




# Fluorinated carbon nanotubes: a low-cost hole transport layer for perovskite solar cells

Mustafa K. A. Mohammed<sup>1,\*</sup> , Ali K. Al-Mousoi<sup>2</sup>, Anjan Kumar<sup>3</sup>, Abdul Rasool J. Katae<sup>4</sup>, Omar A. Khaleel<sup>4</sup>, Duha S. Ahmed<sup>4</sup>, and M. Khalid Hossain<sup>5</sup>

<sup>1</sup>University of Warith Al-Anbiyaa, Karbala 56001, Iraq

<sup>2</sup>Department of Radiology and Ultrasonography Techniques, College of Medical Techniques, Al-Farahidi University, Baghdad 10011, Iraq

<sup>3</sup>Department of Electronics and Communication Engineering, GLA University, Mathura 281406, India

<sup>4</sup>Applied Sciences Department, University of Technology-Iraq, Baghdad 10011, Iraq

<sup>5</sup>Institute of Electronics, Atomic Energy Research Establishment, Bangladesh Atomic Energy Commission, Dhaka 1349, Bangladesh

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## ABSTRACT

The employment of cost-effective and durable structures is essential for the successful commercialization of perovskite solar cells (PSCs). Identifying a viable substitute for hole-selective materials (HSMs), which represent a significant expense in the production of PSCs, could provide a number of benefits. Carbon nanotube-based PSCs have shown promising potential as an alternative to conventional PSCs due to their unique properties such as excellent stability behavior to be potential for commercialization to produce green energy for human industries. One of the most crucial disadvantages of carbon derivatives as HSMs in CPSCs is their low hole mobility, which in the current study has been targeted. In the current study, to increase the efficiency of CPSCs, net carbon nanotubes (CNTs) were doped with nonmetallic fluorine via a facile synthesis method. It was found that introducing fluorine-doped CNTs (F-CNTs) as HSM for MAPbI<sub>3</sub> perovskite could reach up to an efficiency of 15.29%, higher than the efficiency of 13.70% in devices with a net CNT layer. By doping CNTs with fluorine, the charge-transfer resistance and series resistance are reduced, resulting in lower charge recombination at the perovskite/CNT interface. Also, the CPSCs with F-CNT film were more stable in ambient air because the F-CNTs covered more of the perovskite layer. The future trend of CNT-based PSCs is expected to focus on improving their performance, and exploring their potential for various optoelectronics.

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Mustafa K. A. Mohammed and Duha S. Ahmed have contributed equally.

Address correspondence to E-mail: mustafa\_kareem97@yahoo.com

## Introduction

Solar cells offer clean energy and an alternative to burning fossil fuels; however, recently, photovoltaics based on halide perovskites have surpassed those based on crystalline silicon photovoltaics in popularity due to their low cost, versatility, and superior energy conversion efficiency [1–5]. Perovskite-based photovoltaic devices have a high-performance rate due to a variety of characteristics, including but not limited to their high absorption coefficient, long carrier diffusion lengths, readily adjustable band gaps, and high carrier mobility [6–12]. Using a maximum output efficiency that is more than 25%, perovskite solar cells that are manufactured with metal halide appear to be a significant advancement over earlier generations [1]. Despite this advancement, a lot more work has to be done to make PSC inexpensive, stable, and have greater conversion efficiency. The cost-effectiveness of a material utilized as a charge transport layer in PSCs can be determined through various factors such as the cost of starting materials, the ease of processing, and the overall performance of the photovoltaic. One critical component of a perovskite solar cell is the hole-selective material (HSM), which plays a key role in extracting charge carriers from the perovskite layer and transporting them to the electrode. In perovskite solar cells, Spiro-OMeTAD material is commonly used as a HSM, which increases the final price of PSCs. Unfortunately, due to its high cost of synthesis, instability, and need of dopants because of low hole mobility, the use of Spiro-OMeTAD has been encountered a drawback [13–15]. To solve these obstacles, in addition to reducing cell cost and overall stability of the PSCs, the application of stable and available materials with a facile preparation is suggested. Carbon derivatives includes carbon black, graphene, graphite/amorphous carbon, and CNTs suggest low cost, high conductivity, eventual low-temperature processing, chemical stability and environmental kindness materials for using as HSM in PSCs [16–19]. Accordingly, researchers have focused their attention on carbon nanotubes (also known as CNTs), which possess exceptional electrical and optical characteristics and have the potential to bring about the needed improvement [20–24]. In conventional devices using CPSCs, the perovskite active film is placed between two charge-selective materials, one of which

is an electron-selective material (ESM), and the other is a hole-selective material (HSM) [25–28]. Early CPSC-based solar cells commonly use Spiro-OMeTAD and titanium dioxide as HSM and ESM, respectively. CNTs were investigated as a hole transport material as well as a contact electrode as an alternative to traditional p-type hole transport materials such as Spiro-OMeTAD and poly(bis(4-phenyl)(2,4,6-trimethylphenyl) amine (PTAA) due to their high charge carrier mobility and long-range transport [29–33]. The carbon nanotube was included in the design of the cell since there is a possibility that it may slow down the crystallization dynamics of the perovskite layer [23, 24, 33, 34]. In addition, carbon nanotubes have the potential to act as an ion blocker, preventing the passage of iodide into the metal halide perovskite absorber, and so significantly extending the stability of the CPSCs [35]. Despite being used as a hole-selective material, single-walled CNTs (SWCNTs) are not sufficiently charge selective for light-generated holes and cannot provide a strong field to avoid interfacial recombination losses; as a result, a low photogenerated voltage is produced [24, 33, 36]. For this reason, multi-walled CNTs (MWCNTs) were used in this study since their inner walls would remain unharmed and conductive even if their outer nanotube walls reacted with their immediate surroundings [37]. However, CPSCs that use pure CNTs as an HSM often exhibit subpar results due to a misaligned energy level alignment and the fact that the Fermi levels of pure CNTs are much higher than those of perovskite [38, 39]. This reduces the charge transport rate and diminishes the  $V_{OC}$ . In addition, since the pure-CNT coating has poor conductivity, the fabricated solar cells suffer from poor hole transport behavior. The capacity to alter the hole extraction and transfer abilities of MWCNTs can be manipulated using a number of different tactics, one of the most successful of which is the elemental doping approach. Researchers led by Yang have shown that enhancing the amount of boron doping in MWNTs greatly improves the charge extraction capabilities in CPSCs compared to the untreated ones [40]. The CPSC obtained an impressive conversion efficiency of 15.2% using boron-doped MWNTs. Using CsPbI<sub>2</sub>Br as the perovskite material and a P3HT-MWCNT hybrid as the HSM, Wang and colleagues created a CPSC that is both stable and efficient [41]. By acting as a barrier between the CsPbI<sub>2</sub>Br layer and the surrounding air,

the compact P3HT-MWCNT film considerably improves the layer's stability. This group was able to stabilize device efficiency at 8.85% and exhibit good stability by maintaining 85% of their original value for more than 240 h at room temperature. For better carrier mobility and conductivity, Seunghyun Baik and colleagues used multi-walled carbon nanotubes in Spiro-OMeTAD [42]. To prevent unwanted back-electron transport and make full use of the superior charge transport afforded by Spiro-OMeTAD/MWNTs, a hybrid including both Spiro-OMeTAD/MWNTs and pure Spiro-OMeTAD was developed. This group achieved a conversion efficiency above 15% by controlling the MWNT concentration. Kazunari Matsuda and his team proved that the addition of a buffer layer composed of graphene oxide and single-walled CNTs results in a remarkable increase in the overall cell performance of metal halide PSCs [25]. The buffer layer composed of CNTs and graphene oxide functions as an effective HSM. Because of the complementary characteristics of carbon nanotubes and graphene oxide, an energy conversion efficiency of above 13% was attained in a MAPbI<sub>3</sub>-based perovskite solar cell. An MWCNT electrode was employed in the HSL-free carbon-based PSC by Shihe Yang and colleagues [30]. The higher performance may be attributed to the hole transport pathway that is located at the interface of MWCNTs and perovskites. Following some initial tweaking, they were able to attain a performance of 12.7% and a very remarkable fill factor of 80%. Shuhui Yin and his colleagues used CNTs and carbon fiber as counter electrodes for HSL-free perovskite solar cells [20]. It was discovered that CNTs/carbon fiber material significantly improved the contact area between the perovskite and carbon layers, reducing charge-transfer resistance and enhancing hole extraction capability at the perovskite electrode interface. This group achieved a conversion efficiency of 11.8%.

In view of the well-defined morphology, nanosize, and high surface area of CNT, fluorine-modified CNTs (F-CNTs) with varying F ratios were simply prepared and used as hole transport layers for carbon-based PSCs. The MWCNTs have a F-rich surface, while the intact CNT inner walls provide efficient hole transportation from perovskite to the back electrodes. By incorporating CNTs in between the MAPbI<sub>3</sub> film and electrode, CNTs were partially embedded into the perovskite film, acting as a hole extraction and transfer pathway between them. The

CPSC performance and influences of F doping on PCE and ambient stability were elucidated through material characterizations and the current density–voltage (*J*-*V*) test. The CNT bridge approach provides an intimate MAPbI<sub>3</sub>/CNT interface, leading to a champion efficiency of 15.29% with an FF of 79.05%. The unencapsulated cells showed excellent stability under environmental conditions for 100 days when they were maintained in the dark.

## Experiments

### Functionalization of carbon nanotubes

Pristine CNTs with properties of 99 wt% purity, with 8 nm diameter, and 50–200 nm lengths, were bought from the SkySpring Nanomaterials, Inc., USA, and used as received. To prepare the functionalized CNTs, the CNTs and the acids mixture (sulphuric and nitric acids) in a volume (*v/v*) ratio of 3:1, were used. To 100 mg of pristine CNTs, a mixture of concentrated sulphuric (95%) and nitric acids (65%), which were obtained from J. T. Baker Company, and Central Drug House, England, respectively, were used. The resulting concoction was ultrasonically vibrated for 30 min before being diluted with distilled water, vacuum filtered through a cellulose nitrate membrane (0.22 μm), and baked at 110 °C overnight [43].

### Solution preparation

5 mL of ethyl alcohol (EtOH, 99.8%, Merck), 35 μL of 2 M hydrochloric acid (HCl, 37%, Merck), and 350 μL of titanium tetraisopropoxide (TTIP, 99.99%, Sigma-Aldrich) were mixed through vigorous stirring for 30 min at 0 °C. The obtained solution was used as a compact titanium oxide (c-TiO<sub>2</sub>) precursor. To prepare a mesoporous titanium oxide (ms-TiO<sub>2</sub>) precursor, 100 mg TiO<sub>2</sub> paste (30 NR, Dyesol) was dispersed into 850 mg EtOH by ultrasonication for 10 min and then by stirring overnight at room temperature (RT). 1.42 mol of lead iodide (PbI<sub>2</sub>, 99.9%, LumTec) was dissolved in 1 mL of mixed solvents of *N,N*-dimethylformamide (DMF, 99.8%, Merck) and dimethyl sulfoxide (DMSO, 99.8%, Merck) with a volume ratio of 9:1. The PbI<sub>2</sub> solution was heated at 80 °C for 40 min to ensure its complete solution. Thereafter, 1.42 mol of methylammonium iodide

(MAI, 99.8%, LumTec) was mixed with  $\text{PbI}_2$  precursor and stirred at RT for 15 min to form  $\text{MAPbI}_3$  stock solution. To prepare undoped CNTs-based HSM film, 4 mg of CNTs (diameter  $\sim 8$  nm, length  $\sim 5$ –20 nm, SkySpring Nanomaterials, Inc.) was dispersed in 1 mL of chlorobenzene (CB, 99.8%, Merck). To synthesize fluorine-doped CNTs (F-doped CNTs) materials, 250 mg of CNTs was added to 100 mL of diluted hydrofluoric (HF) acid solution with different concentrations (0, 0.3, 0.45, 0.6 M) of molar ratio. The obtained mixtures were stirred at room temperature for 24 h before being washed with distilled water until neutral. Then, the washed powders were dried at 100 °C for 24 h and used as F-doped CNT materials in the current study. 4 mg of each of the F-doped CNTs with different levels of doping was dispersed in 1 mL of CB and used as HSM stock solutions.

### Solar cell fabrication

A c- $\text{TiO}_2$  layer was formed on the pre-patterned and prewashed FTO substrate by spin coating 40  $\mu\text{L}$  of c- $\text{TiO}_2$  precursor at 4000 rpm for 30 s, followed by annealing at 450 °C for 1 h. To prepare the ms- $\text{TiO}_2$  layer, 60  $\mu\text{L}$  of m- $\text{TiO}_2$  precursor was spin coated over the c- $\text{TiO}_2$  layer at 3000 rpm for 20 s, followed by heating at 500 °C for 1 h. 60  $\mu\text{L}$  of  $\text{MAPbI}_3$  stock solution was coated on the m- $\text{TiO}_2$  with a speed of 1000 rpm for 10 s and 6000 rpm for 30 s to fabricate the perovskite layer. During the later spin-coating program, 150  $\mu\text{L}$  of CB anti-solvent was instantly poured on the perovskite layer to complete perovskite formation. To make HSM films, different HSM films were drop cast onto the  $\text{MAPbI}_3$  layer and dried at 70 °C for 5 min. A 100-nm gold electrode was evaporated on the HSM in high vacuum [44].

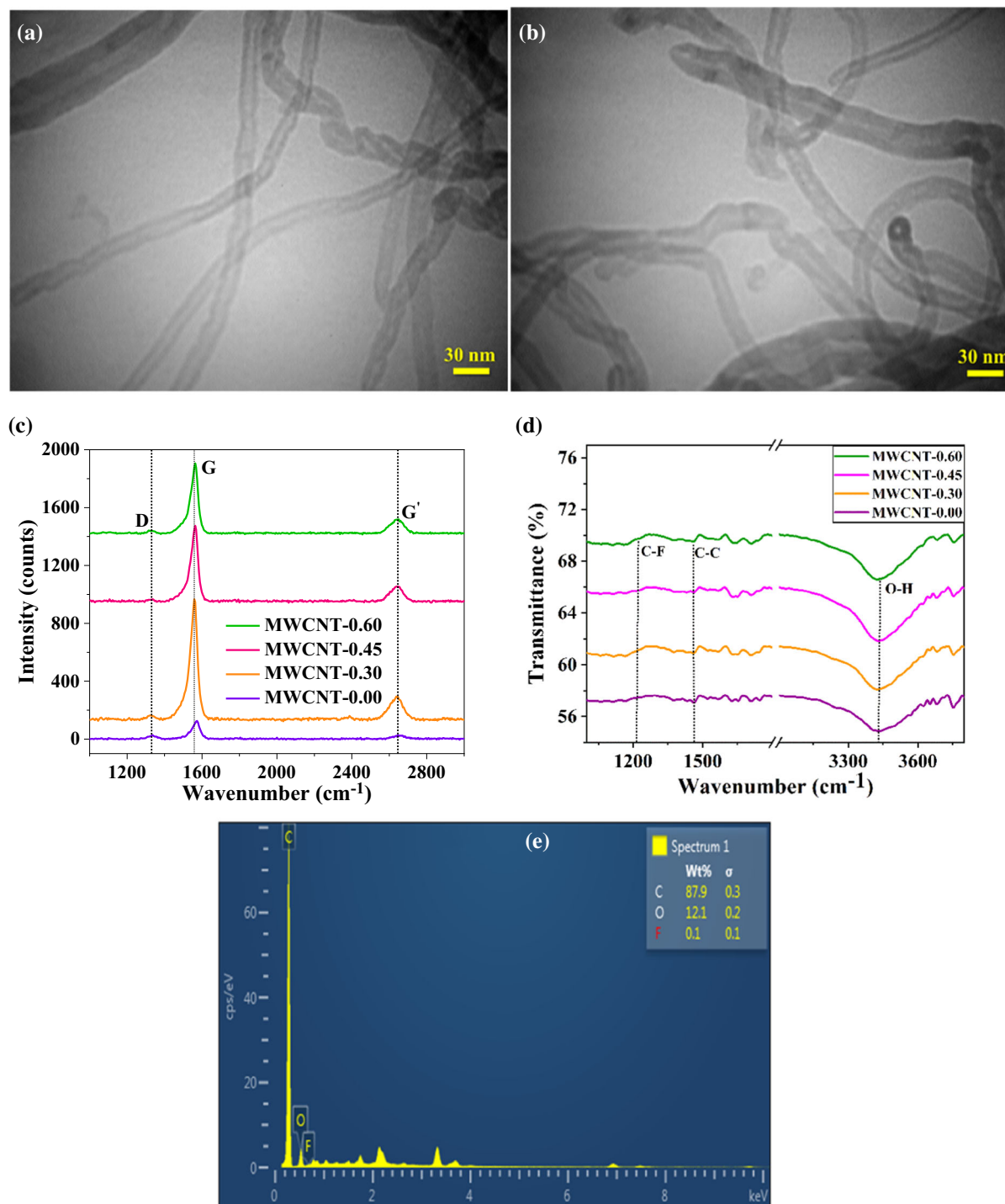
### Characterization

The absorbance spectra of samples were recorded by a Lambda 950 photo-spectrometer. An FLS920P Edinburgh spectrometer recorded the photoluminescence (PL) spectra of samples. To record cross-sectional FESEM images and EDS of samples, a Mira3-XMU FESEM equipment was used. In addition, to record TEM images of samples, a TEM Philips EM 208S instrument was employed. A PANalytical 80 X-ray machine was used to examine the XRD pattern of the perovskite layer. The FT-IR spectra of samples were recorded using an AVATAR

thermometer. The Raman spectra of samples were measured with a TakRam N1-541 TESCAN spectrometer. The  $J$ - $V$  curves of photovoltaic devices were investigated under simulated sunlight illumination. The PSCs were measured with an active area of 0.08  $\text{cm}^2$ . A Solartron 1260 galvanostat was used to measure the electrochemical impedance of perovskite solar cells under short-circuit and simulated sunlight irradiation conditions. The external quantum efficiency (EQE) spectra of PSCs were collected using a calibrated Newport IPCE instrument.

## Results and discussion

Doping CNTs with nonmetallic substituents has been deemed a useful approach for enhancing CNTs' performance since it can affect the electrical structure and electrochemical characteristics of the pure carbon nanomaterials [45]. In this work, we report the simple synthesis of fluorine-doped CNTs (F-CNTs) and characterization via FT-IR, EDS, and Raman measurements. Furthermore, it is well known that electrical conductivity, which is associated with the widely delocalized  $\pi$ -electrons in the sidewalls, is critical to the performance of CNTs [46, 47]. C-F bond formation is advantageous in optoelectronics because it preserves the delocalized  $\pi$ -system of fluorine-added CNTs. Figure 1a and b displays the morphologies of pure and F-CNTs, respectively. The identical tubular structure of pure and F-doped CNTs is well seen in TEM images, demonstrating that F loading did not drastically change the morphology of CNTs. Different samples of CNTs were analyzed using Raman spectroscopy in order to examine the structural defects caused by the addition of F. Figure 1c demonstrates the presence of the distinctive D-band (defects from edges, vacancies, and functional groups) and G-band (vibration mode of the  $\text{sp}^2$  hybrid C skeleton) at 1332 and 1573  $\text{cm}^{-1}$ , respectively. The relative intensities of the D and G bands ( $I_D/I_G$  values) are 0.046, 0.034, 0.021, and 0.039 for pristine CNTs, F-CNTs-0.30, F-CNTs-0.45, and F-CNTs-0.60, respectively. The value of  $I_D/I_G$  for F-CNT- $x$  is lower than that of pure CNT, implying effective modification by fluorination [40]. For the F-doped CNT- $x$ , the intensities of D-band and G'-band further decrease, which might be resulted from the improved reduction degree. FT-IR patterns of the F-CNT- $x$  (Fig. 1d) reveal an absorption band at



**Figure 1** TEM images of **a** pristine CNTs and **b** F-doped CNTs with a 0.45 M HF acid. **c** Raman pattern and **d** FT-IR spectra of CNTs doped with different concentration of HF. **e** EDS spectra of fluorine-doped CNTs with a 0.45 M HF acid.

$1221\text{ cm}^{-1}$  attributed to the stretching vibration of covalent C–F bonds, which is in good agreement with the literature [48]. After fluorinating CNTs, the band attributed to the C=C bond weakens, while the hydroxyl O–H bond strengthens. It has been

established by analysis of the FT-IR spectroscopy of F-CNT- $x$  that fluorine doping took place predominately on the CNT surface. Elemental atomic composition analysis employing EDS revealed the

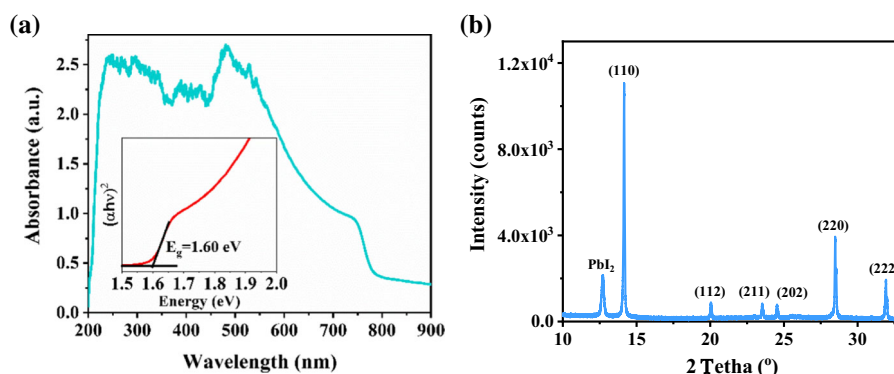
existence of carbon, oxygen, and fluorine, as demonstrated in Fig. 1e.

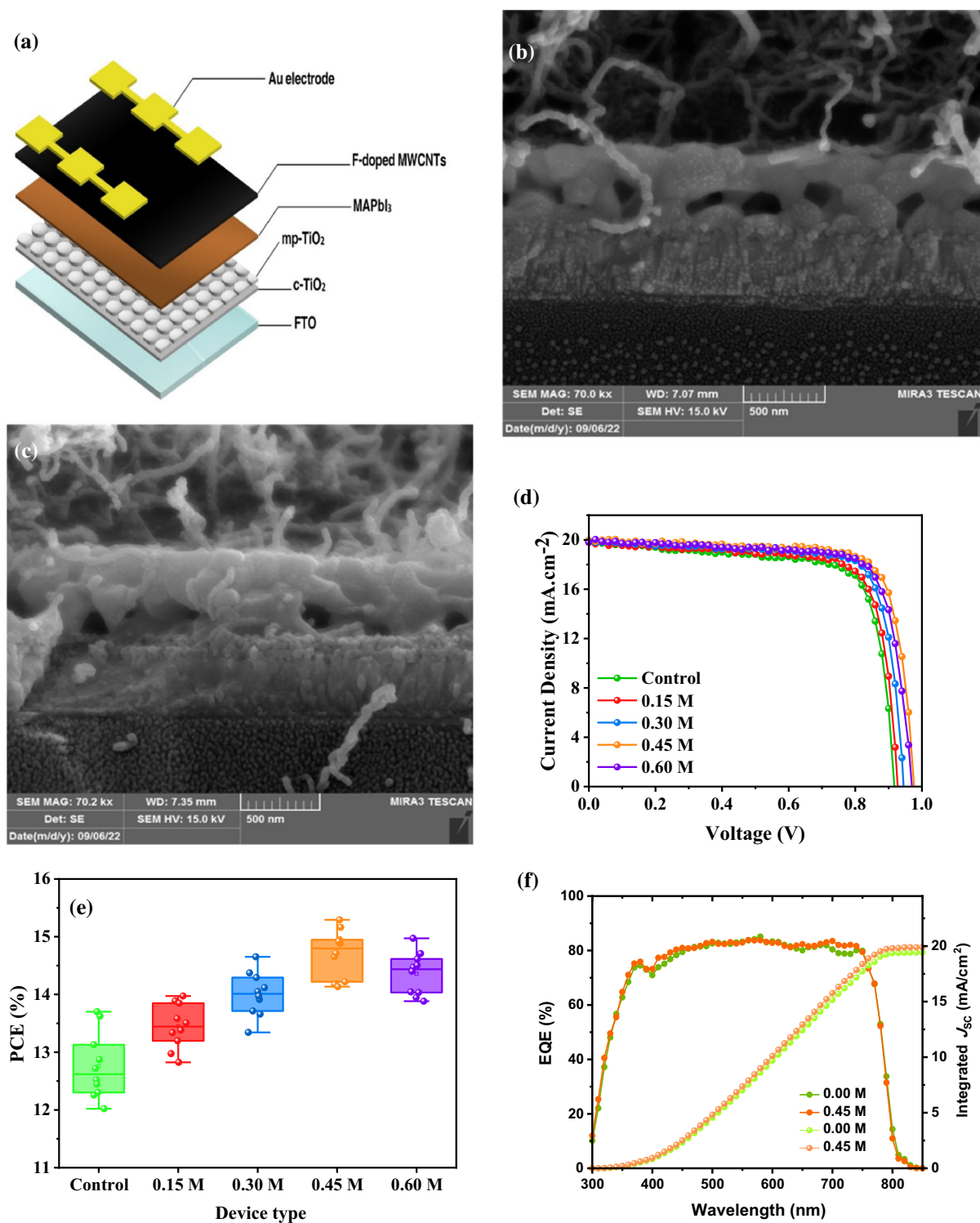
Figure 2a displays the absorption spectrum of a  $\text{CH}_3\text{NH}_3\text{PbI}_3$  perovskite coating on an FTO substrate. To be useful as a light harvester, perovskite must have an absorption property in the visible wavelength range, which it achieved. The spectrum also showed the expected absorption shoulders and absorption beginning at 747 and 782 nm, respectively, which agrees with the earlier reported data, and as calculated from Tauc curve (inset of Fig. 2a) yields an energy bandgap of 1.60 eV [49]. As shown, the bandgap of the  $\text{MAPbI}_3$  film is well matched the sunlight spectrum to efficiently absorb photons and convert them into electrical charges. The bandgap of prepared perovskite is within the optimal range for efficient absorption of visible light, making it a promising absorber for PSCs. The XRD profile of the perovskite deposited on an FTO glass is shown in Fig. 2b. Diffraction peaks at  $2\theta = 14.1^\circ$ ,  $28.4^\circ$ , and  $31.8^\circ$ , correlated to the (110), (220), and (222) planes, respectively, indicate that the material is polycrystalline and has a tetrahedral geometry, as was previously described [50, 51]. A well-oriented polycrystalline structure in the perovskite can facilitate the transport of charge carriers within the material. The partial reaction between  $\text{PbI}_2$  and MAI results in the presence of  $\text{PbI}_2$  in the perovskite, as evidenced by the peak at  $12.38^\circ$  in the perovskite pattern.

Figure 3a is a diagram showing the assembly of a CNT-based perovskite solar cell. A 450-nm-thick  $\text{MAPbI}_3$  film was produced using a one-step anti-solvent technique on a substrate made of FTO/c-TiO<sub>2</sub>/ms-TiO<sub>2</sub>. When chlorobenzene/CNT-containing HF acid was dropped over the  $\text{MAPbI}_3$  film, a partial  $\text{MAPbI}_3$ -CNT composite top layer was

formed, with a relatively thick  $\text{MAPbI}_3$  bottom layer (Fig. 3b–c). Partially embedded CNTs facilitated fast hole-electron separation and carrier transfer, which may also reduce recombination and improve carrier conduction [52]. As shown in Fig. 3b, pristine CNT-based CPSC showed balky gaps and cracks at the CNT/perovskite interface, leading to an insufficient connection between the  $\text{MAPbI}_3$  and HSM, resulting in poor carrier transportation and low CPSC performance [53]. On the other hand, with F-doped CNT-0.45 film (Fig. 3c), CNTs could bridge the  $\text{MAPbI}_3$  film, forming a seamless interface. The CNT bridging approach forms a hole-electron separation highway in the  $\text{MAPbI}_3$  film and a charge transport highway in the CNT film, giving rise to a high PCE. Figure 3d displays the  $J$ - $V$  plots of the best-performing CPSCs with pristine CNTs and F-added CNT- $x$  HSMs. The key photovoltaic parameters are described in Table 1. The champion PCE (backward scan) of the F-CNT<sub>0.45</sub>-based devices was 15.29%, with a 14.71% average PCE, exhibiting that high-performance device can be fabricated employing only CNT film as the HSM. The champion PCE of the control devices with pristine CNT film was 13.70% (backward scan). Later in the study, differences in  $V_{\text{OC}}$  and FF, as well as a slight enhancement in  $J_{\text{SC}}$ , will be examined in detail to explain why the efficiency of the F-added MWCNT devices varies from that of the pristine CNT devices. According to previous studies, a pristine CNT film has a work function of about 5 eV [54], making it close to the value reported for Spiro-OMeTAD, which is about 5.2 eV [55]. In context of this, it shouldn't be a surprise that the  $V_{\text{OC}}$  value found is close to the value found in the Spiro-OMeTAD devices [38]. Although it was already known that the CNT film transported holes, our findings show that an additional F dopant significantly boosts

**Figure 2** **a** UV–Vis absorbance of  $\text{MAPbI}_3$  perovskite layer. Inset: the Tauc curve to calculate the energy bandgap. **b** XRD profile of  $\text{MAPbI}_3$  perovskite layer deposited on FTO glass.





**Figure 3** a Device illustration of CNT-based perovskite solar cells. Cross-section FESEM micrographs of CPSCs with b undoped CNTs film and c fluorine-doped CNTs film with 0.45 M HF acid. d *J-V* characteristics of the best-performing

CPSCs with various CNTs hole-selective materials. e PCE statistical distribution of different CPSCs. f EQE spectra devices with undoped and 0.45 M HF-modified CNT layers.

**Table 1** Photovoltaic parameters of carbon-based PSCs fabricated with different CNTs hole-selective layers

HF Concentration (M)		<sup>a</sup> V <sub>OC</sub> (V)	<sup>b</sup> J <sub>SC</sub> (mA/cm <sup>2</sup> )	<sup>c</sup> FF (%)	PCE (%)
0.00	Average	0.88 ± 0.02	19.61 ± 0.38	74.09 ± 1.26	12.76 ± 0.54
	Best	0.91	19.82	75.95	13.70
0.15	Average	0.91 ± 0.02	19.64 ± 0.31	75.25 ± 0.54	13.45 ± 0.39
	Best	0.93	19.80	75.89	13.97
0.30	Average	0.93 ± 0.02	19.66 ± 0.19	76 ± 0.49	14.01 ± 0.38
	Best	0.95	19.95	77.31	14.65
0.45	Average	0.95 ± 0.02	19.71 ± 0.22	78.71 ± 0.30	14.71 ± 0.41
	Best	0.97	19.94	79.05	15.29
0.60	Average	0.94 ± 0.02	19.65 ± 0.22	77.57 ± 0.71	14.36 ± 0.37
	Best	0.97	19.82	77.87	14.97

<sup>a</sup>V<sub>OC</sub> is open-circuit voltage

<sup>b</sup>J<sub>SC</sub> is short-circuit current density

<sup>c</sup>FF is fill factor

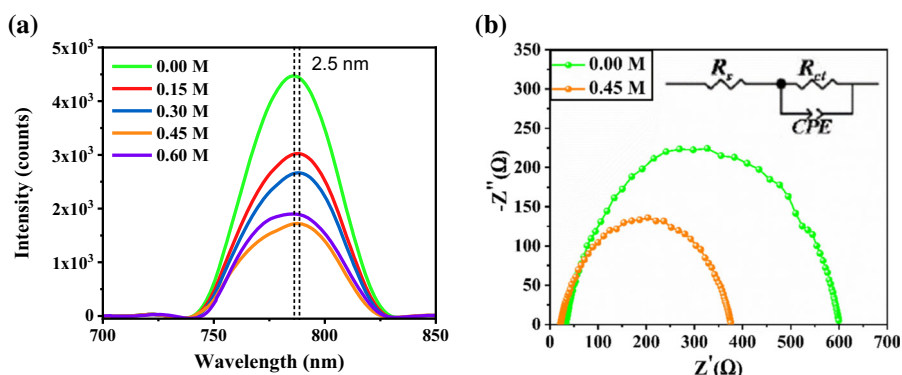
device performance. Furthermore, the fact that the F-CNT provides a higher PCE than pure CNT demonstrates that the F additive also improves the charge collection and transportation in the cells. To further investigate the reproducibility of the CPSCs, the statistics on the PCE several of the different devices based on F-doped CNT-*x* are demonstrated in Fig. 3e. Clearly, the F-CNTs' base CPSCs show small standard deviations and hence enhanced cell reproducibility. Figure 3f also displays the spectrum response of EQE of the C-PSCs treated with HF acid and those not treated. The J<sub>SC</sub> determined from the J-V profiles agrees well with the integrated J<sub>SC</sub> - estimated from the EQE: 19.42 mA cm<sup>-2</sup> for the pure-CNT cell and 91.88 mA cm<sup>-2</sup> for the HF-treated cell.

Using a steady-state PL experiment, we look at how holes are extracted from MAPbI<sub>3</sub>/HSM multi-layer interfaces. The PL patterns for PSCs formed on FTO glass with different CNT-based HSM films are shown in Fig. 4a. A PL peak appears in the MAPbI<sub>3</sub> film at a wavelength of 782 nm, which is in tune with the bandgap of the MAPbI<sub>3</sub> material. PL quenching is clearly seen at the CNT/MAPbI<sub>3</sub> interface, proving that the CNTs are capable of extracting the holes from the MAPbI<sub>3</sub> film. The MAPbI<sub>3</sub>/F-CNT-0.45 device exhibits greater PL quenching than the CNT-only device, indicating a greater hole collection capacity for the F-CNT layer from the perovskite layer. The emission spectra of F-CNT-based devices also show a slight red shift of around 2.5 nm compared to that of a pristine CNT-based device. This little blue change is indicative of CNTs' ability to passivate the surface-trap state in the MAPbI<sub>3</sub> layer [41].

The Nyquist curves (Fig. 4b) were recorded for control and optimized CPSCs under short-circuit state and full-sun irradiation conditions (*Z'* vs *-Z''*), where *Z'* and *Z''* are the real and imaginary parts of the device impedance, respectively [56]. The EIS curves were fitted with an equivalent circle as shown in inset of Fig. 4b, and the obtained parameters were listed in Table 2. As shown, in the high-frequency region, the arc of CPSC with F-CNT is smaller than that of pristine CNT-based CPSC, implying that the modified CNT film of perovskite suppresses the charge-transfer resistance (*R<sub>ct</sub>*) and confirms the role of F-CNT as a hole highway [57]. The value of *R<sub>ct</sub>* was reduced 567.4–354.2 Ω after fluorine being doped into CNT layer. Also, series resistance (*R<sub>s</sub>*) showed a notable reduction for F-modified CNT, which provides interface engineering at MAPbI<sub>3</sub>/CNT and facilitates charge transfer.

Investigations of the long-term stability of CPSCs using CNTs as the HSL that were kept in the dark and at ambient conditions with a humidity of 50% were performed. For an example comparable CPSC device, Fig. 5a shows the variations in photovoltaic performances over the long-term stability experiments. It should be noted that the CNT-based device demonstrated good long-term stability, maintaining 98% of its original efficiency after 100 days. On the other hand, HSL-free CPSC showed fast degradation only after 30 days of testing. The findings highlight that the removal of HSL is critical for the ambient stability of the CPSC device. Humidity degradation of perovskite solar cells is one of their most critical obstacles. Therefore, improving the hydrophobic

**Figure 4** **a** Steady-state PL spectra of carbon-based perovskite solar cells. **b** EIS measurements for control (without F doping) and optimized (with 0.45 F content) devices.



**Table 2** The fitted EIS parameters of different carbon-based PSCs

Device name	$R_s$ (ohm)	$R_{ct}$ (ohm)	$n$	$P$ ( $\times 10^{-6}$ )
0.00 M	33.1	567.4	0.84	3.2
0.045 M	21.7	354.2	0.81	3.5

behavior of PSCs can increase their resistance against humidity degradation. One technique that can demonstrate the hydrophobic behavior of PSCs is measuring the contact angle of the water droplet, and researchers frequently use this technique [58–60]. Figure 5b and c shows that CNT layers possess high water contact angles. The hydrophobic property of CNTs can efficiently hinder moisture from permeating into the MAPbI<sub>3</sub> layer. So, the high stability of the CPSC-based device in the environment should be mostly credited to the hydrophobic CNT.

Fluorine doping of CNT increases the potential of this material for employment as a hole transport layer. As summarized in Table 3, developing F-doped

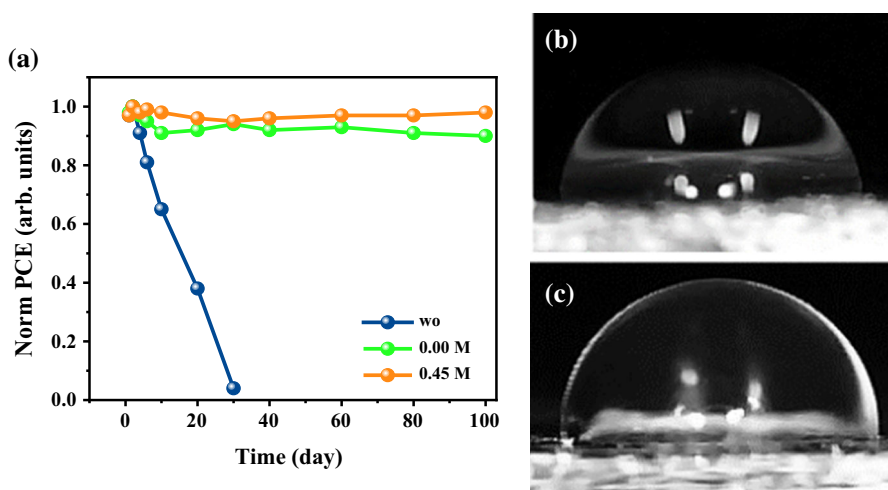
CNT increases the charge-transfer process at the perovskite/CNT interface by reducing the  $R_{tr}$  of the PV device, photovoltaic parameters, hydrophobic behavior of CPSCs, and humidity stability of CPSCs.

Table 4 presents an overview of current and pertinent studies related to the utilization of SWCNT or MWCNT in perovskite-based photovoltaic devices. The multifunctional capabilities of carbon nanotubes in solar cells are noteworthy, as they can enhance charge transport, reduce recombination rates, and offer protection for perovskite materials, ultimately resulting in increased stability and efficiency. CNTs possess the potential to serve as additives in HSM for planar and mesoporous PSC architectures.

## Conclusion

In summary, we report a CNTs-bridging approach for the preparation of carbon-based MAPbI<sub>3</sub> PSCs with considerable efficiency by a cost-effective method. The CNT bridged between the MAPbI<sub>3</sub> film

**Figure 5** **a** Stability test of different CPSCs at environment with relative humidity of 50% in dark conditions. Contact angle of water droplet on **b** CNT-0.00 and **c** CNT-0.45 layers.



**Table 3** Effects of fluorine doping of CNT on photovoltaic and stability of CPSCs

CNT type	Charge transfer	$R_{tr}$	$V_{OC}$	$J_{SC}$	FF	PCE	Hydrophobicity	Stability
Pure CNT	L	H	L	L	L	L	L	L
F-doped CNT	H	L	H	H	H	H	H	H

L and H refer to lower and higher, respectively

**Table 4** Comparison between our findings and previously reported studies that utilized CNT as HSMs

Authors	Structure	Study type	$V_{OC}$ (V)	$J_{SC}$ (mA/ cm <sup>2</sup> )	FF (%)	PCE (%)	Stability
AbdulMohsin et al. [61]	ITO/ZnO/MAPbI <sub>3</sub> /PANI•SWCNT/Au	Simulation	0.56	11.00	30.00	5.00	Not reported
Wei et al.[30]	FTO/TiO <sub>2</sub> /MAPbI <sub>3</sub> /MWCNT	Experimental	0.88	15.60	80.00	12.67	90% stable after 10 days under 20% RH
Wang et al. [25]	FTO/c-TiO <sub>2</sub> /ms-TiO <sub>2</sub> /MAPbI <sub>3</sub> /SWCNT/GO/PMMA/Au	Experimental	0.95	19.40	72.00	13.30	95% stable after 10 days under 70–80% RH
Ryu et al.[62]	FTO/c-TiO <sub>2</sub> /ms-TiO <sub>2</sub> /Cs <sub>0.06</sub> (MA <sub>0.17</sub> FA <sub>0.83</sub> )Pb(I <sub>0.84</sub> Br <sub>0.16</sub> ) <sub>3</sub> /CNT/C	Experimental	1.00	18.97	71.00	13.57	90% stable after 240 min under 80% RH
Zheng et al. [40]	FTO/c-TiO <sub>2</sub> /ms-TiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> /MAPbI <sub>3</sub> /B-MWCNT	Experimental	0.92	21.50	77.00	15.23	98% stable after 80 days in dry air
Wang et al.[52]	FTO/c-TiO <sub>2</sub> /ms-TiO <sub>2</sub> /MAPbI <sub>3</sub> •SWCNT/SWCNT•C	Experimental	0.97	22.36	72.22	15.73	100% stable after 90 days under 65 ± 5% RH
Salehi-Abar et al. [19]	FTO/c-TiO <sub>2</sub> /ms-TiO <sub>2</sub> /MAPbI <sub>3</sub> /MWCNT/Cr <sub>2</sub> O <sub>3</sub> /Au	Experimental	1.00	20.80	78.10	16.29	99% stable after 50 days under 40% RH
Mohammed et al. [63]	FTO/ TiO <sub>2</sub> /MAPbI <sub>3</sub> /CNT/Au	Simulation	1.10	19.19	86.62	18.30	Not reported
Kenfack et al. [64]	ITO/Cu <sub>2</sub> O/ MAPbI <sub>3</sub> /SWCNT•[6]CPP/Au	Simulation	1.00	34.65	72.80	25.31	Not reported
This work	FTO/c-TiO <sub>2</sub> /mp-TiO <sub>2</sub> /MAPbI <sub>3</sub> /F-CNTs/Au	Experimental	0.97	19.94	79.05	15.29	98% stable after 100 days under 50% RH

C, carbon; MWCNT, multi-walled CNT; c-TiO<sub>2</sub>, compact TiO<sub>2</sub>; ms-TiO<sub>2</sub>, mesoporous TiO<sub>2</sub>; SWCNT, single-walled CNT; PANI, polyaniline; P3HT, poly(3-hexylthiophene); Cr<sub>2</sub>O<sub>3</sub>, chromium(III) oxide; GO, graphene oxide; PMMA, polymethyl methacrylate; B, boron

and back electrodes, leading to improved hole extraction and transport capability and developing an intimate CNT/MAPbI<sub>3</sub> interface. Due to such characteristics, an efficiency of 15.29% with an FF of 79.05% was achieved. Moreover, CPSCs based on the CNT layer showed remarkable long-term stability under ambient conditions (50% RH, 25 °C) for about 100 days. The hydrophobic and dense CNT coating protects the MAPbI<sub>3</sub> perovskite material from ambient humidity. The current study is a significant step forward in the development of CPSCs, as it has improved their efficiency and stability. This progress could pave the way for the practical use and industrialization of carbon-based PSCs in the future.

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## Data availability

Data will be available based on reasonable request.

## Declarations

**Conflict of interest** There is no conflict of interest by any author.

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