmittance (TAT) (Fig. 7a). TAT is the ratio of module AVT and transparent area ratio, where a TAT of 1 is equivalent to a complete transmittance of the light incident on the transparent area. For both patterning methods, the same fraction of light incident on the transparent area is transmitted. Moreover, TAT is used to compare transparent areas of translucent modules to glass/ITO in transmission of light. For the glass/ITO stack TAT is represented with the dotted line at 87 % and represents the highest value that can be achieved for the transparent area of translucent modules with remaining ITO in P4. With 80 % TAT at 10 % transparent area ratio and about 75 % for increased transparent area ratios, it shows that less light is transmitted by the transparent area of translucent modules than by the reference glass/ITO stack alone. This is a combined effect of additional absorption by residual material in the transparent area, seen in Fig. 3 and Fig. 6, and an increased scattering at the air-ITO interface due to higher surface roughness caused by deterioration due to the ablation process.

The transparency of semi-transparent solar cells is usually reported as simple average of visible transmittance (AVT) for 400–800 nm wavelength range, based on the spectral transmission measurements. For architectural applications as in building integrated PV (BIPV), the visual appearance of a transmissive device

can also be described by light transmittance (LT) given by the international standard for glass in buildings in ISO 9050:2003.¹⁹ In the definition of LT, the wavelength range from 380–780 nm is combined with the spectral human eye sensitivity in order to represent the visual appearance of the total light transmission. LT is comparable to proposed corrected AVT.⁷ In our perovskite translucent modules the LT is increasing from 7 to 37 % with increasing transparent area ratio of 10 to 50 % (Tab. 1). We report both AVT and LT values, even though in cases of relatively flat transmittance curves as seen here, these values are comparable. In applications of semi-transparent solar cells, the goal is to optimally balance light transmittance and PCE of the devices. Therefore, the product of AVT and PCE is used to quantify such performance.⁷ In Fig. 7b we compare the 4 cm^2 translucent modules with reported translucent cells and semi-transparent small size perovskite cells and modules using AVT versus $\text{PCE} \times \text{AVT}$ (light utilization efficiency).

3.3 Color neutrality

Aside from light transmittance or luminance of the modules, an important aspect in applications of TPV, is also their color or chromaticity. Considering the homogeneous transmittance spectra in the visible region (Fig. 6), we can express the color neutrality in the CIE color space. Here, the laser patterned and mechanically scribed modules with 50 % targeted transparent area have CIE 1931xy coordinates following white light illumination of (0.348, 0.348) and (0.348, 0.346), respectively. In contrast to semi-transparent solar cells based on a thin perovskite layer or high band-gap perovskites, color neutral modules open up a whole variety of applications. This includes the implementation of a tinted packaging to allow for a selective adaptation of the transmitted light color which as a subtractive method would reduce achieved LT.

3.4 Translucent module processing cost

The manufacturing of translucent perovskite modules using the method presented in this work would require one additional process step for the partial area ablation. The low cost of this processing step and the versatility for custom made designs is advantageous over fabrication methods where the translucent pattern is created during the coating and deposition processes of the module fabrication. The cost of the lost materials due to ablation is rather low considering cost estimations for perovskite solar modules in the range of 30–50 US $\$/\text{m}^2$ with a material share of about 75 %.^{16,24} Based on these estimations¹⁶, with a high performance laser ablation step (1MHz repetition frequency, 100 μm spot size, 25 % pulse overlap) a translucent module with 50 % TAR can be processed within 90 s/m^2 at an additional cost of 5–10 US $\$/\text{m}^2$. The ablated material of a few gram per square meter from the perovskite module can be collected for recycling as in the P1–P3 patterning steps. This demonstrates that perovskite modules offer the advantages of TFPV including the possibility to vary neutral color transparency of modules while maintaining the efficiency of the opaque area at a low additional cost.

Fig. 5 a) Schematic showing cross-section of translucent module with red line showing the simulated PCE change over opaque area b) Simulation results showing fit between decrease in normalized opaque area PCE and decrease in W. Fitting lines for damage edge width of 5–80 μm is shown, where most points fit the 20–40 μm region.

Fig. 6 Specular and total transmittance curves for laser and mechanically patterned translucent modules and for reference glass/ITO sample. Inset shows images of both laser (left) and mechanically (right) patterned translucent modules with a targeted transparent area ratio of 50 %.

4 Conclusions

By the addition of one processing step of the industry established fabrication method of laser or mechanical patterning, highly efficient perovskite modules can be transformed into color neutral, translucent modules. Here, the resulting translucent modules achieve a light transmittance of up to 37 % and maximum power point power conversion efficiencies of 5.1 %. Through detailed characterization of the fabricated modules, we determine the patterning method, patterning design and perovskite stack to be the three main factors in limiting light utilization efficiency of these translucent devices. Finally, laser and mechanical patterning show comparable results with laser patterning offering faster processing and variation and precision in patterning design. The presented fabrication methods have the advantage to introduce transparency to perovskite devices by combining freedom in patterning design and level of transparency with an ease of transferability of advances in opaque module performance, encapsulation and stability to translucent modules. Further device and process optimization is going to reduce the detrimental effects of the limiting factors and leading to an improvement of the performance of translucent perovskite modules towards theoretical boundaries.⁷ Progress in solar cell stability and a sustainable management of material toxicity will determine the potential of perovskite solar cells for future applications.

Silicon based photovoltaics with transparencies of up to 20 % are already part of the BIPV market. However, such technologies are not easily adapted to consumer preference of various shapes and translucency without apparent design patterns.¹⁵ This report and previous studies on small area cells exhibit that perovskite solar cells offer a versatility in design and color while maintaining high power conversion efficiency in comparison to other available transparent PV technologies.

Fig. 7 a) Measured transmittance of light incident on the transparent area. The errors are originating in the determination of the extension of the transparent region. b) Comparison of measured PCE versus LT or AVT⁷ for the translucent modules and cells, and published data on semi-transparent perovskite devices (adopted from Ref.⁷).

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Conflict of interest

There are no conflicts to declare.

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Table of content

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This work defines possibilities and limitations of industry known ablation process applied to perovskite PV resulting in translucent and efficient modules.