

An Advanced Study of Carbon Nanotube Applications in Perovskite Solar Cells

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DOI: <https://doi.org/10.36348/sjet.2026.v11i04.007>

Received: 08.10.2025 | Accepted: 16.12.2025 | Published: 11.04.2026

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Abstract

Halide perovskites have emerged as significant materials for the light-absorbing layer in many optoelectronic devices due to their remarkable optoelectronic capabilities. To enhance device performance for broader acceptance, it is imperative to identify novel solutions. A viable approach is the integration of carbon nanotubes (CNTs), which have demonstrated exceptional adaptability and effectiveness. In these devices, carbon nanotubes (CNTs) fulfill many roles, such as supplying conductive substrates and electrodes, as well as enhancing charge extraction and transport. The forthcoming generation of photovoltaic devices, metal halide perovskite solar cells (PSCs), presents significant potential. Despite substantial advancements, concurrently attaining optimal efficiency, stability, and affordability continues to pose a challenge, necessitating the creation of innovative materials termed CNTs, which, due to their exceptional electrical, optical, and mechanical characteristics, have attracted significant interest as prospective materials for highly efficient PSCs. The integration of CNTs into perovskite solar cells enhances adaptability, facilitating advancements in device performance and durability for many applications. This article offers a comprehensive examination of current developments in carbon nanotube technology and its incorporation into perovskite solar cells, functioning as transparent conductive electrodes, charge transporters, interlayers, hole-transporting materials, and back electrodes. Furthermore, we identified significant obstacles and provided recommendations for future improvements in perovskite solar cells with CNTs.

Keywords: Perovskite, CNTs, Optical, Efficiency, Electrodes, Transparent.

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1: INTRODUCTION

The utilization of renewable and sustainable energy sources is increasing in prevalence. Increasing apprehensions over climate change and the exhaustion of fossil fuel supplies are the principal catalysts in this context [2,3,4,5]. Solar energy is a cost-effective and feasible solution to fulfill future energy requirements without compromising sustainability. The optoelectronic potential of metal halide perovskites is swiftly advancing [7,9]. Their exceptional optoelectronic characteristics, such as a fluctuating bandgap, substantial absorption coefficient, and elevated carrier mobility, predominantly account for this. Researchers have demonstrated a prolonged and balanced carrier diffusion duration with little nonradiative loss [10]. Various optoelectronic devices, including photodetectors, light-emitting diodes, and solar cells, derive significant advantages from their distinctive characteristics [16-17]. Metal halide perovskite solar cells are emerging as a highly promising

alternative to conventional silicon-based photovoltaic technology. This is due to the ability to conduct research utilizing economical, scalable, low-temperature, solution-processing techniques [13,14].

A p-i-n or n-i-p configuration of a single-junction perovskite solar cell generally has the following components: a back electrode, a perovskite light-absorbing layer, an electron transport layer (ETL), and a transparent conductive metal oxide electrode. A commonly utilized substrate for subsequent layer deposition is a transparent conductive metal oxide that envelops a stiff plastic or glass base. The perovskite layer is interposed between two electron transport layers (ETLs) and the hole transport layer (HTL), regardless of whether solution-based or thermal evaporation methods are employed. The metal back electrode of the perovskite solar cell is vaporized to complete the assembly. Researchers have consistently modified the perovskite composition and crystal growth, enhanced device

interfaces, and developed novel device topologies to optimize the performance of their devices [5,7].

In a brief period, the worldwide scientific community has significantly improved the photovoltaic conversion efficiency of perovskite solar cells, increasing it from 3.8% to 30%. The crystalline silicon solar cell maintains commendable commercial performance despite its restricted stability and insufficient scalability. The predominant perovskite solar cells (PSCs) exhibiting excellent efficiency reported in the literature are constructed on a surface area of 0.01 cm². Research indicates that the efficiency of perovskite solar cells declines with increasing area [17,18,19]. The efficiency of perovskite/silicon heterojunction tandem solar cells has attained 28.6% [20], while the efficiency of perovskite/perovskite tandem solar cells has surpassed 32% [21], but the theoretical maximum efficiency reported above 35% [22].

Metal halide perovskites rapidly dissolve in many polar solvents owing to their robust ionic bonding and low formation enthalpy. This characteristic enables the successful and economical fabrication of research samples at low temperatures for solution processing applications. Due to their exceptional fault tolerance, films produced by solution processing preserve the distinctive optoelectronic characteristics of perovskites. Consequently, they cannot undergo processing akin to conventional semiconductors, which need a high-temperature vacuum [23].

Spray deposition, ink-jet printing, gravure printing, blade coating, and slot-die coating are scalable manufacturing processes employed to make large-area perovskite solar cells. A minimodule of 21 cm² achieved a power conversion efficiency above 18% using thermally co-evaporated deposition [26]. Furthermore, semitransparent perovskite solar cells may be easily fabricated utilizing perovskites, owing to their elevated absorption coefficient and capacity for bandgap modulation. Windows and facades of buildings and vehicles that produce power might represent a viable use for these PSCs [27,28]. In addition to ongoing efforts to achieve opaque and semitransparent perovskite solar cells with high-performance single junctions, there is a growing focus on developing tandem solar cells utilizing perovskites. Researchers aim to substantially improve the cost-efficiency ratio of solar technology while concurrently minimizing spectral losses with these cells [29,30].

The power conversion efficiencies of perovskite–silicon tandem solar cells currently reach 28% under standard sunlight conditions, whereas perovskite–perovskite solar cells have an efficiency of 24.8% [31]. Advancements have occurred in device designs and power conversion efficiency; nonetheless, much progress is necessary for perovskite solar cells to

achieve their full potential. These cells must exhibit enhanced resistance to deterioration, reduced cost, and be free of lead [32]. The significance of resolving the stability issue in PSCs has been well acknowledged [33,34]. Their stability may be influenced by environmental conditions like UV radiation, temperature stress, and moisture infiltration. Metal electrodes and selective charge transport layers are two internal variables that significantly influence the device's long-term performance. Recent years have witnessed significant enhancements in device stability [35,36].

Numerous factors influence the energy payback period of PSCs, including stability, material costs, and manufacturing expenses. Currently, one of the priciest elements of PSCs is transparent conductive oxide electrodes, often made of indium tin oxide or fluorine tin oxide [37]. Researchers concur that their lack mechanical flexibility renders them unsuitable for application in flexible PSCs. Moreover, the utilization of metal back electrodes, often made of silver (Ag) or gold (Au), can be a costly and energy-demanding procedure necessitating high-vacuum evaporation techniques. Moreover, during device operation, unwanted electron and hole recombination often transpires inside the perovskite layers and at their interfaces, constraining the improvement of device performance [38]. Utilizing innovative materials and enhancing the designs of current components are essential for addressing these challenges.

Numerous carbon materials have been utilized to address the previously mentioned issues with PSCs [39]. Researchers focusing on PSCs find carbon nanotubes (CNTs), a type of one-dimensional carbon nanomaterial, particularly intriguing. Since the early 1990s, carbon nanotubes have been utilized in numerous applications across electrical and optoelectronic devices [40,41]. The structural characterization of carbon nanotubes indicates that they are fundamentally hollow cylinders formed by the rolling of graphene sheets in one or multiple layers. Carbon nanotubes can be classified into three primary categories: single-walled, double-walled, and multi-walled nanotubes [42]. SWCNTs can be categorized into three types based on their chiral angles (θ): zigzag ($\theta = 0^\circ$), chiral ($0^\circ < \theta < 30^\circ$), and armchair ($\theta = 30^\circ$) [43]. The diameters and chiral angles of SWCNTs determine their classification as metallic or semiconducting. The electrical conductivity of metallic single-walled carbon nanotubes typically ranges from 10^4 to 10^6 S m⁻¹. At room temperature, their thermal conductivity can reach up to 3500 W m⁻¹ K⁻¹ [44].

Carbon nanotubes exhibit a tensile strength approximately 100 times greater than that of steel, alongside an exceptionally high Young's modulus, which ranges from 1 to 4 TPa. These properties arise from the presence of covalent sp² bonds between carbon atoms [45]. A number of articles have briefly described the key properties of carbon nanotubes. The utilization

of carbon nanotubes in perovskite solar cells is attributed to their notable optical and electrical properties, as well as their exceptional mechanical flexibility. Included are PSCs featuring a mesoporous n-i-p structure, a planar n-i-p structure, an inverted p-i-n structure, or a mesoscopic structure lacking a hole transport layer. Recent advancements have demonstrated significant success in employing carbon nanotubes in various components of devices to enhance the performance of perovskite solar cells. The components of perovskite light absorbers consist of additives, interlayers, transparent conductive electrodes, back electrodes, and materials for electron and hole transport [46,47].

Power conversion efficiency exceeding 25% was attained through the full integration of carbon nanotubes into perovskite solar cells (PSCs). In light of these promising results, we are reassessing the function of carbon nanotubes in perovskite solar cells. Recent papers have reported encouraging reviews of carbon materials utilized in perovskite solar cells (PSCs); however, comprehensive research specifically addressing carbon nanotubes (CNTs) in PSCs remains lacking. This review summarizes and critically evaluates recent developments in the incorporation of carbon nanotubes into perovskite solar cells. This paper aims to summarize recent advancements in carbon nanotube (CNT)-based perovskite solar cells (PSCs). In conclusion, we identify the primary challenges and offer insights on potential advancements for perovskite solar cells utilizing carbon nanotubes in the future [48].

2: Structure of Carbon Nanotubes

Carbon nanotubes are an artificially fabricated allotrope of carbon. Graphene in two dimensions can be regarded as a precursor material for fullerenes, carbon nanotubes, and three-dimensional graphite [49]. The defining features of carbon nanotubes are their high area ratio and hollow cylindrical structure. The atoms constituting the tube's walls exhibit sp² and sp³ hybridization, resulting in a mixed hybridization state. Four atoms are associated with the sp³ hybrid orbital, exhibiting a bond angle of 109.5°, while three atoms are linked to the sp² hybrid orbital, characterized by a bond angle of 120°. Linear defects characterized by sp³ hybrid orbitals exist within the sp² hybrid lattice. The structure is preserved within a carbon nanotube environment. A

carbon nanotube surface with five-membered rings exhibits a convex form, whereas a surface containing seven-membered rings displays a concave shape. This results in an uneven and flattened structure of the carbon nanotube. The capped end of a carbon nanotube is formed when the tube's terminus is decorated with a heptacyclic or pentacyclic ring. Members with seven or five rings frequently encapsulate the cylinder within large fullerene and hemispherical molecules [50].

Carbon nanotubes are classified as single-walled or multi-walled based on the number of wall layers present. In comparison to single-walled carbon nanotubes (SWCNTs), multi-walled carbon nanotubes (MWCNTs) exhibit greater chemical inertness. Additionally, multi-walled carbon nanotubes exhibit cleaner surfaces. The radii of nanotubes exhibit an inverse relationship with shear and Young's moduli [51]. Defect centers, characterized by configurations of five or seven members, typically form at the ends of the tubes, on the surfaces of the layers, and between the layers of single-walled carbon nanotubes during expansion. The intensity of chemical reactions induced by defects escalates, and the complexity of the surface's chemical composition increases with the number of carbon nanotube layers [52]. Multi-walled carbon nanotubes, the predominant type of carbon nanotubes, consist of multiple coaxial cylindrical tubes containing several layers, which can range from a few to tens of layers. Moreover, additional scholarly articles have elucidated diverse carbon nanotube structures and their applications.

The chiral vector, denoted as Ch , serves as a standard method for characterizing the atomic structure of single-walled carbon nanotubes. The chiral vector, denoted as Ch , is defined by the equation $\text{Ch} = n\mathbf{a}_1 + m\mathbf{a}_2$, where n and m represent the chiral indices. The large arrows indicate armchair, chiral vectors, and zigzag formations. SWCNTs can be categorized into three distinct types based on their axial orientations (n, m): zigzag form ($m = 0$ or $n = 0$), armchair form ($n = m$), and chiral form ($n \neq m$). Carbon nanotubes exist in three distinct forms, as illustrated in the image [53]. Chairs can exhibit various structures, including armchair, zigzag, chiral, double-walled, and multi-walled configurations (Figure 1) [54, 55].

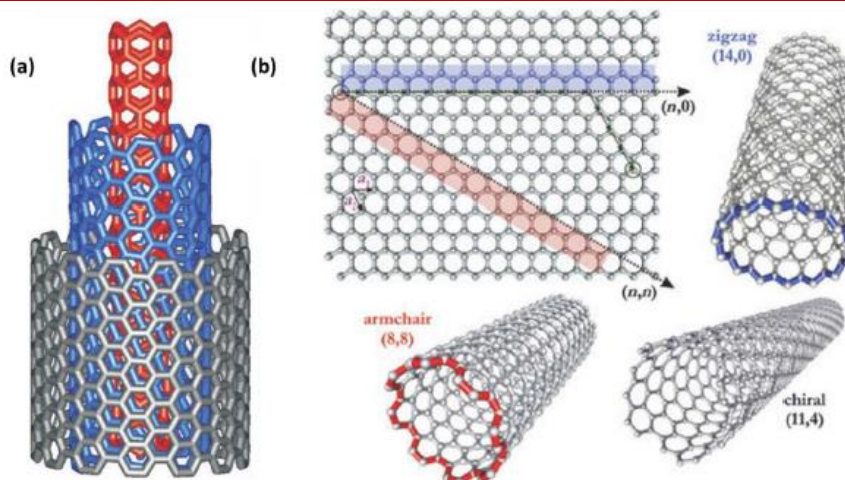


Figure 1: (a) Structure of a multi-walled carbon nanotube featuring three shells with differing chirality. A graphene sheet is rolled, producing three distinct forms of carbon nanotubes (CNTs). Copyright © 2004 John Wiley & Sons

3: CNTs Synthesis

A: Laser ablation

A laser can also be employed to synthesize nanotubes. The production of nanotubes utilizes a laser, as indicated by the terminology. The laser ablation approach represents a more advanced technology that addresses the limitations of the electric arc method. This is pertinent to the synthesis of both multi-walled carbon nanotubes (MWCNTs) and single-walled carbon nanotubes (SWCNTs). Nanotubes are frequently produced in large quantities utilizing this method. The synthesis process entails the ablation of the carbon target through the application of a pulsed laser. Pulsed laser ablation is performed on a graphite target using a catalyst and an inert gas. The process results in the synthesis of carbon nanotubes. Thess *et al.*, synthesized ropes of SWCNTs through the laser ablation method [21].

In these experiments, nanotubes measuring hundreds to several microns in length and 5 to 20 nm in size were synthesized. Researchers indicate that carbon nanotube structures can be validated through transmission electron microscopy and X-ray diffraction techniques. Two proposed methods for the fabrication of carbon nanotubes are direct ablation and carbon particle suspension in the reaction zone [35]. The synthesis of SWCNTs is adversely impacted by the use of a high-intensity UV laser. Multiple factors influence the synthesis via laser ablation, including the catalyst, laser power, temperature, wavelength, selection of inert gas, fluid dynamics, and pressure surrounding the carbon target. It is essential to maintain control over each variable during the synthesis of nanotubes. The laser ablation method for carbon nanotube production exhibits several deficiencies. One disadvantage is the elevated final temperature of the synthesized molecules [21-35].

Faizah A. AlMalki and colleagues employed a Q-switched pulsed Nd laser. A graphite target submerged in deionized water was subjected to ablation using a YAG laser with a wavelength of 1064 nm. The laser exhibited a pulse repetition rate of 1 Hz and a pulse

duration of 9 ns. A graphite pellet with a purity of 99.91% (sourced from National Spectroscopic Electrodes Co.) and a diameter of mm was positioned at the bottom of a glass jar containing 3 mL of deionized water (D-W). The laser energy ranged from 60 to 220 mJ, delivered in 75 pulses [59,60]. The synthesis of pristine single-walled carbon nanotubes (SWCNTs) is highly desired because of their significant applications in electronics and biomedicine. The synthesis of this specific tube via the AD approach involved immersing the anode in various metal catalysts that are not required for the formation of MWCNTs (Figure 2a) [61].

B: Hydrothermal/Sonochemical Method

Using a sonochemical technique, Jeong *et al.*, produced SWCNTs. The catalyst, ferrocene, was combined with the carbon source, p-xylene, and the nucleation site, silica powder, to collect CNTs following ultrasonication at room temperature [62]. The sonochemical approach was used by Raja and Ryu to synthesize CNTs. Sonicating dichlorobenzene and ZnCl₂ particles in an ultrasonic water bath was a straightforward process. The system's maximum output was 50 W at 40 KHz [63]. Sodium hydroxide, distilled water, polyethylene glycol, and ethyl alcohol were among the precursor materials used in the hydrothermal synthesis of CNTs by Wang *et al.*, In a 250 mL flask, these ingredients were coupled. Using a magnetic stirrer, the reaction mixtures were vigorously mixed for 30 min. Once the first half hour was over, the precursor was transferred to a reactor to undergo further treatment for 20 h at 160 °C. The researchers removed the material when it cooled [64]. The hydrothermal approach was used by Razali *et al.*, to synthesize CNTs. The carbon sources, catalyst, and solvent utilized were ferrocene (2 g), sulfur (4 g), and a NaOH solution (10 MW). For 24 h, the precursor was heated to 200 °C in an autoclave reactor made of Teflon–stainless steel. The samples were collected for further analysis after chilling [65]. Carbon nanotubes (CNTs) are a recent development in nanotechnology and have become a cornerstone of scientific study owing to their extensive use in the fields

of spintronics, optical materials, and optoelectronics. Currently, carbon nanotubes (CNTs) are extensively produced using a variety of well-established techniques that include elevated temperatures. Carbon nanotubes (CNTs) were produced utilizing a hydrothermal approach at a low temperature of 160 °C. The precursors used for this synthesis were ethanol, polyethylene glycol (PEG), and sodium hydroxide. The reaction time for the synthesis was 20 h. The structure of the prepared CNTs was evaluated using a SEM analysis (Figure 2b) [66].

C: Arc Discharge Method

Arc discharge refers to the phenomenon where an electric current passes through the air or another medium, resulting in a continuous discharge of electricity. This process is characterized by the formation of a conductive plasma channel, which allows for the flow of current between two electrodes. Arc discharges are commonly observed in various applications, including welding, lighting, and electrical equipment failures.

The production of nanotubes originates from the arc discharge process, one of the earliest known technologies. Arc evaporation is considered the most effective method for synthesizing multi-walled carbon

nanotubes [67]. The generation of an electric arc requires two electrodes and the application of direct current between them during the arc discharge process [68]. The procedure initiates with the positioning of electrodes inside a vacuum chamber, which is subsequently filled with inert gas. Carbon deposition on the electrodes may occur rapidly as a result of the continuous gas flow. The presence of plasma within the chamber facilitates the deposition process. Two essential factors are (1) the pressure of the inert gas and (2) the current, which must be precisely regulated for the process to function effectively. During the activation of the power source, the inert gas pressure in the reaction chamber remains constant. An electric arc is generated when the positive and negative electrodes approach, collide, and subsequently separate. The electrodes generate plasma when subjected to high temperatures through arcing. Subsequently, carbon nanotubes adhere to the negative electrodes. This approach enables the production of carbon nanotubes with single or multiple walls, as well as those with multiple layers. Unlike MWCNTs, which are synthesized by merely regulating the pressure of an inert gas, the production of SWCNTs requires the use of a catalyst. This method enables the growth of nanotubes measuring several microns in length and 2–20 nm in diameter [69].

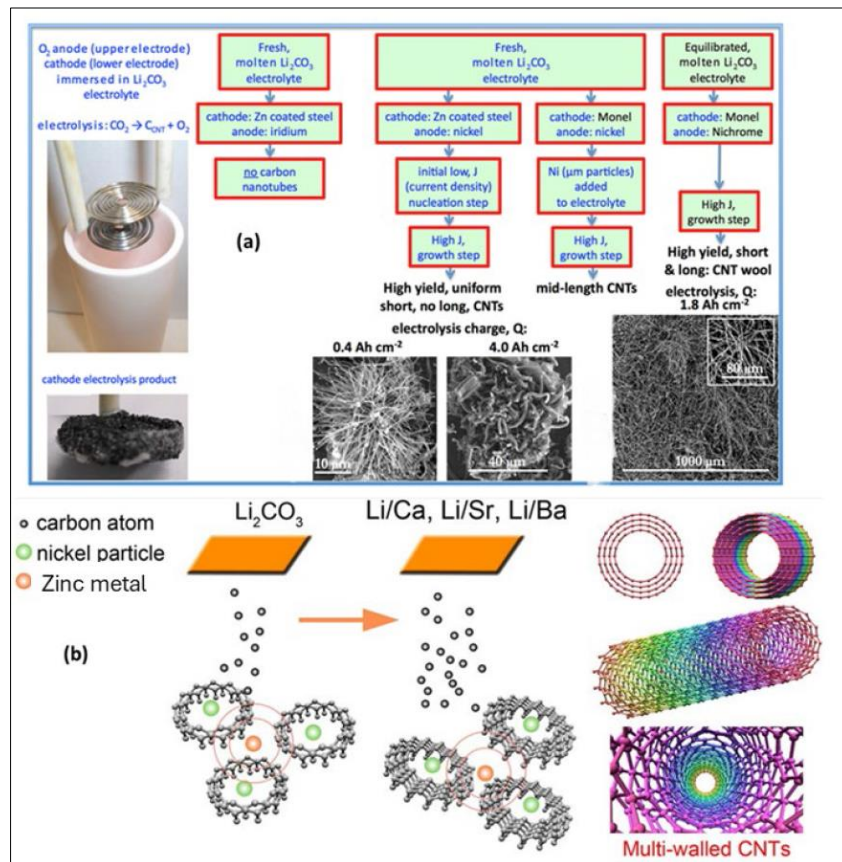


Figure 2: (a) Schematic representation of innovative synergistic pathways for the electrolysis of macroscopic-length CNT “wool” in molten carbonate, resulting in a high yield. Reproduced with authorization from [71]. Copyright © 2017 Elsevier. Methods for synthesizing carbon nanotubes in solutions containing either pure lithium carbonate or salts, including alkaline earth carbonates. Reproduced with authorization from [72]. Copyright © 2019 IOP Publishing

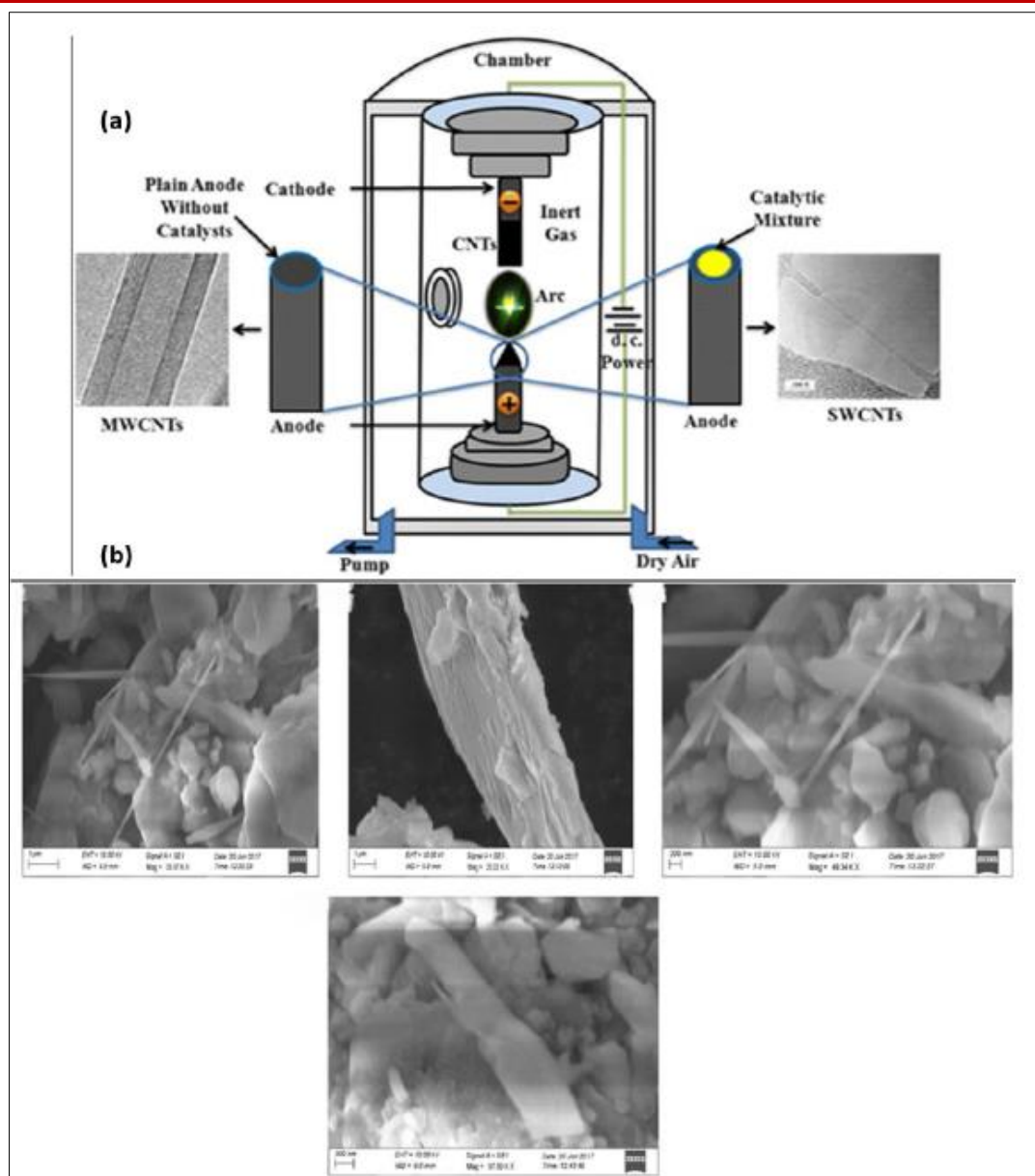


Figure 3: (a) Schematic representation of the apparatus for synthesizing single-walled carbon nanotubes (SWCNTs) and multi-walled carbon nanotubes (MWCNTs). SEM images of carbon nanotubes. Copyright © 2016 Springer Nature

D: Electrolysis-Technique

Researchers synthesized carbon nanotubes through electrochemical deposition. An electrochemical reactor was constructed using a tri-chamber electrochemical cell. Nanoparticles coated with a Fe/Ni alloy were utilized to construct the anode and cathode, while the calomel electrode functioned as the standard reference. The electrode's volume consisted of a mixture comprising 40% methanol and 60% benzyl alcohol. The cathode facilitated the production of carbon nanotubes through the application of a potential difference of approximately 1000 volts between the anode and cathode [70]. Johnson and colleagues synthesized carbon nanotubes through molten electrolysis, utilizing CO₂ as the precursor material (Figure 3a). Molten lithium carbonate was utilized for synthesis purposes. A crucible

made of pure alumina was utilized to conduct the reaction. A range of metals functioned as the cathode, with nickel acting as the anode. The cathode produced carbon nanotubes, while the anode generated oxygen as a result of electrolysis. The synthesis strategy involved two steps: dissolution and electrolysis. Lithium carbonate was produced during the dissolution process. Subsequently, carbon nanotubes were synthesized from lithium carbonate [71]. Li *et al.*, synthesized carbon nanotubes using CO₂-based molten salt electrolysis. This method utilized Li₂CO₃, CaCO₃, SrCO₃, and BaCO₃ for the synthesis of CNTs (Figure 3b). The synthesis method entailed the introduction of various combinations of these substances into a crucible. Nickel wire functioned as the anode, while galvanized iron wire acted as the cathode. The crucible was heated to a

temperature of 750 °C. The deposition of CNTs was accomplished by incrementally increasing the current density from 6 mA/cm² to 200 mA/cm² [12].

E: Chemical-vapor-deposition

The chemical vapor deposition method employs an appropriate catalyst and carbon source [53-54]. This method is highly effective for the synthesis of carbon nanotubes. Chemical vapor deposition yields carbon nanotubes characterized by low crystallinity, leading to the formation of disordered and misaligned graphene sheaths. Transmission electron microscopy (TEM) was employed to identify this. Inductively coupled plasma vapor deposition represents an alternative method that may be employed. The ICP system generates plasma that is responsible for the production of nanotubes. This method allows the CNTs to exhibit crystallinity at temperatures reaching 500 °C. There are two issues associated with the application of chemical vapor deposition for the synthesis of carbon nanotubes. One concern is the overconsumption of precursors during this process [51-60]. This method employs significantly higher temperatures compared to PECVD. Nanotubes are synthesized exclusively at low temperatures through plasma-assisted chemical vapor deposition (CVD). The formation of microstructure is significantly dependent on low-temperature synthesis. Furthermore, nanotubes produced via PECVD exhibit significantly reduced contamination levels compared to those synthesized

through CVD. Figure 4a–e presents a schematic representation of the carbon nanotube (CNT) production process via fluidized bed chemical vapor deposition (FCCVD). Ferrocene serves as the catalyst precursor, while sulfur acts as the growth promoter, and ethylene functions as the carbon source, all of which are continuously introduced into a reactor alongside a carrier gas. The metallocene catalyst precursor decomposes at a higher temperature, leading to the formation of catalyst nanoparticles through the collision of metal clusters. The decomposition of carbon precursors occurs at high temperatures, with metal nanoparticles serving as catalysts. Simultaneously, specific regions of the catalyst nanoparticles undergo the development of hot zones on their surface, potentially due to the segregation of sulfur. Carbon that is completely absorbed in particular regions of the molten material solidifies to create carbon caps, which subsequently extend into elongated tubes as carbon continues to be supplied. The FCCVD process is characterized by the synthesis of catalyst nanoparticles and the continuous growth of CNTs through the ongoing provision of the catalyst precursor and carbon source. The FCCVD technique allows for precise adjustments of growth parameters, including the catalyst precursor and its introduction method, the carbon source, growth temperature, and the type and flow rate of the carrier gas. This degree of control facilitates the manipulation of morphology, structure, quality, and yield of carbon nanotubes (CNTs) [75].

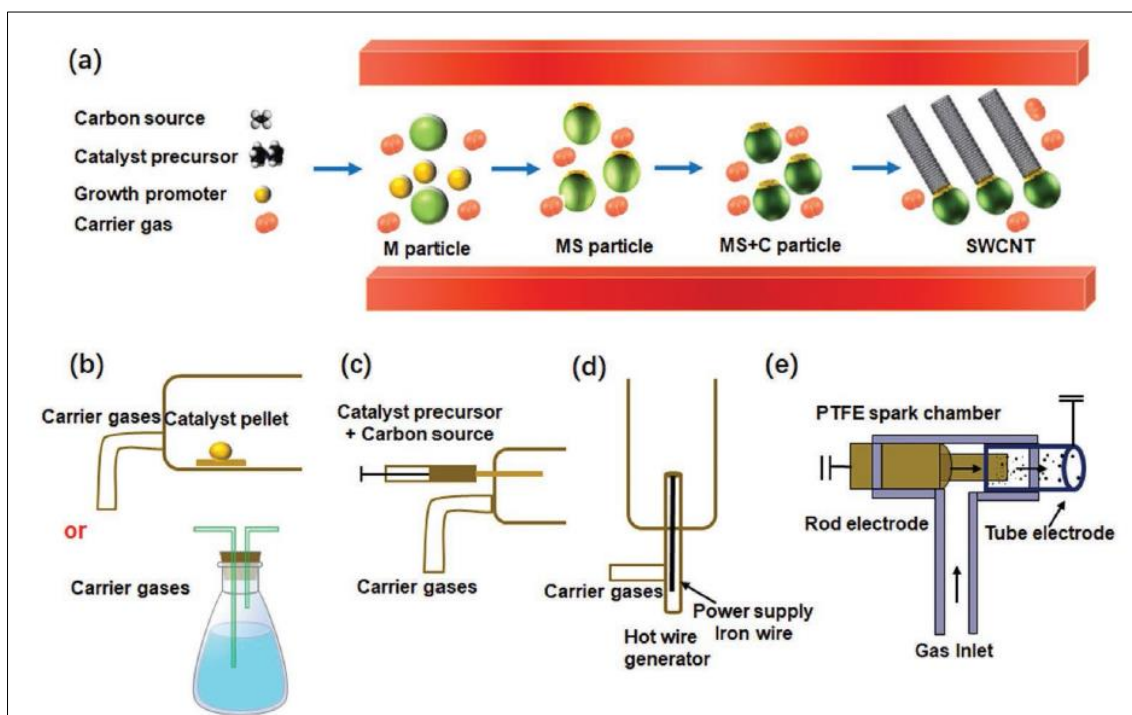


Figure 4: (a) Schematic representation of carbon nanotube synthesis via fluidized catalytic chemical vapor deposition (FCCVD), and (b–e) different methods for catalyst introduction into the reactor: supersaturated vapor. Reproduced with authorization from [75]. Copyright © 2019 John Wiley & Sons

4: Perovskite-Solar-Cell

The perovskite film serves as the light-absorbing layer in inverted PSCs. A compact and

homogeneous perovskite coating is essential to prevent undesired contact between the upper and lower charge transport layers [59]. High-quality films are crucial for

minimizing nonradiative recombination at the interfaces caused by defects and energy levels. The VOC and FF values of PSCs are significantly constrained due to these factors. Enhancements in the quality of perovskite films have emerged from rigorous optimization of manufacturing processes, precise adjustments of the band gap, and effective flaw passivation. The Schottky–Queisser limit indicates that photovoltaic solar cells incorporating a perovskite material with a suitable band gap may attain power conversion efficiencies exceeding 30% [61]. Altering the chemical composition of the perovskite may achieve this outcome. A transition from perovskites utilizing methylammonium lead iodide to those employing formamidinium has been adopted in high-efficiency inverted perovskite solar cells. There are several known types of perovskites: mixed-cation perovskites that include FA and Cs, triple-cation perovskites that incorporate cesium and FAMA, and mixed-cation perovskites that consist of FAMA and Cs. Various perovskite types require distinct production methods for the fabrication of perovskite films. The current gold standard for inverted perovskite solar cells is a one-step procedure that consistently yields high-quality perovskite films with minimal waste [61–64]. The significant influence of manufacturing conditions on the full conversion of PbI₂ into perovskite renders the conventional two-step solution method, typically employed for standard PSCs, inapplicable to inverted perovskite films. The produced perovskite films frequently exhibit a rough surface, complicating the uniform application of a thin PCBM or C60 layer over the entire perovskite layer [64,65]. As a result, a significant quantity of charge carriers is lost due to inadequate coverage. Vacuum deposition, blade coating, and slot-die coating represent scalable techniques commonly employed in the production of large-scale perovskite solar modules, in contrast to the methods utilized for small-scale cell fabrication in laboratory settings [66–68].

5: Carbon-Nanotube-Based-Perovskite-Solar-Cells

Carbon nanotubes and various one-dimensional semiconducting materials with distinctive architectures have been extensively utilized in perovskite solar cells. Cylindrical carbon nanotubes represent a specific category of carbon nanomaterials. Their length, diameter, and chirality represent several dimensions along which these nanotubes can exhibit variation in shape and size. Structures may be classified as single-walled, double-walled, or multi-walled. Carbon nanotubes exhibit significant potential in photovoltaic

applications due to their unique structure and exceptional properties, including high electrical conductivity, large surface area, excellent light transparency, and strong resistance to chemical and thermal changes. Their distinctive, extended π -system facilitates a highly mobile and direct pathway for charge transport. Research on carbon nanotubes has focused extensively on their potential applications in perovskite solar cells. Carbon nanotubes, in their fundamental state, serve as enhancements for charge extraction and stability or function as a layer for charge collection.

The electrical properties of carbon nanotubes determine their significance in solar cells. Metallic carbon nanotubes facilitate a direct conduction pathway for electrical charges. The addition of this specific type of carbon nanotube to the active layer diminishes the overall performance of the device. Semiconducting carbon nanotubes are preferred for the photoactive layer due to their capacity to minimize charge recombination and facilitate electron transfer to an external circuit.

The initial study on the integration of carbon nanotubes into perovskite solar cells was conducted in. In this research, the metal counter electrode in the n-i-p architecture of the solar cells was substituted with a laminated CNT network. The carbon nanotubes exhibited an efficiency of 6.9%, serving the dual function of both the electrode and the hole-selective layer. The CNT film's transparency facilitated the utilization of its dual-sided light power, resulting in enhanced efficiency. The absence of charge selectivity and the elevated resistance of the CNT film are probable factors contributing to the low efficiency. The incorporation of spiro-OMeTAD resulted in a power conversion efficiency of 9.9%, reflecting enhanced performance.

The researchers created a perovskite solar cell (PSC) by layering a double single-walled carbon nanotube (SWCNT) film on lead halide perovskite (LHP) and subsequently depositing spiro-OMeTAD. The absence of the metal top electrode may result in a solar cell efficiency of 15.5%. The device structure was obtained from a cross-sectional view using a scanning electron micrograph. Scanning electron microscopy of a cross-section of the solar cell revealed elongated projections, presumed to be SWCNTs, that detached from the film during the cutting of the device. This study offers extensive insights into the role of carbon nanotubes as a charge-conducting layer in perovskite solar cells.

Table 1. Advantages and disadvantages of different synthesis methods CNTs.

Methods	Merits	Demerits
Laser ablation	The incorporation of a metal catalyst into the carbon target leads to the synthesis of single-walled carbon nanotubes (SWCNTs) characterized by a narrow diameter distribution and elevation.	It is unsuitable for large-scale manufacturing.
Chemical vapor deposition	An extensive technique that also demonstrates how a multivariable process can be adjusted in a number of ways, including alcohol catalytic CVD, high-pressure CO disproportionation, plasma-enhanced CVD, thermochemical CVD, aerogel supported CVD, aerosol-assisted CVD, and hybrid laser-assisted thermal CVD.	Process parameter modifications are required to regulate the diameter distribution and yield of SWCNTs.
Electric arc discharge	Because of the high temperature employed, it is possible to make nanotubes with few structural errors. It is also an easy and affordable method that can yield a huge number of nanotubes.	The regulation of chirality in CNTs is not possible. If metal catalysts are used, it is necessary to purify the synthesized nanotubes. This procedure requires a high temperature.
	PECVD enables the formation of carbon nanotubes (CNTs) at much lower temperatures, sometimes even at room temperature, in comparison to thermal CVD (700–1000 °C). One potential avenue for exploration is the use of heat-sensitive substrates such as polymers or flexible electronics. The use of plasma in PECVD facilitates a heightened level of reactivity, hence enabling the improved manipulation of the properties shown by the carbon nanotubes (CNTs). The manipulation of plasma characteristics has the ability to affect the chirality, or handedness, of carbon nanotubes (CNTs), therefore affecting their electrical properties.	The use of high-energy plasma has the potential to induce the development of flaws inside carbon nanotubes (CNTs), which may have implications for their electrical and mechanical characteristics.
Catalyst method	It requires a low temperature.	It produces a low yield.
Arc discharge	It involves the mass production of carbon nanotubes (CNTs) and fullerenes.	It is a high-temperature process.
Electrocatalysis	This method has the following: a straightforward experimental setup; a controllable synthesis process through the use of electrolysis modes; inexpensive raw materials; controllable CNT structures; and the ability to dope carbon phases and morphologies in a single step through the optimization of electrolysis conditions and the electrolytic bath composition. Comparing electrolysis to other common CNT synthesis methods, this method is less expensive.	There are two primary issues with this approach, taking into account its drawbacks. The first is the issue with the graphite cathode breaking during the electrolysis process, and the second is the buildup of electrolysis byproducts in the bath, including carbon nanomaterials (the cathode), alkaline metal (the anode), and chlorine gas (the anode). These two issues hinder the ongoing performance and lead to process instability.
Hydrothermal/solvothermal	When compared to previous techniques of CNT synthesis, this method has three major advantages. First of all, the starting materials are readily obtained and stable at room temperature; second, CNT development may be achieved using this method even at low temperatures of 150–180 °C; and third, a hydrocarbon gas or carrier gas is not needed for the CNT growth process.	It produces a low yield.

The mechanical flexibility and exceptional charge transport properties of carbon nanotubes render them highly useful. This makes them suitable for application in portable devices, where they can be integrated into flexible perovskite solar cells (Table 2). Researchers manufactured photovoltaic solar cells utilizing a flexible fiber architecture. The configuration places the light-harvesting perovskite absorber between p-type flexible nanotubes and n-type core wire. The outermost layer of the cathode consisted of carbon nanotubes derived from a spinnable array in a dry state. The fiber-shaped perovskite solar cell demonstrated stability under bending conditions, achieving a power conversion efficiency of 3.3%. Despite not yet reaching an efficiency of 10% or higher, there is optimism regarding the potential of photovoltaic fabric.

Perovskite solar cells incorporating carbon nanotubes, both single-walled and multi-walled, as the hole-selective layer exhibit remarkable performance. Functionalization enhances the dispersion of carbon nanotubes in solution, rendering them a suitable alternative for p-type dopants in commonly utilized hole-transporting materials such as spiro-OMeTAD and P3HT. Researchers demonstrated the potential of using carbon nanotubes as an alternative doping process for

spiro-OMeTAD through a two-layered configuration of CNTs coated with polymers and incorporated into an organic matrix. In comparison to a device utilizing undoped spiro-OMeTAD, the device exhibited significantly greater charge extraction efficiency. The poor performance of the device, in comparison to a state-of-the-art PSC, can likely be attributed to the inability of the nanotubes to establish a sufficiently linked percolation network in this configuration. The fill factor is limited due to the device's performance being affected by series resistance. The insufficient direct interfacial contact between the LHP absorber and the carbon nanotubes resulted from their dispersion within the spiro-OMeTAD layer. As a result, they were unsuccessful in transferring holes from the interface. The carbon nanotubes established a highly interconnected network that directly interfaced with the low-halide perovskite layer following the sequential deposition of spiro-OMeTAD and CNTs. The absorber efficiently facilitated the transfer of photogenerated holes, enabling selective hole transport.

The primary advantages of utilizing carbon nanotubes in solar cells are their inherent robustness and longevity. Various methods have been outlined for the extraction of photogenerated charges from perovskite

solar cells, utilizing the charge transport characteristics of carbon nanotubes. A crucial advancement for the long-term viability of the technology involves replacing complete carbon nanotube structures synthesized in a solvent for application in the hole-selective layers and other expensive components of perovskite solar cells.

This would enhance the environmental sustainability and cost efficiency of the PSCs. Devices incorporating carbon nanotube components have the potential to enhance power conversion efficiency, positioning them as a viable and beneficial choice for reliable, high-performance electronics.

Table 2. Properties of CNTs.

Property	Comments	Value
Intrinsic mobility	Dopant concentration at 10^{17} cm^{-3} is over a hundred times more than silicon's at 300 K	At ambient temperature for individual CNTs: above $10^5 \text{ cm}^2/\text{Vs}$
Surface area	More than the $1200 \text{ m}^2/\text{g}$ value for activated carbon	$1600 \text{ m}^2/\text{g}$
On/off current ratios	Temperature one thousand times greater than that of a bilayer structure	Over 10^5
Thermal conductivity	Diamonds' is about $1500 \text{ Wm}^{-1} \text{ K}^{-1}$ greater	Reaching $3500 \text{ Wm}^{-1} \text{ K}^{-1}$.
Fracture stress	Size of the steel wires is about 50 times smaller after density normalization	50 GPa
Electrical conductivity	Comparable to that of mercury and other metals	$104 \times 10^6 \text{ S cm}^{-1}$
Free carrier concentration	Lower compared to graphene ($\sim 10^{20} \text{ cm}^{-3}$) and the majority of metals (such as silver, which has $\sim 10^{22} \text{ cm}^{-3}$)	$\sim 10^{17} \text{ cm}^{-3}$
Current carrying capacity	Copper has a 1000-fold lower value	Surpassing 109 A cm^{-2} for a single CNT
Young's modulus	Of a single-crystal diamond at ambient temperature	1–2 TPa

6: Carbon Nanotubes Utilized as Hole Transport Layers

The p-type characteristics of the hole transport layer impede the movement of photogenerated electrons while simultaneously attracting and retaining holes, facilitating their transfer from the active layer to the electrode. Consequently, an effective HTL will exhibit well-aligned energy levels, excellent thermal stability, and elevated hole mobility. Spiro-OMeTAD and PEDOT: PSS are often utilized as highly effective hole transport layers (HTLs) in p-i-n and n-i-p perovskite devices. Various factors have driven researchers to seek alternate hole transport layers (HTLs); nevertheless, these alternatives are sometimes prohibitively expensive, unstable, or excessively hygroscopic. Carbon nanotubes have recently been recognized as very promising hole transport layers in perovskite devices due to their superior hydrophobic properties, affordability, and remarkable durability.

A carbon nanotube sheet, coated with poly(methylmethacrylate), was utilized in an experiment

conducted by researchers. The efficiency of the perovskite solar cells was 5.82% when this film served as a hole transport layer (HTL). The researchers observed that the contact resistance between the perovskites and CNTs was lowered as a result of the increase in contact due to the shrinking of PMMA. In comparison to the PSC utilizing Spiro-OMeTAD as the hole transport layer, the device exhibited a significantly elevated VOC of 1.45 V. Notwithstanding this, the CNT layer demonstrated minimal charge carrier selectivity and was predominantly regarded as a conductor.

A feasible resolution to this problem is the development of hybrid hole transport layers by the integration of hole transport materials into carbon nanotube films. The double hole transport layers are constructed in n-i-p devices using single-walled carbon nanotubes covered with poly(3-hexylthiophene) and polymethyl methacrylate. Functionalization of P3HT enhanced the p-type characteristics and altered the electrical properties of SWCNTs. Simultaneously, PMMA was utilized on the surface of P3HT/SWCNTs to

inhibit oxygen and moisture from penetrating the perovskite layer by functioning as an encapsulant. The devices exhibited a final efficiency of 14.2%, improved stability, and the development of a hole-conducting protective layer.

Even though PMMA can't increase CNTs' hole transport capacity due to its insulating features, it could make PSCs more stable and efficient thanks to its protective and interfacial-enhancing characteristics. Swapping out PMMA with hole-conductive polymers is one way to boost the photovoltaic performance of devices. Researchers used a double-hole transport layer in their n-i-p devices [71]. The HTL was composed of a combination of P3HT/SWCNTs and undoped Spiro-OMeTAD. With a PCE of 15.4%, the perovskite solar cell outperformed its predecessor, which employed just undoped Spiro-OMeTAD and had a PCE of 6.8%. The addition of a hole transport bilayer containing single-walled carbon nanotubes sped up the charge extraction process, leading to an improvement in solar efficiency.

In the subsequent investigation, $\text{FA}_{0.83}\text{MA}_{0.17}\text{Pb}(\text{I}_{0.83}\text{Br}_{0.17})_3$ was utilized as the light-absorbing layer and SnO_2 as the electron-accepting layer in order to enhance the power conversion efficiency to 18.8%. This device outperformed others that employed undoped Spiro-OMeTAD or Li-doped Spiro-OMeTAD as HTLs. Scientists utilized ethylene vinyl acetate, an affordable and effective non-conjugated polymer, to distribute SWCNTs and MWCNTs in organic solvents effectively [75-76]. The process produced conductive films composed of superior SWCNTs and MWCNTs functionalized with EVA. Then, to achieve PCEs of 17.1% and 16.8%, respectively, perovskite solar cells' double hole transport layers were adjusted to use two films rather than P3HT/CNTs [77]. Combining the double HTLs with other hole transport materials, such as carbon nanotubes, results in highly efficient and stable perovskite solar cells.

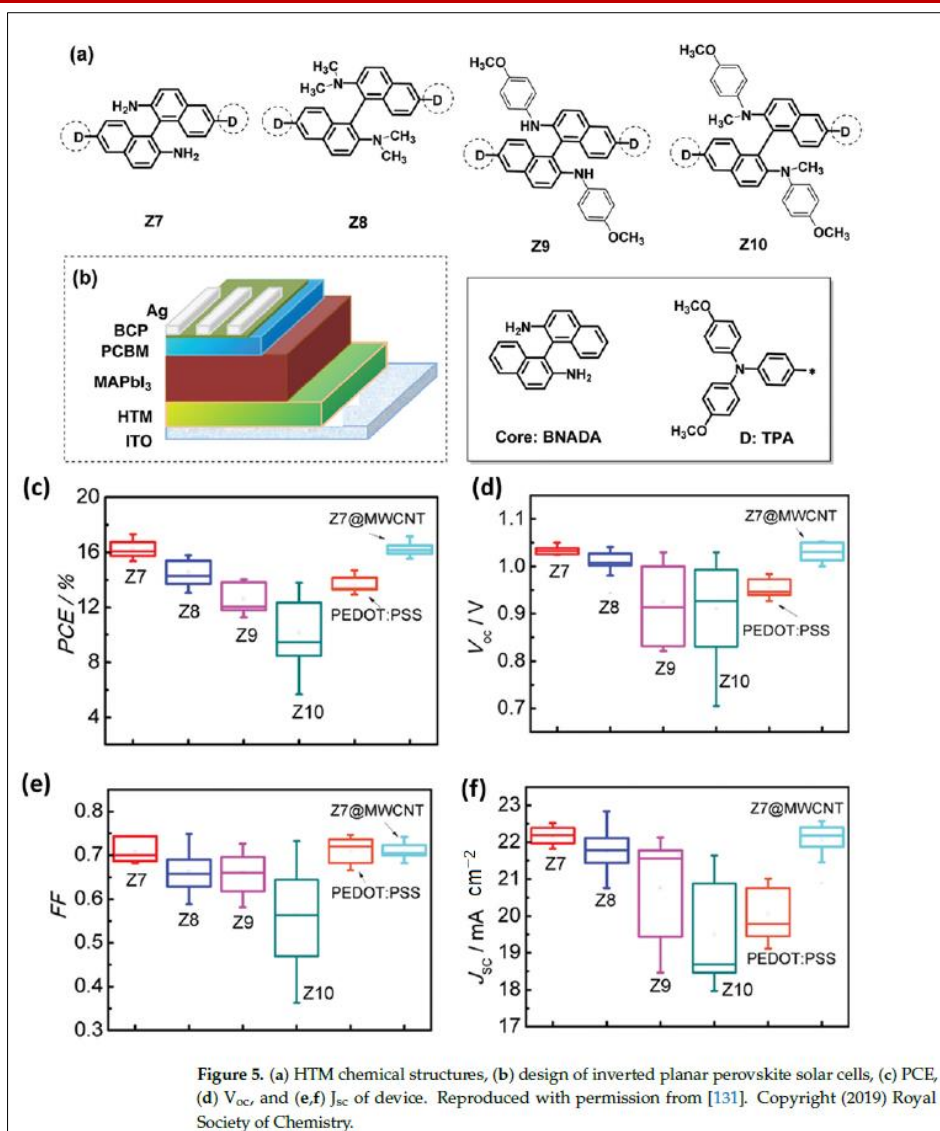
Researchers combined PEDOT: PSS with single-walled carbon nanotubes to create new hybrid

hole transport layers [73]. We designed and manufactured new hole transport molecules with different side chains based on binaphthylamine. Figure 5a,b shows that although the created materials are structurally and optoelectronically identical, their film characteristics are different. Physisorption allowed for the attachment of two kinds of hole transport molecules to multi-walled carbon nanotubes; these molecules were optimized with amine and methyl units.

Inverted perovskite solar cells were made using these functionalized carbon nanotubes. Similar to the original hole transport material's efficiency (17.32%), an unencapsulated device utilizing a hole-transporting material combined with multi-walled carbon nanotubes reached a power conversion efficiency of 17.17% [79-81].

A maximum power conversion efficiency (PCE) of 17.17%, a voltage of 1.052 V, a flux factor (FF) of 0.72, and a current density (J_{sc}) of 22.57 mA/cm² were all reached by the HTM@MWCNTs device (Figure 5c, 5d, and 5f, respectively).

In addition, the former shows higher device stability than the latter. An innovative method for improving the stability of inverted perovskite solar cells is to include carbon nanotubes into hole transport materials [74]. The end result was a robust chain structure that appeared to incorporate very small molecules. When comparing stability and efficiency, the p-i-n PSC without encapsulation achieved 17.2% whereas the PSC with a PEDOT:PSS-based hole transport layer reached 14.69%. Additionally, it was found that a hybrid material composed of carbon nanotubes and nickel oxide produced an outstanding layer for transporting holes. By incorporating carbon nanotubes into a solution that already included a precursor of nickel oxide, a hybrid hole transport layer (HTL) was produced [75]. This hybrid HTL showcased enhanced hole extraction and higher conductivity while preserving the optical and surface morphological characteristics of NiOx. The improved p-i-n planar device had a power conversion efficiency of 16.9%.



7: CONCLUSIONS AND FUTURE OUTLOOK

Due to its advantageous characteristics and fast efficiency improvements, metal halide-based perovskite solar cells (PSCs) have garnered significant attention from the photovoltaic industry. Perovskite solar cell designs offer several applications for one-dimensional carbon nanotubes (CNTs) owing to their remarkable electrical, optical, and mechanical characteristics. Included in these components are charge transporters, interface modifiers, perovskite additives, hole transport techniques, and charge-collecting electrodes. The use of CNTs in these components has significant potential for enhancing the performance and stability of PSCs. Exploring the potential of carbon nanotubes in perovskite solar cells is a burgeoning study domain; although significant progress in recent years, it remains imperative to address and examine numerous crucial issues.

Carbon nanotubes are crucial in photovoltaic solar cells due to their transparency and conductivity, facilitating little energy loss when sunlight reaches the

perovskite light absorbers. This study primarily aims to examine the features of single-walled carbon nanotubes (SWCNTs). However, the research indicates that SWCNT sheets lack sufficient transmittance and conductivity for use in perovskite solar cell applications. High-purity metallic single-walled carbon nanotubes are in high demand due to their superior conductivity compared to semiconducting or mixed nanotubes, and they can be acquired by synthesis or post-treatment methods. Additional research is required to develop high-performance PSCs, and the whole potential of DWCNTs in PSCs has yet to be realized. To enhance the conductivity and transmittance of CNTs, it is essential to choose appropriate dopants that will yield enduring benefits. Prior to adding more layers, it is essential to consider the smoothness of the surface of the CNT transparent films and the compatibility of the selected charge connections at the interface for researchers' work. A proficient method to achieve these goals is to integrate carbon nanotubes with other molecules, such as conducting polymers. Carbon nanotubes (CNTs) may serve as optimal transparent top contacts for the

development of efficient semitransparent devices and perovskite-based tandem solar cells. Advancements in integrated solar cells and the power conversion efficiency of photovoltaic technology rely on this research direction.

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