

Enhanced Selective Contact Behavior in a-Si:H/oxide Transparent Photovoltaic Devices via Dipole Layer Integration

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Transparent photovoltaic (TPV) devices have the potential to revolutionize photovoltaic (PV) technology by enabling on-site generation while minimizing visual impact. However, a major challenge in the development of TPV, as well as for many PV technologies, is the open-circuit voltage (V_{oc}) deficit, which limits their efficiency. In this work, the development of wide-bandgap inorganic-based TPV devices is reported with a focus on low-cost, earth-abundant, stable, and nontoxic materials. The device structure consists of an ultrathin hydrogenated amorphous silicon (a-Si:H) absorber and metal-oxide layers as selective contacts. Herein, novel approach is presented to significantly improve device performance, especially in V_{oc} , by introducing molecular dipoles in the device electron-transport layer. By incorporating polyethyleneimine or poly(amidoamine) G_1 and G_2 dipoles, V_{oc} (from 410 mV up to 638 mV) is significantly increased without sacrificing the average photopic transmittance of the device, leading to a record efficiency for this particular approach in TPV. Measurements confirm excellent long-term stability. This approach can potentially allow tuning the work function of the selective contacts enabling the use of low-cost, earth-abundant materials that are not optimized for a particular absorber. Furthermore, this solution circumvents the issue of low V_{oc} by a simple interface treatment.

scaled up into industrial processes, presenting a high technology readiness level.^[2] Recently, different works have explored the potential of using ultrathin hydrogenated amorphous silicon films as UV-selective absorbers for TPV applications, improving on power conversion efficiency (PCE) with respect to state-of-the-art devices, which have mostly relied on oxide-based technologies.^[3–5] This alternative can be implemented without sacrificing much in terms of transparency due to the ultrathin absorber thickness and the use of transparent and color-neutral oxide-based charge-transport layers (CTLs).^[6,7] This potentially allows its application onto windows, facades, and building surfaces, where it can generate electricity from sunlight without significantly affecting the appearance of the substrate.^[8] These types of transparent solar cells open up new possibilities for integrating solar energy into architectural design, sustainable construction, and the development of multifunctional optoelectronic devices.^[9]

Moreover, it is important to highlight that reducing manufacturing costs is a key objective allowing for this technology to become cost-effective, more accessible and competitive in the energy market.^[10] By addressing these challenges, hydrogenated amorphous silicon solar cells could become a more attractive option for clean and renewable energy generation in the future.^[11]

1. Introduction

Currently, one of the promising approaches in the field of materials for solar cells is the use of amorphous silicon for the manufacture of transparent solar cells.^[1] Amorphous silicon is a noncrystalline form of silicon that has the advantage of being highly transparent to visible light and that has already been

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The integration of dipoles^[12] has been explored in organic electronic devices, such as organic solar cells,^[13,14] organic light-emitting diodes and ferroelectric materials,^[15] to enhance carrier injection, and improve charge-carrier mobility.^[16] In solar cells and other electronic devices, careful incorporation of dipoles through material selection, deposition techniques, and device engineering optimizes their performance.^[17] Different research works have evidenced that they serve as passivation layers on solar cell surfaces, reducing charge carrier recombination and improving efficiency.^[18,19] Furthermore, they facilitate charge separation in solar cells, leading to more efficient charge extraction and power generation. Ongoing research continues to explore innovative ways to integrate dipoles, pushing the boundaries of efficiency and performance further in these devices. As for inorganic solar cells, there has been less research on the matter, although it seems a very promising approach in order for photovoltaic (PV) devices to meet their theoretical limits and maximize PCE especially by maximizing their open-circuit voltage (V_{oc}).

In this work, different molecular dipoles have been implemented into device architectures, specifically at the electron-transport layer (ETL)/absorber interfaces of a-Si:H/oxide TPV devices. These devices have been characterized and compared against a reference structure without the interfacial dipoles. The reference device architecture consists of soda lime glass (SLG)/fluorine-doped tin oxide (FTO)/aluminum-doped zinc oxide (AZO)/ZnO/a-Si:H/ V_2O_5 /AZO and the studied devices incorporate molecular dipoles embedded as an interfacial nanometric film between the AZO and ZnO films. TPV characterization of these devices has been carried out. Specifically, the temperature-dependent V_{oc} characteristics have been studied and suggest that the V_{oc} improvement can be correlated to the dipolar moment of the interfacial layer at the electron-selective

contact. Additionally, scanning transmission electron microscopy and energy dispersive X-ray spectroscopy (STEM–EDX) measurements have been carried out. Optical characterization has confirmed the high transparency of devices and that the incorporation of interfacial dipoles has negligible effect on the optical characteristics of the devices. This research is potentially relevant for the advancement of thin-film TPV technologies as well as for conventional PV technologies, and other technologies in general that rely on advanced CTL/active layer/CTL architectures.

2. Results and Discussion

Planar ultrathin TPV devices are fabricated with the following architecture: SLG/FTO/ETL/a-Si:H/ V_2O_5 /AZO. The most innovative focus of this study consists in the introduction of molecular dipoles at the ETL. A reference device is prepared using an ETL composed of AZO/ZnO and in parallel samples are fabricated intercalating molecular dipoles AZO/dipole/ZnO. Three different molecules are studied: a branched conjugated polymer, polyethyleneimine (PEI), and two different generations (G_1 , G_2) of the dendrimeric macromolecule poly(amidoamine) (PAMAM) (Figure 1). For the different samples, AZO, ZnO, and V_2O_5 films have been deposited in parallel in a single process to be able to make accurate comparisons focusing on the effect of the dipolar molecule incorporation. In this case, a 30 nm a-Si:H film is deposited while a hole-transport layer (HTL)/TCO stack of V_2O_5 /AZO is deposited on top to complete the cells (Figure 1). It is relevant to stress that these devices are made without any catalogued critical raw material and at relatively low temperatures, being 220 °C the maximum process temperature used for the deposition of a-Si:H. Additionally, the different deposition techniques used allow for the conformal and

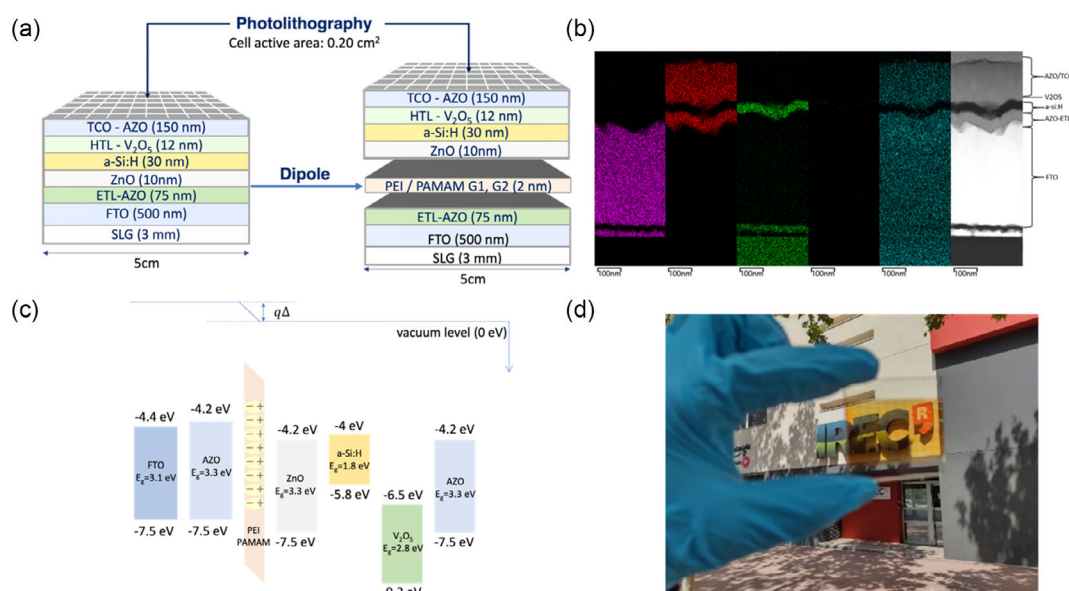


Figure 1. a) (left) Schematic illustration of the TPV device reference architecture (not to scale); (right) schematic illustration of the TPV device reference architecture with dipole layer. b) STEM–EDX—cross section of reference device. c) Band diagram of the different films that conform the reference device. d) Pictures of completed dipole device, taken outdoors in broad daylight. The picture shows the TPV device, showing a board with the Institut de Recerca en Energia de Catalunya (IREC) logo located at the building entrance.

controlled deposition of the different nanometric films that compose the device.

We begin by analyzing the optical properties of the completed devices and later continuing with the different optoelectronic characterizations performed.

2.1. Optical Characterization and Current Density–Voltage and V_{oc} – T Characteristics of TPV Devices

In this section, the complete device structure is characterized by measuring their reflectance and transmittance by optical spectrophotometry. The complete devices were fabricated with the same conditions using different dipole layers in the architecture including PEI and dendrimer PAMAM Generation 1 (G_1) and Generation 2 (G_2) leaving the rest of the structure unchanged. The measurement allows the determination of the transmittance spectra of complete TPV devices, which will serve as input for calculation of average photopic transmittance (APT) and color rendering index (CRI).

The optical measurements of reflectance and transmittance for the completed devices were conducted in the wavelength range of 300–800 nm as depicted in **Figure 2**. These spectra were employed to determine the values of APT and CRI, as per the equations provided in the methodology outlined by Traverse et al.^[20] All these parameters are summarized in **Table 1**. From the transmittance measurements, it has been possible to calculate the CIE color coordinates. The coordinates as well as the CIE color maps are provided in Figure S7, Supporting Information. As a standard in TPV characterization, from optical measurements and external quantum efficiency (EQE) measurements, we provide the photon balance check graphs in Figure S8, Supporting Information. These measurements confirm no over-estimation of generated photocurrent.

Light utilization efficiency (LUE) was calculated using Equation (1):

$$LUE = PCE \times APT \quad (1)$$

APT was calculated using Equation (2):

Table 1. Summary of TPV parameters extracted from optical and electrical characterization of the different devices studied in front illumination configuration.

Device	CRI	APT [%]	V_{oc} [mV]	J_{sc} [mA cm ⁻²]	FF [%]	PCE [%]	LUE [%]
Reference	89	39.8	407	5.1	39	0.80	0.32
PEI	89	39.9	535	4.8	45	1.17	0.47
PAMAM G_1	90	39.4	591	5.5	46	1.51	0.56
PAMAM G_2	89	39.5	638	4.8	49	1.50	0.59

$$APT = \frac{\int T(\lambda)P(\lambda)S(\lambda)d\lambda}{\int P(\lambda)S(\lambda)d\lambda} \quad (2)$$

where λ is the wavelength, T is the transmittance, P is the photopic response, and S is the solar photon flux (AM1.5 G). For detailed information about APT, LUE, CRI, and calculation, see ref. [20].

Figure 3 shows the improvement of the V_{oc} values and the other parameters as well as the efficiency of the solar cells. All the devices using dipolar interlayers exhibit a statistically relevant improvement in the PV performance, compared to the reference solar cell.

An essential factor in enhancing the performance of transparent devices is bifaciality (see Figure S1–S4 and Table S1, Supporting Information). This means that solar cells can generate close to the same efficiency regardless of the incident direction of light, even harnessing the energy reflected from surfaces. This allows us to consider superstrate configurations, which could be advantageous when depositing them on various substrates and streamlining processes. This becomes especially significant when considering industrial scalability for the next generations of TPV technology. Stability measurements were performed and confirmed excellent stability for the devices (Figure S5, Supporting Information).

Dipole layers used for this experiment are readily known as conjugated polyelectrolytes (CP). CPs are polymeric structures that possess charged groups of either positive (cationic) or negative (anionic) charge.^[21] Cationic polymers are preferred to

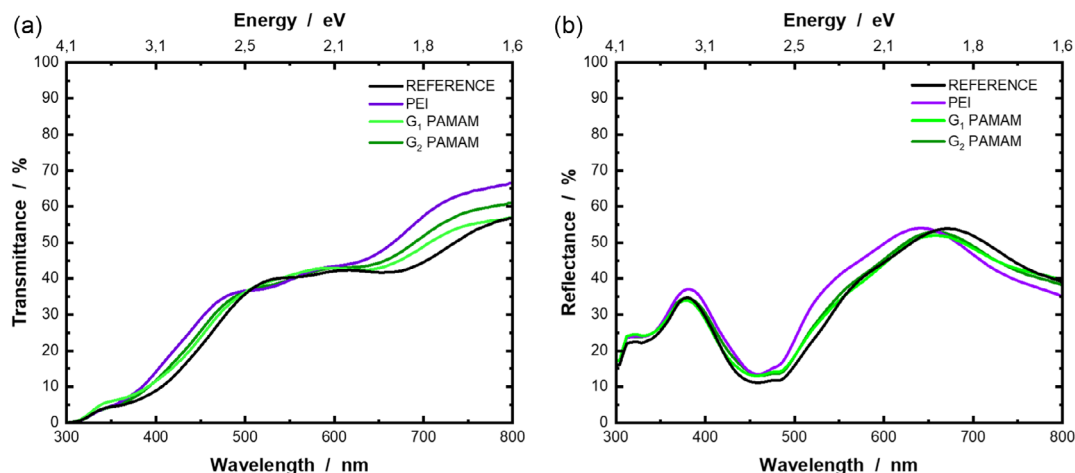


Figure 2. a) Transmittance spectra; b) reflectance spectra from 300 to 800 nm.

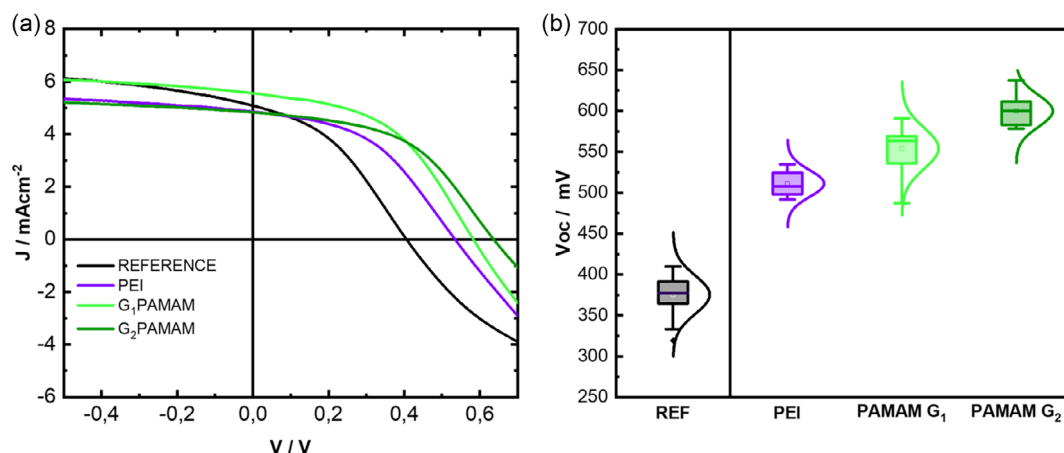


Figure 3. a) J - V characteristics of the studied devices; b) box plot showing V_{oc} statistics for all subcells measured in the different devices.

generate proper band alignment for electron extraction, therefore acting as ETLs. In contrast, anionic polymers are generally used for enhancing hole extraction properties.^[22,23] In both PEI and PAMAM films, presence of amino groups (i.e., $-NH_2$) with nitrogen in sp_3 hybridization provides a cationic nature to the film.^[19,24] Counter anion is understood to be critical to enhance the dipole effect on these cationic polymer sites.^[25] Nevertheless, Lewis basicity of amino groups combined with slightly acidic solvents (e.g., anhydrous methanol, pH 6) promotes the formation of a conjugated charged system necessary for the formation of dipole systems.

Equal chemical nature of the cationic sites, as well as counter anions in PEI and PAMAM G_1 and G_2 (i.e., protonated amino/methanolate systems), allows for qualitative interpretation of the PV results. In this sense, the total number of amino sites per monomer/macromolecule will be used as the main difference between dipole layers. For instance, branched PEI is generally drawn to have four amino sites per monomer. Dendrimers in contrast form hyper-branched treelike structures with increasingly higher surface amino sites per generation. For example, PAMAM G_1 has eight amino sites per macromolecule, while for PAMAM G_2 the number of amino sites is doubled to 16 per macromolecule (see

Figure 4). PV characterization has shown that as the number of amino sites per monomer or molecule increases, a significant increase in V_{oc} , fill factor (FF), and in general a significantly higher efficiency is obtained compared to the reference sample. This can be linked to the Helmholtz equation^[26]:

$$\Delta = \frac{1}{\epsilon} \cdot N \cdot p \quad (3)$$

The Helmholtz potential of the film is directly proportional to the number of dipoles (N) and dipole moment (p) and inversely proportional to the dielectric constant (ϵ). As the dipole is formed by the amino/ethanolate-conjugated system, the change in dipole potential is in essence driven by the number of dipoles of the film. This number will also depend on packing factor of monomers/molecules. Nevertheless, dipole potential is increased by a factor 2 as the dendrimer generation is increased.

Another significant result is the higher FF values of the solar cells implementing dipolar interlayers at the electron-selective contact. The FF value has been usually associated to the quality of the transparent electrode. Since all the devices studied here used the same TCO, the increased FF could be attributed to a

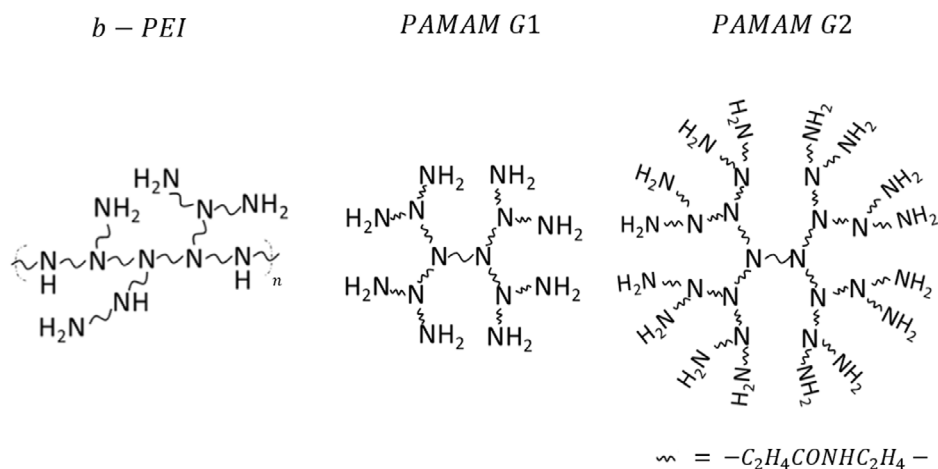


Figure 4. Schematic representation of branched polyethyleneimine (b-PEI) and polyamidoamine (PAMAM) Generation 1 and Generation 2.

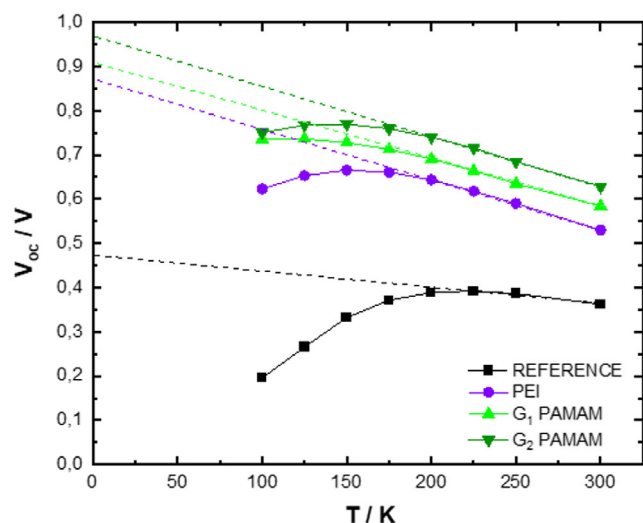


Figure 5. Temperature-dependent open-circuit voltage (V_{oc}) measurements in a-Si:H devices under 1 sun, the dotted lines represent the linear extrapolation to $T=0$ K, which gives an estimation of the activation energy E_a .

Table 2. Values of activation energy, saturation V_{oc} , and temperature for the studied devices.

Sample	E_a [V]	V_{oc}^{sat} [V]	T (V_{oc}^{sat}) [K]
Reference	0.47	0.41	225
PEI	0.87	0.66	150
PAMAM G_1	0.91	0.74	125
PAMAM G_2	0.97	0.79	150

much easier extraction of the photo-generated carrier (i.e., electrons at the electron-selective contact).

Temperature-dependent V_{oc} characteristics are shown in **Figure 5**. The electrical measurements were carried from 300 K down to 100 K, sampling every 25 K. At room temperature, the V_{oc} experiences a linear decrease. The extrapolated values of activation energy (E_a) for all the studied devices are presented in **Table 2**. The extrapolated E_a serves as an indicator of the maximum achievable quasi-Fermi level separation. If E_a approaches the absorber bandgap, V_{oc} is dominated by bulk recombination and thus interface recombination becomes negligible. Conversely, when E_a is significantly smaller than the bandgap, a substantial portion of total recombination is expected to take place at the interfaces. For the bandgap of 1.7 eV in a-Si:H, one would expect high E_a values in solar cells with optimum interfaces. Thus, the low E_a value of the reference device suggests that a high degree of recombination takes place at the interfaces. In contrast, it seems evident from the higher E_a values that interface recombination was significantly mitigated for devices incorporating dipole layers.

2.2. Spectral Response

Spectral response measurements were performed on the different devices studied to compare and analyze their response.

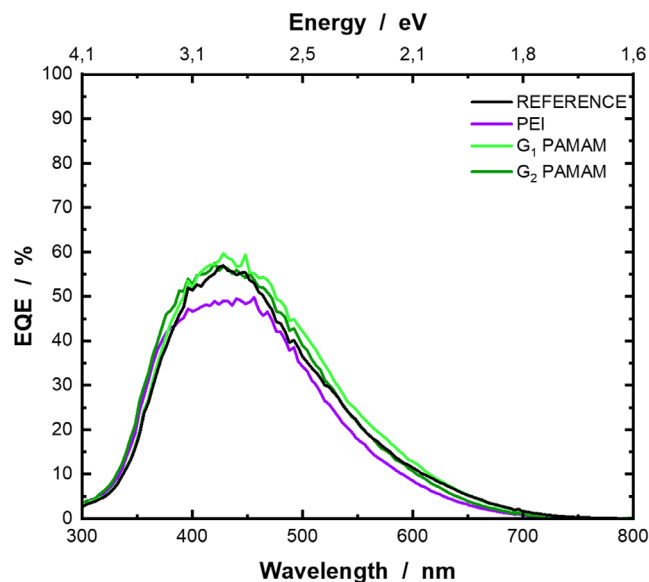


Figure 6. External quantum efficiency of the reference device compared with the devices incorporating molecular dipoles. The spectra show that the dipoles do not significantly affect the devices spectral response as compared with the strong modification observed in the increased V_{oc} and FF as seen on the J - V characteristics.

In **Figure 6**, the EQE resulting from spectral response measurements of the studied devices are presented. As already seen in previous experimental work,^[6,7] the response is centered at around 400–430 nm. From the EQE, a calculation of the integrated short-circuit current density (J_{sc}) is performed to compare this value with those extracted from the current density–voltage (J - V) measurements (**Figure S9**, Supporting Information).

When comparing the EQE of the different devices, it can be concluded that the introduction of interfacial dipoles did not affect the spectral response (hence, the J_{sc}) of the resulting devices. This fact is also supported when inspecting the J - V characteristics of the different devices. Looking at the transmittance spectra obtained from optical spectrophotometry, it is clear that the changes coming from the incorporation of the interfacial dipoles are negligible, and that indeed the most relevant change is the strong increase in V_{oc} and also a somewhat significant increase in the FF of the TPV devices, when compared against the reference. Going back to the EQE measurements, it is confirmed that increased dipolar strength does not affect the spectral response.

3. Conclusion

In conclusion, this study has addressed V_{oc} deficit. Circumventing this issue by incorporating dipole layers in the architecture using ultrathin a-Si absorber layers in TPV solar cells, the results have shown a significant improvement in the performance of these devices, the most important coming from a significantly strong increase in the V_{oc} and the FF, which has led to a higher overall solar cell efficiency. The studied dipoles PEI and PAMAM G_1 and G_2 have positively increased the V_{oc} and FF values. Future work could explore the use of higher dipole

momentum molecules to approach the V_{oc} limits of the light absorber. Furthermore, these alternative CTL structures minimize parasitic absorption, avoiding the need of harmful and costly dopants to provide quasi-Fermi level splitting. This increased V_{oc} has been postulated to stem from the increased dipolar moment that tunes the energetics at the ETL/absorber interface and thus improves on contact selectivity in contrast with the use of the standard unfunctionalized oxide ETL. Further studies will try to pinpoint the origin of this improvement to confirm the mechanism that enables the V_{oc} increase. Furthermore, the use of ultrathin a-Si:H absorber layers has allowed wavelength selectivity in the UV–blue range, providing promising results in terms of efficiency. In summary, this approach presents an interesting route for the improvement of devices reducing V_{oc} deficit with the introduction of an interfacial layer, offering alternative applications and an efficient power source for low-power devices, with the potential to expand the possibilities in the field of PVs.

The high V_{oc} was particularly noteworthy for devices with FTO/AZO/ G_2 -PAMAM/ZnO/a-Si:H(30 nm)/ V_2O_5 /AZO architecture, which demonstrated the highest value, close to V_{oc} = 640 mV, a PCE of 1.5%, and AVT of 40%. To the best of our knowledge, this result yields a record PCE when compared to state-of-the-art devices that take this approach toward UV-selective TPV using oxide-based CTLs. This compares with a reference device without any dipolar molecules showing PCE = 0.8% with V_{oc} = 410 mV only. We have also observed that the V_{oc} increases proportionally with the generation of the PAMAM dendrimer (i.e., proportional to the magnitude of dipole moment).^[27] These encouraging results bring the devices closer to their fundamental maximum V_{oc} , around 900 mV, as already achieved in opaque PV for p–i–n a-Si:H solar cells while avoiding the use of doped a-Si:H CTLs.

These results demonstrate the potential of molecular dipoles to significantly improve the performance of TPV solar cells^[28] and pave the way for the development of cost-effective and aesthetically pleasing TPV devices for distributed energy applications.

4. Experimental Section

Sample Preparation: Transparent solar cells (fabricated in this work present the structure SLG/FTO/AZO/(dipole)/ZnO/a-Si:H/ V_2O_5 /AZO. Commercial SLG/FTO substrates (Merck; R_{sh} = 10 Ω sq^{−1}) were used as substrate and transparent back-contact. Prior to deposition, substrates were rinsed in distilled water, dried in Ar flux and afterward cleaned by subsequent 10 min ultrasonic baths in isopropanol and ultrapure H_2O at room temperature, followed by a drying step in Ar flux and concluding with a 5 min O_3 treatment. A 75 nm AZO film was grown in an ALD system (Savannah S200, Cambridge Nanotech) at 135 °C, using precursors diethylzinc (DEZ 95%, STREM), ultrapure H_2O , and trimethylaluminum (TMA 99%, STREM) and using N_2 as purging gas. A total of 21 supercycles at a 19:1 DEZ:TMA ratio were performed, with 19 H_2O/N_2 /DEZ/ N_2 (20 ms/3 s/15 ms/3 s) cycles and 1 H_2O/N_2 /TMA/ N_2 (50 ms/1 s/20 ms/5 s) cycle. After the AZO deposition, the dipolar molecules studied were deposited on top (except for the reference device, without any dipolar film). For the PEI film, fresh solutions of PEI (Merck; 20%) dissolved in methanol (0.01 wt%) were spin coated at 5000 rpm for 50 s and immediately after were transferred to a hot plate at 90 °C to remove any solvent. This method yielded a 2 nm film. For the PAMAM films, fresh solutions of PAMAM (Merck; 20 wt%) were prepared dissolving PAMAM in methanol

(0.01 wt%). This was done identically for both G_1 - and G_2 -PAMAM. These solutions were spin coated at 5000 rpm for 50 s and immediately transferred to a hot plate at 90 °C to remove any solvent. The following step consisted in depositing an interfacial ZnO film of 8 nm, which helped making a better contact with the absorber/ETL and also served as a capping or protection layer so that the dipolar films were not degraded in the heat and plasma that the films would see in the plasma enhanced chemical vapor deposition (PE-CVD) of a-Si:H. Afterward, intrinsic a-Si:H ultrathin films of 30 nm were deposited by PE-CVD (Elettrorava) with a radio frequency power supply of 13.56 MHz at 6 W. Substrate temperature was measured at 220–250 °C during deposition, with a mixed 36/12 sccm SiH_4/CH_4 atmosphere and with deposition time of 4 min, respectively. To finish the solar cells, the HTL and front contact were deposited in a single sequential ALD process without breaking vacuum. V_2O_5 was grown at 165 °C, using tetrakis(ethylmethylamino)-vanadium (IV) ($C_{12}H_{32}N_4V$) (TEMAV 99%, STREM) and ultrapure H_2O using again N_2 as carrier and purging gas. Immediately after 125 supercycles H_2O/N_2 /TEMAV/ N_2 (2 s/3 s/3 s/3 s), without breaking vacuum and keeping T = 165 °C, AZO was deposited at a 19:1 ratio for 75 supercycles. Once the sample was fully grown, the active area cells were isolated using a photolithographic process followed by a wet chemical etching process that effectively isolated the cells from each other. First, photoresist at room temperature was spread on the surface of the samples and was statically spin coating for 30 s at 30.000 rpm. Immediately after, it was taken to a furnace at 105 °C for 20 min to evaporate the residual solvent. Then, UV photolithography was performed with a positive mask with square patterns defining a cell active area of 0.20 cm². Afterward, a two-step etching process was performed. The first stage used HCl at 10% to remove the AZO/ V_2O_5 layer. The final etching used an isotropic etching solution of HF + HNO_3 to attack and remove the rest of the stack (a-Si:H/ZnO/dipole/AZO) down to the FTO, which remained unaffected. As a result, the active area protected with the resin would remain and the subcells would be effectively isolated from each other allowing for J–V characterization without the need of a mask.

Characterization: Spectrophotometry measurements were carried out with a UV–vis–NIR spectrophotometer (Perkin Elmer Lambda L950) with an integrating sphere, in single-beam configuration, in transmittance and reflectance mode with spectral range from 300 to 800 nm to measure the completed device total transmittance and reflectance. This data was also used to calculate the device APT, CRI, and CIE color coordinates and color map. J–V measurements under illumination were carried out using an AAA class solar simulator with AM1.5G illumination calibrated using an National Renewable Energy Laboratory-certified Si reference solar cell (Abet Technologies Model 15 150). Electrical measurements were carried out with a source-measure unit (Keithley SMU 2400) in four-wire sense mode, remotely controlled by the software Tracer (ReRa solutions) using an IEEE 488 GPIB instrument control device (National Instruments GPIB-USB-HS). V_{oc} – T measurements were carried out in the same manner. Temperature was controlled using a Linkam system and measurements were carried out from 300 K down to 100 K in temperature intervals of 25 K. Spectral response measurements were acquired using a spectral response system (Bentham PVE300) calibrated with Si and Ge photodiodes. For optoelectronic measurements the samples were placed in a dark opaque matt to avoid any back-reflections that could alter the characterization of the transparent solar cells.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

dipoles, interfaces, open-circuit voltage (V_{oc}) deficits, selective contacts, transparent photovoltaic (PV)

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