

FLEXIBLE MORPHOCOLOR® TECHNOLOGY INTEGRATED FOR COLOURED PEROVSKITE SOLAR CELLS

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ABSTRACT: The microstructured flexible film based MorphoColor® technology has been developed to be integrated into a flexible perovskite solar cell device. The concept is that the conducting front contact is substituted by a conducting interference layer coating on the microstructured substrate. This enables a MorphoColor® to be combined on a single flexible substrate with or as the conducting front contact.

A high angle dependent colour stable terracotta-coloured flexible microstructured PET film based on ITO and TiOx sputtered layers with a measured sheet resistivity of $22 \Omega/\square$ was designed and produced to match a conventional terracotta tile colour. Spin-coated perovskite, however, formed a non-conformal, inhomogeneously thick layer on the coated microstructured PET-based substrate. Thus, for a perovskite device to be manufactured on top of this microstructured system the perovskite should be deposited with a more conformal coating method (e.g. sputtering or spray coating). Alternatively, the configuration of the sputtered coatings has been adjusted so a conventional MorphoColor® coating with insulating dielectric materials is coated on the microstructured PET-side, and the conducting ITO coating is coated on the opposite smooth surface of the PET.

KEYWORDS: BIPV, ITO, sputtering, interference layers, coloured PV, perovskite

1 INTRODUCTION

The licensed technology MorphoColor® [1] used for coloured solar modules relies on a sputtered thin-film filter in combination with micro-structured substrates and is inspired by the morpho-butterfly [2, 3]. The advantages of MorphoColor® are the high saturated colours, high angle-dependence colour stability and minimal efficiency losses (5-10 %). These parameters are critical to ensure a satisfying aesthetic appearance yet maintain a high conversion efficiency of the solar module.

The MorphoColor® technology has been highly developed on textured glass and more recently on micro-structured film [4]. The glass-based technology is a commercial product with a wide colour palette, fabulously showcased by the recent installation at the St. Pauli Millerntor football stadium in Hamburg [5]. The flexible film technology, where the PET (polyethyleneterephthalat)-based film system is laminated into a glass-module system, is under development for crystalline silicon photovoltaic, PV, modules.

Spectrally matching MorphoColor® to a perovskite technology has previously been demonstrated [6]. The new goal here is to *integrate* MorphoColor® technology into a *flexible* perovskite solar cell. The concept is that the conducting front contact is substituted by a conducting MorphoColor® coating. This enables a MorphoColor® to be combined on a single flexible substrate with or as the conducting front contact. Thus, the flexible micro-structured substrate integrates both the electrical front contact and enables a colour-designed cell ready for processing with a thin film solar cell technology.

The concept is relevant as it has the potential to yield a product with a lower number of manufacturing steps to produce in roll-to-roll systems, which could be then processed into a perovskite flexible module. Due to the flexible and light structure and the designed colour the technology can be used as a 3D-shaped solar module, suitable, for example, as a curved roof-tile for BIPV or a

formed car bonnet in VIPV.

2 METHOD

2.1 Application and initial investigation of the layer stack

Lacquer printed PET substrates of thickness $125 \mu\text{m}$ are provided by temicon GmbH. The microstructure of the lacquer used is created using nanoimprint lithography on a roll-to-roll system [7].

For the conductive interference layer systems, sputter coating was carried out on the Fraunhofer ISE vertical inline plasma sputter pilot line using DC magnetron planar targets (ITO (10 wt.% SnO_2 , 90 wt.% In_2O_3) und TiO_2 (99.5 %, purity)). A second ITO target (ITO (3 wt.% SnO_2 , 97 wt.% In_2O_3)) was also trialled on the horizontal inline plasma system sputter pilot line. The oxygen flow rate was optimized to attain maximised conductivity and solar transmittance for each material thin film. The systematic investigation of the ITO optimisation is presented here.

To determine the refractive indices of the thin film materials used, corresponding individual layers are deposited on glass substrates, and the spectral reflectance and transmittance are measured by a Bruker VERTEX 80 Fourier spectrometer equipped with an integrating sphere. The refractive index data are then fitted to the measured data via the exponential Cauchy model [8] or in the case of ITO a Cauchy and Drude model [9, 10]. For the determination of the individual layer thicknesses of an applied layer stack, a flat glass substrate is co-coated in each case. The reflectance and transmittance spectra of this reference are determined by Fourier spectrometry, after which the layer thicknesses are fitted using the transfer matrix formalism.

The sheet resistance of the film stacks and the individual layers were measured by a 4-point probe.

The standard dielectric configuration of the layer stack is similar to the systems described in [2] consisting of sputtered of Si_3N_4 , TiO_2 , and SiO_2 .

2.2 Designing the thin film stack

A genetic algorithm is used to assist the design of MorphoColor® layer systems. The individual generated layer designs are evaluated based on the evaluation criteria ΔE (the difference in colour of the input $L^*a^*b^*$ colour parameters [11] and the actual $L^*a^*b^*$ parameters of the designed system), the weighted spectral transmittance (in the AM1.5 pervoskite absorbing spectra) and the colour angular stability of the designed film system (defined as the variation of the hue [12] as function of angle of incident).

For the design of the initial conducting terracotta layers (described in 3.2 and 3.3 and shown in the left-hand schematic of figure 4) spectral data from a pervoskite device was not matched but only an “absorbing” final layer with a constant refractive index of 1.5 (similarly used when designing layers for a silicon solar device system).

For photographs and colour analysis of these sputter-coated systems, a black absorbing lacquer of refractive index 1.5 was sprayed on the coated, structured side and the reflectance and photographs were taken from the uncoated side of the film-substrate.

For the design of the modified film system (described in 3.4 and right-hand schematic in figure 4) the pervoskite thin film system with the pervoskite absorbing curve based on [13] was used in the model (see [14] for details). When a weighted pervoskite spectrum is used it is also weighted with the absorption spectra from the same data.

2.3 Processing the pervoskite solar cell device

With the goal to examine the subsequent processing of the spin-coated pervoskite-stack on the microstructure flexible substrate rather than to have an optimised operation device, the clean temicon microstructured PET film and flat glass, both sputter-coated with conducting MorphoColor® are further processed with the following procedure. PEDOT (Clevios™ AI 4083), an organic semiconductor, was coated onto the substrates. For each 50 mm × 50 mm substrate, 1 mL of PEDOT solution was filtered through a 0.45 μm PTFE filter and deposited onto the substrate to ensure complete surface coverage.

The substrates were then statically spin-coated at 3000 rpm for 60 s, with an acceleration of 1000 rpm s⁻¹. Since PEDOT is water-based, the spin coating was carried out in the fume hood in the clean room. Finally, the PEDOT substrates were annealed in air at 140 °C for 10 minutes.

For the pervoskite layer, a triple-cation pervoskite with the stoichiometry $\text{Cs}_{0.05}\text{FA}_{0.85}\text{MA}_{0.1}\text{PbI}_{2.9}\text{Br}_{0.1}$ was used. To prepare 1 mL of a 1.45 M pervoskite precursor solution, 18.8 mg of CsI (99.9%, trace metals basis, Sigma-Aldrich), 701.88 mg of PbI_2 (99.99%, trace metals basis, TCI), 16.2 mg of MABr (>99.99%, trace elements basis, Dyenamo), and 212 mg of FAI (99.99%, trace elements basis, Dyenamo) were weighed into a vial. A magnetic stirring bar was then added, followed by DMF and DMSO in a 4:1 volume ratio (800 μL DMF and 200 μL DMSO). The solution was stirred for approximately 2 hours inside the glovebox on the day of the experiment to ensure complete dissolution.

For deposition, 300 μL of the precursor solution was statically spin-coated using a two-step program:

- Step 1: 2000 rpm for 10 s, with an acceleration of 200 rpm s⁻¹
- Step 2: 6000 rpm for 20 s, with an acceleration of 200 rpm s⁻¹.

The entire pervoskite preparation and deposition

process was carried out inside a nitrogen-filled glovebox.

300 μL of chlorobenzene (CB; 99.8%, anhydrous, Sigma-Aldrich) was rapidly and continuously dispensed onto the substrate 7 seconds before the end of the spin-coating process to induce crystallization. Following deposition, the substrates were annealed at 100 °C for 60 minutes.

Scanning electron microscope cross sections of the fully coated substrates were taken using a FIB-SEM Zeiss Auriga 60.

3 RESULTS

3.1 Sputtered ITO optimization on flexible microstructured PET

The thin film ITO and TiOx sputtered on the flexible temicon PET substrate are amorphous.

The two ITO targets (10 wt.% and 3 wt.% SnO_2) were systematically trialled with a variation of oxygen flow rate. The resulting thin films resistivity and absorption coefficient per nm is plotted on a graph for the two different ITO targets and oxygen flow rate combinations (see figure 1).

An optimum film using a 10 % wt. SnO_2 target and a 6 sccm O_2 flow rate (Ar flow rate of 100 sccm and pressure 1.8×10^{-3} mbar) yielded an absorption of 4.8×10^{-4} % per nm and a resistivity of 5.8×10^{-4} Ωcm was then chosen to be used in conducting interference film systems subsequently described in this paper.

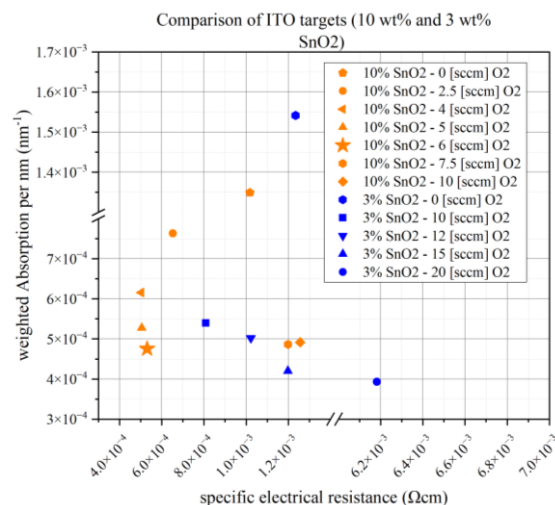


Figure 1: Plot of averaged absorption % per nm for a pervoskite AM1.5 weighted absorption spectra versus specific electrical resistivity for two different ITO targets (10wt.% SnO_2 and 3wt.% SnO_2) as a function of oxygen flow rate.

3.2 Terracotta conducting MorphoColor film thin stack design

A conductive MorphoColor thin film system with a terracotta-coloured reflectance, constant over a wide viewing angle and good electrical conductivity (sheet resistance was measured as 22 $\Omega/\square$) was achieved on a micro-structured film by using ITO and TiOx as alternating low and high refractive index layers in a 5-layer system. For this film system however, there is a high parasitic spectral absorption. This is because the overall multi-layer system consists in total of 250 nm of

amorphous ITO leading to a measured parasitic spectral absorption of 9 %.

To reduce the amount of ITO in the interference film stack, alternative film systems were investigated which consisted of the standard dielectric film system and only the final layer of conducting ITO (130 nm in thickness). This increased the sheet resistance of the system to $45 \Omega/\square$ (which is still well below a $60 \Omega/\square$ of commercial flexible PET products) but decreased the total parasitic absorption to only 5%. It should be noted the ITO deposited in the multi-layer stack system had less absorption losses than in the individual ITO films. This is hypothetically due to several reasons including that the deposition temperature is higher during than in the multi-layer system due to the longer sputtering process, the growth of the ITO film is on a sputtered layer (rather than the substrate – in this case the microstructure lacquer on the PET) and a longer exposure

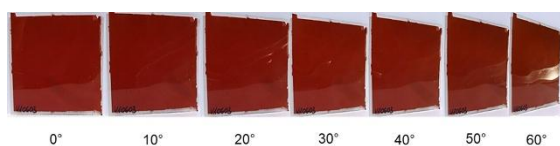


Figure 2: Angular behavior of the colour of a 6 cm² structured temicon film coated with a conducting MorphoColor® thin-film system. The viewing angle increases from 0° to 60° in 10° increments. The two-colour regions in the respective images can be attributed to the reflection of the surroundings.

to the partial oxygen plasma during sputtering of the multi-layer system.

Figure 2 shows a photograph of the conducting thin-film system deposited on the temicon micro-structured film and sprayed with a black lacquer (to simulate an absorbing solar device layer) photographed at 0-60° angles to the normal. The constant colour observed at increasing viewing angles demonstrates the high angle-dependent colour stability of these conducting MorphoColor® film systems on these microstructured PET films.

3.3 Spin-coated pervoksite devices on the MorphoColor® thin film stacks

Next the compatibility of the spin-coated perovskite on the coated structured PET substrate was trialed. The same conducting MorphoColor® thin film system was deposited on both glass and temicon structured PET and subsequently spin coated with a perovskite device. SEM cross sections are shown in figure 3 and the schematic of the whole film system can be seen in the left-hand schematic in figure 4.

It can be seen in the SEM pictures that whilst the sputter coated-smooth glass is homogeneously covered by the spin-coated layers, the microstructure on the PET film effects the spin-coating of the layers so that they are unconformably thick in the “troughs” of the microstructure and not fully or very thinly coated on the microstructure “peaks”.

3.4 Design and Production of alternative integrated MorphoColor configurations for the perovskite flexible device.

As the microstructured surface proved difficult to spin-coat the subsequent perovskite device layers, an alternative MorphoColor® multi-layer system was

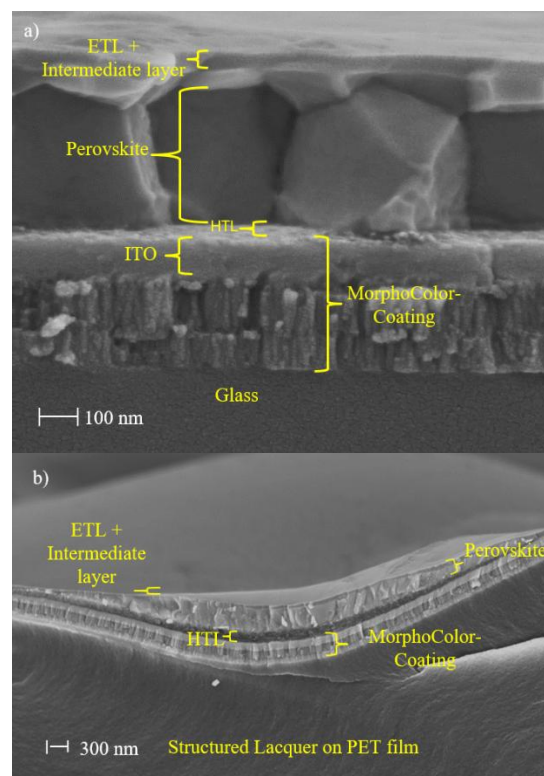


Figure 3: Scanning Electron Microscope pictures of the thin film system on a) a smooth glass substrate b) a micro-structured PET film. The coating in this case is as shown in the schematic figure 4a.

proposed. The original system is shown in figure 4a and the alternative system in figure 4b. The alternative modified version has a dielectric interference film on the microstructure side of the temicon PET substrate. On the smooth side of the PET substrate the ITO is then sputter coated and subsequently processed with the spin-coated perovskite layers. The spin-coated layers are then conformal as they are not on the microstructured side.

Microstructured PET films were coated with this proposed configuration with terracotta, blue and grey interference film stacks on the microstructured side and an ITO exhibiting a conductivity of $40\text{-}60 \Omega/\square$ on the smooth

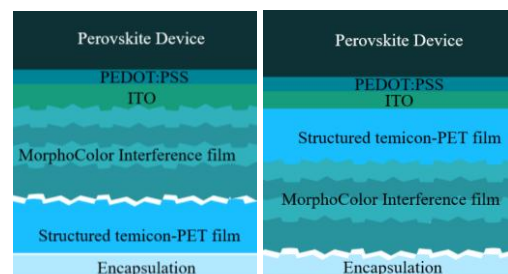


Figure 4: Left: schematic of the complete film system configuration used in the coating shown in the SEM pictures and described in sections 3.2 and 3.3 where the MorphoColor® coating is conducting and integrated into the pervoskite device. Right: schematic of the modified thin film configuration, to avoid processing on a microstructured surface, where the MorphoColor® remains on the microstructured side but the conducting coating and rest of the pervoskite device is processed on the smooth side of the same PET temicon film.

Table I: Three different coloured MorphoColor® coatings deposited in the configuration shown in the right schematic of figure 4. The simulated loss in transmittance (ΔT_{sol}) in the portion of the AM1.5 solar radiation spectrum that is absorbed by a perovskite solar device compared to an equivalent flexible PET system without the MorphoColor® coating is tabulised.

Colour	ΔT_{sol}
Terracotta	7.0%
Blue	3.7%
Grey	2.7%

side, the loss in transmission when integrated into a perovskite device are calculated and compared to that of a non-coloured flexible perovskite device on a standard flexible PET substrate in table I.

4 CONCLUSIONS

Conducting thin films of sputtered amorphous ITO were optimized on temicon microstructured PET substrate films to find suitable conductivity in conjunction with minimized parasitic absorption in the spectral range of a typical perovskite absorption spectrum. Optimum deposition parameters using a 10 % wt. SnO₂ target and a 6 sccm O₂ flow rate were identified.

Electrically conducting terracotta-coloured coatings on microstructured flexible PET substrates were made using ITO/TiO_x/ITO/TiO_x/ITO and standard dielectric MorphoColor topped with ITO film.

Due to the higher absorption of the amorphous ITO on the flexible substrate which cannot be exposed to a subsequent anneal due to temperature restrictions of the substrate, it was determined to be spectrally optimal (less parasitic absorption) to use standard dielectric interference coating with the conducting ITO as the top layer, to minimize the absorption of the film system.

Thus, the flexible micro-structured substrate integrates both the electrical front contact and enables a colour-designed cell ready for processing with a flexible thin film solar cell technology. The proven high angle dependent colour stability terracotta colour which was achieved blends into a roof of conventional terracotta tiles.

The direct integration of the conductive MorphoColor® thin-film system into a perovskite solar cell module by means of spin coating of the functional layers of the perovskite solar cell was only successfully realized on a planar substrate. For the micro-structured surface, the spin-coated perovskite could not be homogeneously and conformally coated. One option would be to use a more conformal coating technique for the perovskite such as a sprayed or sputtered process, which allows more conformal coating.

An alternative second option was identified where the multi-layer system with the interference film and microstructure is on the front-side and the conducting layer on the opposite smooth side of the PET substrate. Microstructured PET films were coated with this proposed configuration with terracotta, blue and grey interference film stacks on the microstructured side and an ITO on the smooth side, the loss in transmission when integrated into a perovskite device were calculated and compared to that of a non-colored flexible perovskite device on standard PET. Therefore, incorporating these films should yield perovskite solar devices with 93 % for terracotta and 95.3% for blue the performance compared to the non-

coloured device.

The different thin film configurations of the MorphoColor® coating allow a single PET microstructured temicon film to be used to manufacture a coloured flexible perovskite solar device which then can be encapsulated. This limits the thickness and number of films being encapsulated into these flexible devices.

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ACKNOWLEDGEMENTS

This work has received funding from the Horizon Europe Framework (HORIZON) research and innovation program under the grant agreement No 101122327 "SMARTLINE PV".