Emerging Internet of Things driven carbon nanotubes-based devices

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Received: 23 August 2021 / Revised: 28 October 2021 / Accepted: 9 November 2021

ABSTRACT

Carbon nanotubes (CNTs) have attracted great attentions in the field of electronics, sensors, healthcare, and energy conversion. Such emerging applications have driven the carbon nanotube research in a rapid fashion. Indeed, the structure control over CNTs has inspired an intensive research vortex due to the high promises in electronic and optical device applications. Here, this in-depth review is anticipated to provide insights into the controllable synthesis and applications of high-quality CNTs. First, the general synthesis and post-purification of CNTs are briefly discussed. Then, the state-of-the-art electronic device applications are discussed, including field-effect transistors, gas sensors, DNA biosensors, and pressure gauges. Besides, the optical sensors are delivered based on the photoluminescence. In addition, energy applications of CNTs are discussed such as thermoelectric energy generators. Eventually, future opportunities are proposed for the Internet of Things (IoT) oriented sensors, data processing, and artificial intelligence.

KEYWORDS

carbon nanotubes, electronics, sensors, electronic skins, Internet of Things, artificial intelligence

1 Brief introduction of carbon nanotubes

Carbon nanotubes (CNTs) have demonstrated extraordinary electronic [1,2], optical [3–6], and mechanical [7,8] and integrated into functional devices such as integrated circuits [9, 10],

biomedical imaging [11–14] and sensing [15–17], electromechanical systems [18], and super-stretchable ropes [19] for three decades [20–22]. The Internet of Things (IoT) era has enabled the broad applications of CNTs, such as universal sensors,

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data processors, wearable healthcare devices, and energy conversion and storage.

The properties of CNTs are determined by their chirality; therefore, the preparation of structure and morphology controlled CNTs become the most important goal in the community. To this end, there have been a number of reviews focusing different aspects of controlled CNT preparation, from chemical vapor deposition (CVD) growth [23–36] to solution-based post separation [37–43]. However, few reviews exist for listing the IoT-driven CNT applications.

In this review, we focus on the IoT related applications of CNTs (Fig. 1) including integrated circuits, synaptic arrays, chemical sensors, electronic skins, and DNA recognition. The emerging trends and future opportunities of CNT research are conveyed in the concluding remarks.

Figure 1 The emerging applications of CNTs based devices driven by the IoT.

Prior to the devices, we first look at the fundamentals of CNTs. Single-walled carbon nanotubes (SWCNTs) [44] are considered as cylinders rolling from a monolayer of graphene through the chiral vector $C_h = na_1 + ma_2$, where a_1 and a_2 are the unit vectors of the spatial lattice of graphene (Fig. 2(a)). According to the chirality, SWCNTs can be classified into chiral (n, m) , and achiral type. The achiral carbon nanotubes include the armchair (n, n) , and zigzag $(n, 0)$ types. According to their conductivity, the CNTs can be classified into metallic, semi-metallic, and semiconducting types [34, 45].

Due to the unique helical structure and property determined by chirality (n, m) , SWCNTs have become one of the important materials with considerable potential in many applications, such as nanoelectronics [9, 46–48], photonics [49], environment [50], energy [51], and bioimaging [52]. Nevertheless, the fabrication of high-purity identical SWCNTs has impeded the advancement in their use over the past three decades.

The electronic structure of a carbon nanotube has stemmed from that of graphene [58]. To start with, one needs to know the crystal structure of graphene in real space (Fig. 2(b)) and reciprocal space (Fig. 2(c)). In the extended first Brillouin zone of graphene, the conduction and valence bands touch in the Fermi level with six points [59]. The carbon nanotube shows different electronic structure compared with graphene because of the quantum confinement of electrons [60]. The momentum normal to the tube axis is required to be quantized, which provides periodic boundary conditions for the solution of energy equations. Such a quantization in a carbon nanotube results in the generation

of a series of discrete sub-bands as indicated by the cutting of parallel lines.

In the zone-folding model, the cutting lines were superimposed with the band structure of graphene (Fig. 2(d)). When these lines cross the Fermi point, the carbon nanotube is metallic (Figs. 2(e) and 2(f)). When these lines do not cross the Fermi point, it is semiconducting (Figs. 2(g) and 2(h)).

Specifically, (n, n) nanotubes (armchair tubes) are always metallic, and (n, m) nanotubes with $n - m = 3j$, where $j = 1, 2, 3, \dots$, are nearly metallic with a small, curvature-induced gap that has a $1/d^2$ dependence. Tubes with $n - m \neq 3j$ are semiconductors. The semiconducting SWCNTs (s-SWCNTs) exhibit chirality-dependent transition energy (E_{it}). The band gap (i.e., 0.5–1.5 eV) of s-SWCNTs is proportional to the reciprocal of the diameter (E versus $1/d$ relationship). The schematic band structure and optical transition were demonstrated for metallic (Figs. 2(i) and 2(j)) and semiconducting carbon nanotubes (Figs. 2(k) and 2(l)).

Here, the van Hove singularities (VHS) lead to narrow absorption and emission energy bands. The typical Kataura plot exhibits the relationship between transition energy and diameter, which was generally used to assign the chiral index (n, m) of SWCNTs [61, 62]. A sharp density of state (DOS) peak illustrates a narrow linewidth. Therefore, the nature of the transition in SWCNTs is the exciton transition rather than the band transition under strong electron interaction.

The small-sized bound electron hole (a few nanometers in diameter) with a considerable amount of binding energy (several hundreds of millielectronvolts) can be classified as Frenkel exciton; the foregoing is also the result of one-dimensional (1D) confinement along the nanotubes [63, 64]. Therefore, the customization of carbon nanotube with specific diameter and chirality becomes necessary for their optical and opto-electronic applications.

The conductivity of metallic SWCNTs (m-SWCNTs) can reach 10,000 times than that of copper [22, 65]. The m-SWCNTs exhibit a ballistic transport behavior, i.e., slight or no electron scattering occurs through the tube. Heat is not generated without phonons inside the tube lattice, viz., CNTs can conduct high currents while avoiding excessive temperature rise. This conductive property is advantageous to integrated circuits because the m-SWCNT network can be utilized to form transparent electrodes. S-SWCNTs transport electrons in a dispersed manner, although experiments show that they exhibit high mobility. Furthermore, CNTs have been employed as conducting electrodes in electrochemical systems. The large specific area of CNTs ensures the sufficient access of reactive species to environmental changes [66] and clean energy production [67].

During hydrogen evolution reactions, CNTs sustain their morphology via bubble generation [68]. In addition, CNTs have excellently performed as electrodes in supercapacitors. For instance, N-doped CNTs grown on a carbon cloth are used to host nickel nanoparticles and molybdenum carbide, thus forming a composite for electrode materials in a symmetric supercapacitor [69]. The deposition of electrode materials on the carbon cloth is compatible with the concept of flexible energy storage. Moreover, CNT-based composites enhance heating resistance. This conductor-supercapacitor integration leads to the fabrication of a self-powered heater capable of attaining temperatures exceeding 100 °C.

The CNTs also exhibit the excellent mechanical properties [7, 70], whose elastic modulus can reach 1 TPa, which is equivalent to that of a diamond and approximately five times that of steel. If other engineering materials are used as matrix with CNTs, the resulting composite material can exhibit improved

Figure 2 The crystal structure, electronic band structure, and optical transition of carbon nanotubes generated by the derivation of graphene. (a) The formation of carbon nanotube by rolling up of a graphene ribbon. (b) Real space and (c) reciprocal space view of hexagonal crystal structure. (d) The electronic structure of carbon nanotube generated by the zone folding of Brillouin zone of monolayer graphene. (e) Band structure (top) and first Brillouin zone (bottom) and (f) energy contour of graphene. The nanotube circumferential boundary condition was employed for the depiction of band structure of carbon nanotube. The solid parallel lines indicate the allowed states of a metallic carbon nanotube. (g) Band structure (top) and first Brillouin zone (bottom) and (h) energy contour of graphene. The lines indicate the allowed states of a semiconducting carbon nanotube. (i) Optical transitions in energy versus wave vector (E vs. k) in the k space and (j) the density of states of a metallic carbon nanotube. (k) Optical transitions in E - k space and (l) the density of states of a semiconducting carbon nanotube. (a) Reproduced with permission from Ref. [53], © Elsevier Ltd. 2016. (b) and (c) Reproduced with permission from Ref. [54], © Gao, W. L. et al. 2019. (d) Reproduced with permission from Ref. [55], © National Academy of Sciences 2010. (e) and (g) reduced with permission from Ref. [56], © IEEE 2003. (f), (h), (i), and (k) Reproduced with permission from Ref. [57], © Optical Society of America 2016. (j) and (l) Reproduced with permission from Ref. [34], © American Chemical Society 2020.

performance in terms of superior strength, flexibility, resistance, and isotropy [22]. When CNTs are used in polymer composites, metal powders, and ceramic substrates, the structure can be enhanced because CNTs can withstand 30% of the induced strain [71]. Moreover, functionalizing CNTs with silane molecule can enhance the solubility (chemical compatibility) of polymer-based composites. However, the presence of catalyst impurities (metal particles in general) may reduce the coefficient of heat conductivity by generating scattering sites.

The unique properties of CNTs have enabled many of IoT based applications. The understanding of structure–property relationship has driven the enormous research in the controlled synthesis of CNTs. We come to the brief discussion of already-reported synthesis achievements for chirality and structure regulation of CNTs.

2 Brief summary of controlled synthesis of CNTs

The CVD method is advantageous for the controlled growth of CNTs and mass production, because it affords tunable parameters to modulate the structure and morphology of CNTs [72]. Here we briefly summarized the recent progress of controlled synthesis of SWCNTs associated with the device applications.

The alignment of CNTs in both vertical and horizontal directions as well as CNT network (Fig. 3) are highly required in

integrated circuits, energy, and environmental applications. The vertically aligned CNT forest has been formed through the incorporation of water-assisted growth and regulation of catalysts [73, 74]. The horizontal alignment of CNTs has been achieved guided by the gas flow [75–77] and substrate lattice [78–82].

By continuously displacing the heating furnace assisted with gas follow direction, the longest CNT with half a meter has been produced [87]. Recently, horizontally aligned SWCNTs were also achieved through the solution-based assembling with high density [10, 17, 88–92]. Besides that, other technique such as acoustic-assisted assembly was also developed to prepare single-chirality SWCNTs from an ultralong tube [93]. The random SWCNT network is ideal for fabricating transparent conductive films [94–97] because of their small diameter, good optical and electrical properties, and excellent flexibility. Recently, the floating metal catalyst CVD method is developed to achieve high quality, density, and purity SWCNTs deposited on substrate [98–103]. The high intertube Schottky junction resistance [103], existence of aggregated bundles of SWCNTs, and performance–yield tradeoff [102] are key issues, which may lead to a degraded electronic and optoelectronic performance of the films.

The preparation of uniform-structure SWCNTs remains one of the most important challenges. Thanks to the continuous efforts, chirality pure CNTs are becoming increasingly accessible. There are several breakthroughs in both chirality-specific growth and

Figure 3 Selected synthesis strategies and growth mechanisms of CNTs with controlled geometry and structure. The schemes were adjusted and pieced together from references, including structure control from Ref. [34], morphology control from Ref. [30, 74, 81, 82], and growth mechanism from Ref. [83–86]. Reproduced with permission from: Ref. [34], © American Chemical Society 2020; Ref. [30], © WILEY-VCH Verlag GmbH Co. KGaA, Weinheim, 2010; Ref. [74], © American Association for the Advancement of Science 2004; Ref. [81], © American Chemical Society 2005; Ref. [82], © Macmillan Publishers Limited 2015; Ref. [83], © National Academy of Sciences 2009; Ref. [84], © Magnin, Y. et al., some rights reserved; exclusive licensee American Association for the Advancement of Science 2018; Ref. [85], © Chinese Chemical Society 2021; Ref. [86], © Macmillan Publishers Limited 2012.

sorting [104–106]. Li et al. reported the strategy of using intermetallic compounds with defined structures as catalysts to achieve direct synthesis of SWNTs with one dominant (n, m) of 80%–98% purity under proper kinetic growth conditions [27, 106–108]. Additionally, the chirality controlled SWCNT arrays with high density were also accessible by using solid carbide catalysts [36, 105, 109].

The growth mechanism of SWCNTs involving aspects of thermodynamics and kinetics revealed by computer [83, 84, 110–115] and *in-situ* techniques [85, 86, 116–124] became clearer. As to chirality-based sorting, Kataura et al. demonstrated industrial-scale separation of high-purity, single-chirality SWNTs by using mixed-surfactant gel chromatography [125] and the separation of 12 different single-chirality enantiomers with high enantiomeric purity [126]. Zheng et al., who developed the chirality-specific sorting of SWNTs by DNA [127, 128], employed aqueous two-phase extraction to separate 23 kinds of single chirality and helicity SWNTs, and separated the metallic and quasi-metallic SWNTs for the first time [129]. These rapid developments have revealed new findings to digest and new questions to contemplate.

Drawing directly from our first-hand experience and insight, we aim to give a comprehensive and critical review on application driven controlled synthesis of CNTs from three aspects: electronics, sensors, and thermoelectric power generators.

3 Carbon nanotube based electronic devices

The gained insight into the synthesis of CNTs enables the effective application of this knowledge to electronics, photonics, optoelectronics, wearable electronics, the IoT, and artificial intelligence. First, basic electronic devices are introduced, including field-effect transistor (FET), universal sensors, and logic gates.

3.1 Carbon nanotube-based FETs for integrated circuits

Moore's law states that new generations of semiconductors are developed every two or three years, whereas transistor density doubles, and fabrication cost is halved [130]. Dennard scaling provides the amendment to Moore's law, viz., new generation technology affords improved performance and reduces power consumption for the same device configuration [131]. The subthreshold swing limit has been attributed to the thermal electrons extracted by the gate electrode voltage, leading to current leakage, heat generation, and consumption of extra electricity. The 22-nm silicon technology has emerged with a subthreshold swing of $60 \text{ mV} \cdot \text{dec}^{-1}$, which does not match the theoretical limit. Planar FETs do not fully control the channel conductivity through the gate voltage.

A fin architecture as a top gate has validated the size reduction in channel length in a triple-gated fin FET (FinFET). Consumer electronics, such as smartphones and tablets, have satisfied the demands of consumers for entertainment with larger screens, faster data processing, and highly battery capacity. Smartphones have driven the technology update of chips with matured 14-nm and 10-nm FinFET architectures [132], whereas the 7-nm technology [133] and 5-nm node have emerged as test chips. For these, extreme ultraviolet lithography [134] has been developed to define the prototype with a 5-nm fin pitch resolution.

The cost reduction in silicon-based integrated circuits, stemming from increasing the device density, has led to the low cost of consumer electronics. However, the silicon-based complementary metal-oxide-semiconductor (CMOS) techniques have reached their theoretical limit without sufficient space for increasing the transistor density in wafers. The emerging technologies demand high computing capability and low energy consumption from the transistors. Moreover, healthcare and wearable electronics require lightweight, flexible, and stretchable semiconducting materials.

CNT electronics do not require a three-dimensional (3D) fin gate to achieve a 5 nm channel length. Notably, individual s-SWCNTs with a small diameter guarantee a small channel length without entailing a complicated lithography approach [46, 135, 136]. CNT-based high-performance CMOS devices have been scaled down to sub-10 nm nodes (Fig. 4).

The CNT-based integrated circuits with performance

Figure 4 Structure and performance of CNT based field-effect transistors with 5-nm gate length. (a) Architecture of the CNT based transistor. (b) Scanning electron microscopy (SEM) micrograph of an individual CNT bridging over source and drain electrodes with a distance of 5 nm (channel length). (c) Cross-section transmission electron microscopy (TEM) micrograph for the transistor with a gate length of 5 nm. (d) Transfer characteristic of the transistor. V_{Th} means the threshold voltage. SS means subthreshold slope. Reproduced with permission from Ref. [135], © American Association for the Advancement of Science 2017.

comparable to that of silicon-based $0.18\ \mu\text{m}$ CMOS has recently been demonstrated [137]. Moreover, a planar CNT FET [9] has exhibited superior performances, i.e., faster operation and lower power consumption, compared with those of $14\ \text{nm}$ silicon FinFET. Moreover, electrostatic control has been guaranteed with graphene as the top gate electrode for an electrostatic control. The CNT transistor has achieved a reduced subthreshold swing [135] of $73\ \text{mV}\cdot\text{dec}^{-1}$. Furthermore, a strategy based on employing a Dirac source has led to a decreased subthreshold swing of $40\ \text{mV}\cdot\text{dec}^{-1}$ averaged over four decades and a lower on-state voltage of $0.5\ \text{V}$ [138]. The Pd/CNT device features an Ohmic contact, leading to ballistic transport behavior [139]. Indeed, the metal contact with low-dimensional carbon nanomaterials contributes to conductance regulation [140, 141], electrical doping [142, 143], and carrier type reversal [144, 145]. In addition, the electric field could also lead to the increased tunneling probability [146] and ultralow noise level [147]. Besides, the distance between source and gate could determine the work frequency of the transistors [148]. In addition, the length of the Pd gate electrode has been optimized for the on-state current of CNT transistors [149].

Gigahertz-CNT transistors have exhibited the function of a five-stage ring oscillator [137]. Moreover, gigahertz-integrated

circuits with CNT transistors have demonstrated frequency mixers and multipliers [150]. The CNT resonator-based oscillator, an important source of constant alternating current signals, has suffered from nonlinear oscillation with thermal fluctuations [150]. In this case, the *in-situ* vibration of CNT oscillator has been recorded with a photonic microscope equipped with a micrometer optical cavity. The vibrating CNT transistor has exhibited single-electron conductivity with coherent output [151]. In suspended CNT mechanical resonators, interesting phenomena, such as insulating electronic states, have been observed [152].

The high-purity s-SWCNTs are highly required to achieve excellent performance over FETs. The m-SWCNTs are more reactive than s-SWCNTs because their DOS approximates the Fermi level. Introducing oxidative environment in the CVD process was widely proved effective to achieve the enrichment of s-SWCNTs by etching or restraining the nucleation of m-SWCNTs [153]. For example, the use of methanol [154, 155], oxygen [156], water vapor [157], oxidative supports [158], and UV-light [159] produced oxidative species ($\cdot\text{H}$, $\cdot\text{CH}_3$, $\cdot\text{OH}$), which effectively etched the m-SWCNTs (Figs. 5(a) and 5(b)). However, to date, the highest reported content of s-SWCNTs as grown by this strategy is less than 97%, which is still unavailable for high-performance FET such as low-power digital circuits.

Figure 5 Controlled synthesis of s-SWCNT through CVD growth and post purification. (a) Methanol assisted CVD growth. Reproduced with permission from Ref. [154], © American Chemical Society 2009. (b) UV-light assisted CVD growth. Reproduced with permission from Ref. [159], © American Chemical Society 2009. (c) Chirality specific growth by catalyst template. Reproduced with permission from Ref. [107], © American Chemical Society 2017. (d) Twisting the chirality of SWCNTs by reversing the electric field to grow s-SWCNTs. Reproduced with permission from Ref. [160], © Macmillan Publishers Limited, part of Springer Nature 2018. (e) Rate-selected growth of ultrapure s-SWCNTs. Reproduced with permission from Ref. [161], © Zhu, Z. X. et al. 2019.

Because of the complicated competition in the reactivity of SWCNTs between metallicity and diameter, it is very difficult to achieve ultrapure s-SWCNTs with this method. To address this problem, some new pathways for growing s-SWCNTs are developed to approach the daunting requirements of purity for applications.

For example, direct growth of s-SWCNT with specific chirality is another way to improve the purity of semiconducting species. Using the Co₂W₆ catalysts as structural templates, Li et al. achieved a preferential growth of (14, 4) semiconducting SWCNTs with the proper carbon supply [107]. For the (14, 4) samples, the chirality purity was further improved to 98.6% (Fig. 5(c)) with a high s-SWCNT content of 99.8% by the treatment using water moisture [107].

Jiang et al. introduced the electric field to the catalyst to amplify the difference in renucleation barrier between m- and s-SWCNTs, which leads to the dominant s-SWCNT growth over both kinds of tube edges [160]. With this strategy, s-SWCNT arrays with the high purity of 99.6%–99.9% were directly achieved by the electro-renucleation (Fig. 5(d)).

Wei et al. found a nearly 10-fold faster decay rate of m-SWCNTs was shown than that of the s-SWCNTs [161] and realized a spontaneous purification (99.9999%) to obtain long (> 100 mm) semiconducting tubes (Fig. 5(e)). Besides the direct CVD growth, post purification by gas-phase hydrocarbonation [162] and electric breakdown [163] was also developed to achieve such goal. However, the drawback of post etching/breakdown is that all SWCNTs are destroyed to some extent and tube density greatly decreased.

The target of density higher than 125 tube·μm⁻¹ and the purity of s-SWCNTs over 99.9999% required for high performance integrated circuits [164] is still far beyond the present capability of synthesis. In 2013, the SWCNT computer was developed by Shulaker et al. [47], demonstrating that CNTs are prominent among a variety of emerging technologies that are considered for the next generation of highly energy-efficient electronic systems.

However, the purity and density of s-SWCNT array are main problems.

Solution-processed self-assembling [10, 17, 90, 165–169] has been demonstrated to be a promising approach to achieve such target. For example, Peng et al. reported a multiple dispersion and sorting process to realize the extremely high (> 99.9999%) semiconducting purity [10]. Here, a dimension-limited self-alignment procedure was employed for preparing well-aligned SWCNT arrays (within alignment of 9°) with a tunable density of 100 to 200 CNTs/μm on a 10-centimeter silicon wafer (Fig. 6(a)). Top-gate field-effect transistors fabricated on the CNT array show better performance than that of commercial silicon metal oxide-semiconductor FETs with similar gate length [10].

Moreover, memory and processing devices of CNTs could be integrated in one chip [170]. In contrast, conventional computing systems have independent processors, random access memory, and storage. The CNT logic circuits have been transferred to arbitrary substrates, such as polymer, plant leaf, and a human wrist. In this case, read-only memory and full adder logic circuit have been demonstrated.

Thin-film SWCNT transistors have been compatible with biologic interface [171]. Moreover, the morphology of silver electrodes has been optimized to improve the conductivity of CNT transistors [172]. Analog and digital circuits, such as eight-stage shift registers and tunable-gain amplifiers [173], have been demonstrated by employing high-purity s-SWCNT thin films. More remarkable reports are expected to surpass previous limits. Park et al. developed an ion-exchange strategy to fabricate SWCNT arrays of individually positioned carbon nanotubes with a density of 10⁹ cm⁻² (Fig. 6(b)) [167]. A high density of SWCNT transistors was used to electrically test more than 10,000 devices in a single chip, showing a high purity of s-SWCNTs.

3.2 Floated-gated FETs and synaptic arrays

Floating-CNT transistors have considerable potential in

Figure 6 Solution-processed self-assembled SWCNTs. (a) The dimension-limited self-alignment procedure for preparing well-aligned CNT arrays. Reproduced with permission from Ref. [10], © Liu, L. J. et al., some rights reserved; exclusive licensee American Association for the Advancement of Science 2020. (b) Selective placement of carbon nanotubes by an ion-exchange process. The drain-source voltage was set as −0.5 V. Reproduced with permission from Ref. [167], © Macmillan Publishers Limited 2012.

the fabrication of artificial synapses for neuromorphic computing [174,175]. Compared with atomic switches and memristors, transistors have a facile work mechanism and high possibility for integration. The use of 9×8 CNT transistor arrays as synapses has successfully executed selective and parallel weight updates (Fig. 7) [176]. Such an artificial synapse array ensures the feature extraction of photographs through convolution operations, thus contributing to big data processing through neuromorphic computing.

To imitate the human brain, the fabrication of 300×300 synaptic array of SWCNT transistors with reservoir computing, which is basic for recurrent neural networks, has been demonstrated [177]. Artificial synapses have also been applied to signal processing, communication, memory, and machine learning. The synaptic operation has been utilized in localizing sound sources [178]. Therefore, the direct integration of memory and processor into a single device system (i.e., without incorporating an external circuit as an interface compared with the conventional von Neumann architecture) may reduce electric consumption [179]. Furthermore, a VO_2/SWCNT device has been found capable of generating a periodic sub-20-ns pulse signal, mimicking the spiking neuron [180].

A stretchable synaptic transistor array [181] can function as an excellent simulation of the synaptic performances of neurons (Fig. 8). Indeed, the polymer/neuron interface can provide a low contact resistance for efficient conformal brain–machine interface [181]. Besides, CNTs could function as electrodes for the brain–machine interface [182].

In addition, CNT synaptic transistors have been integrated with ferroelectric nanogenerator [183], demonstrating the potential applications of electronic skin.

4 Carbon nanotube-based sensors

The sensing mechanisms of CNT based devices include electrical, optical, electrochemical, and electro-optical types. The electrical sensors can be categorized as chemresistors,

chemicapacitors [184], and transistors [185]. The readers can find the comprehensive reviews elsewhere for chemicapacitors, transistors, photodetectors [186,187], nanomechanical resonators, and electrochemical sensors. In this section, we highlight the recent progress in chemresistors and optical sensors, which possess the feature of simplicity.

4.1 Chemiresistive biosensor for DNA detection

Various types of electronic device architectures exist for the CNT based sensors [188], including transistors, capacitors, and chemiresistors. Among them, the chemiresistors possess the simplicity with two electrodes, which satisfies the requirement of IoT applications [189], i.e., low cost, low energy consumption, and distributed sensor networks. Indeed, the chemiresistors are highly compatible with the integration of radio-frequency identification (RFID) tags [190,191].

The resistance of the CNT changes drastically upon the chemisorption of target molecules such as DNA [192] on the surface of CNTs [193,194]. The CNT chemiresistor based sensors have high promising in the IoT based healthcare system and early diagnosis [195].

The CNT-based chemical resistance biosensor for detecting the DNA sequence of the H5N1 avian influenza virus has been reported [196]. The performance of DNA sensor is assessed based on CNT based chemiresistors on rigid glass (Fig. 9(a)) and flexible substrate (Fig. 9(b)).

Direct-contact printing was used to mount horizontally-aligned CNT arrays [196]. These N doped multi-walled carbon nanotube (MWCNT) array act as a conductor channel after depositing the cross-finger electrodes over them (Fig. 9(c)). The SWCNTs were directly CVD-deposited over substrates and subsequently the electrode deposition leads to the device fabrication (Fig. 9(d)).

The work mechanism of the DNA sensor was briefly depicted. The DNA probe oligonucleotide was fixed onto the sidewall of the CNT (Figs. 10(a) and 10(b)). Then, the unoccupied CNTs were covered in the Triton buffer solution. A particular concentration of the H5N1 DNA solution was eventually dropped on the device for testing [196].

Figure 7 A typical floated-gated FET arrays based on s-SWCNT network film for synaptic parallel computing. (a) A schematic of the artificial neural network for neuromorphic computing. (b) Parallel access to arrays of synaptic crossbars. (c) The design-under-test of the integrated circuits based on CNT transistor arrays. (d) Fake-colored optical micrograph for the 9×8 CNT transistor arrays as magnified region from (c). (e) Optical micrograph for three CNT based float-gated field-effect transistors. Reproduced with permission from Ref. [176], © The Royal Society of Chemistry 2019.

Figure 8 Stretchable CNT-based synaptic transistors. (a) Electrodes arrays for stretchable transistor by inkjet printing. (b) The cross-section view of an individual transistor. (c) The photograph of flexible transistor array over a human hand. Reproduced with permission from Ref. [181], © Molina-Lopez, F. et al. 2019.

Figure 9 Carbon nanotubes based chemiresistors. Photographs of chemiresistor arrays over (a) quartz substrate and (b) polyimide foil. SEM graphs of chemiresistor demonstrating (c) the cross-finger electrodes and (d) s-SWCNTs bridging the electrodes. Reproduced with permission from Ref. [196], © Elsevier B.V. 2017.

The sensor performances based on s-SWCNTs and N doped MWCNTs were compared. As shown in Fig. 10(c), the resistance changes according to the specific binding of the DNA probes. By varying the electric conductance of SWCNTs with surface adsorbents, the resistance is plotted versus different test solutions (Fig. 10(d)). The CNT resistance responded to diverse DNA concentrations (Figs. 10(e) and 10(f)). The declining trend of resistance is the same for the two resistors based on N doped MWCNTs and s-SWCNTs. However, the s-SWCNT based device shows a sensitivity level that is 1,000 times higher than that of N doped MWCNTs, i.e., with 20% resistance upon the probing of 200-pM DNA.

The difference in the concentration range between the two could be attributed to the electronic properties of the metallic and semiconducting CNTs. The additional effect of the Schottky barrier modulation on the metal/CNT interface is significant at the lowest reliable detection concentration.

The detection principle of resistance change is caused by the interaction between the DNA target [196] and CNTs. This detection technique affords the advantages of miniaturization, low power consumption, high accuracy, and label-free detection.

4.2 Gas and solvent sensors

Sensing methodologies utilizing electrical conductivity as a transducing element are well developed technologies with extensive examples in gas phase, liquid phase, and biological

Figure 10 The carbon nanotubes based chemiresistor for DNA sensing. (a) Scheme of DNA recognition mechanism in the chemiresistor based biosensor. (b) Photograph of the probe station for electrical measurement of carbon nanotube chemiresistor. The relative resistance (R/R_0) of the sensors depending on the materials: (c) N doped MWCNTs and (d) s-SWCNTs. The applied voltage was 2.0 V. The relative resistance (R/R_0) of the DNA sensors at different concentrations of DNA T by effective materials: (e) N-MWCNTs and (f) s-SWCNTs. The 0 pM means no DNA incorporation. Reprinted (adapted) with permission from Ref. [196], © Elsevier B.V. 2017.

sensing. Several types of nanomaterials, such as nanostructured-metal, metallic nanoparticles, metal complexes, organic polymers, and carbon-based nanomaterials, have already emerged as active elements for gas sensors. Thus far, transition metal oxide-based gas sensors exhibit efficient detection of carbon monoxide at operating temperatures not exceeding 200 °C. Modified SWCNTs for ammonia gas detection can be highly sensitive and power-efficient when operating at room temperature.

The CNT-based gas sensors are facily fabricated and can be easily scaled up (Figs. 11(a) and 11(b)). The main component is the s-SWCNTs, which are sufficiently long to bridge electrodes (Fig. 11(c)). The gas sensors are highly sensitive and have a detection threshold of 1.5 ppm [197].

The deposition of dispersed CNTs onto the electrode area can be controlled. The sensor with the highest sensitivity possesses a low power consumption of 0.6 μ W [197], suggesting that it is ideal for mobile applications or remote environment monitoring. Furthermore, the gas sensor sensitivity can be further improved by reducing the CNT bundles during the CVD synthesis.

Figure 11 The ammonia gas sensor by the chemiresistor of carbon nanotubes. (a) The photograph of the gas sensor. (b) The X-ray photoelectron spectroscopic data of CNTs interacted with the bonding of oxygen atoms. (c) Scheme of the gas sensing device with the resistance of CNTs. (d) The relative resistance $\Delta R/R_0$ of CNTs under exposure to the different concentrations of ammonia. Reproduced with permission from Ref. [197], © American Chemical Society 2018.

Moreover, CNT sensors are capable of detecting ozone and nitrogen dioxide within 1 min [198]. Hence, these low-cost gas sensors could be used in detecting hazardous gases in the IoT. Success in detecting organic solvent molecules has also been achieved using SWCNTs. Nevertheless, pristine SWCNTs exhibit a nonspecific response to chemical exposure. To achieve the selectivity of targeted analytes, functional groups have to be formed on the surface of nanotubes.

Some aromatic molecules and surfactants with π - π bonds, viz., noncovalent functionalization forms, can physically adsorb SWCNTs. However, noncovalent functionalization has one problem-limited stability. This problem has been successfully resolved with the employment of poly(phenylene ethynylene) (PPE) [199]. The PPE polymer has a molecule of pentiptycene (Fig. 12(a)), which strongly interacts with SWCNTs due to their equal diameter (Figs. 12(b) and 12(c)).

The operation mechanism of the gas sensors is demonstrated in Figs. 12(d) and 12(e). The organic solvent sensor also highly responds to various benzene concentrations (Fig. 12(f)). Moreover, the chemiresistance has an excellent linear proportional relationship with benzene concentration (Fig. 12(g)). The sensors subsequently show successful detection of aromatic hydrocarbons.

4.3 Heavy metal ions sensing by electrochemiluminescence logic gates based on CNTs fluorescence quenching

The precise detection of heavy metal ions is extremely important for environmental protection and remediation. Due to the interaction with protein, Hg^{2+} can cause massive and irreversible damage to the brain, intestines, and kidneys. Both Hg^{2+} and Ag^+ are the primary heavy metal ion pollutants in lakes and rivers. In contrast, Ag^+ can deactivate the sulfhydryl enzyme. Electrochemiluminescent molecular logic gates exhibit satisfactory performances in determining the concentration of these two metal ions.

The work mechanism of logic gates with electrochemiluminescence is briefly depicted as follows. It is different in output signals compared with the conventional CMOS based logic gates

Figure 12 Organic solvent sensors based on PPE polymer/SWCNT composite for the examination of benzene, toluene, and xylene. (a) The molecular structure of pentiptycene analogous to the uppercase letter H. (b) Cross-section and (c) 3D view of the well binding of SWCNT with pentiptycene containing PPE polymer. The SWCNTs were well dispersed along the framework of PPE polymer due to the perfect match of their sizes. (d) Relative conductance and (e) changes in frequency and dissipation as the sensor performances upon the introduction of chemical vapors. The ΔG means the change of conductance with G_0 for initial conductance without gas vapor exposure. ΔF_3 means the changes in frequency at 3rd overtone. ΔD_3 means the dissipation factor at 3rd overtone. The QCM-D means the quartz crystal microbalance with dissipation monitoring. (f) Sensitivity of chemiresistance for PPE polymer/SWCNT composite with various concentrations of benzene. (g) Comparison of chemiresistance response of gas sensors with various polymers under distinct concentrations of benzene. The P1, P2, and P4 mean three types of pentiptycene polymers. The SG65i means the (6, 5)-enriched SWCNTs. Reproduced with permission from Ref. [199], © American Chemical Society 2020.

by the measurement of output voltage, viz., high voltage means 1 and low voltage means 0. Here, the electrochemiluminescence intensity was employed as the output signal and the chemisorption-based binding of fluorescent tags with CNTs as input. When the fluorescent tags were immobilized on CNT surfaces, the electrochemiluminescence is quenched to low intensity, which means 0. When the fluorescent tags were destabilized by the emergence of heavy metal ions and released from CNT surfaces, the electrochemiluminescence is recovered to high intensity, which means 1.

Early-stage logic gates for detecting Pb^{2+} and K^+ ions have also been reported [200]. However, these are complicated to handle and not portable. Logic gates with electrochemiluminescence are better than colorimetric logic gates. Although various ions can form stable complexes by bridging specific nucleotide bases, the mismatch of the base pair remains a significant problem.

A logic gate architecture [201] has been fabricated by employing the merits of the CNTs-aptamer composite. The oxygen-containing groups can easily attach to the ends and defects on the CNT sidewall (Fig. 13(a)). The Ru-silica ($\text{Ru}(\text{bpy})_3^{2+}$ -doped silica) with T rich (S1) and C rich (S2) oligonucleotides as tags were firstly adsorbed on the surface of CNTs in presence of Hg^{2+} . Here, the coordination of T base- Hg^{2+} -T base stabilizes the hybridization of oligonucleotides. Then, the CNTs could quench the fluorescence of the Ru-silica/oligonucleotides. With the input of S1, I^- , or both of them, the Ru-silica/oligonucleotides get liberated from the surface of CNTs, which result in the recovery of the electrochemiluminescence (Fig. 13(b)).

Eventually, a NOR gate was formed (Figs. 13(c) and 13(d)). This protocol could be extended to AND, OR, and INHIBIT logic gates as well (Fig. 13(e)). Therefore, this logic gate could identify different heavy-metal ions in complicated chemical environments.

Figure 13 The carbon nanotubes-based electrochemiluminescence logic gates for Hg^{2+} ions detection. (a) Schematic diagram of the logic gate with the inputs of I^- and S1 tag, and the output of the electrochemiluminescence. The S1 is T rich oligonucleotide and S2 is C rich oligonucleotide. (b) Electrochemiluminescence (ECL) intensity curves of the NOR logic gate (after Hg^{2+} treatment) versus time with different input conditions of S1 and I^- (from top to bottom: no S1 and no I^- ; only S1; only I^- ; both S1 and I^-). (c) The symbol diagram of the NOR logic gate. (d) The truth table of the NOR logic gate. (e) Schematic diagram of three different types of logic gates including AND, OR, and INHIBIT. Reproduced with permission from Ref. [201], © The Royal Society of Chemistry 2016.

4.4 Photoluminescence for DNA sensing

Carbon nanotubes possess intense light-absorption with the first near-infrared window of 750–1,000 nm, which produces *in-situ* fluorescence imaging [202, 203]. Then, the CNTs as fluorophores exhibited the intrinsic fluorescence emission at the second near infrared window of 1,000–1,700 nm [204, 205]. Therefore, the CNTs become emerging nanomaterials as a fluorescent-contrast agent for deep tissue fluorescence imaging [52].

The gel filtration method was employed to realize the enrichment of semiconductor (12, 1) and (13, 3) single-walled carbon nanotubes (Figs. 14(a)–14(c)). Upon the resonance absorption of 808 nm photons, these carbon nanotubes emit photons of 1,200 nm, termed photoluminescence emission (PLE). The photoluminescence intensity of chirality-sorted single-walled carbon nanotubes is ca. 5 times higher than that of unsorted single-walled carbon nanotubes (Fig. 14(d)). These successfully demonstrate the real-time *in vivo* imaging of the whole body of mice (Fig. 14(e)). Besides, it can be applied for indicating the tumor vessels with high resolution. The CNT based optical PLE method has guaranteed the tumor recognition and sensing [206].

Figure 14 Typical photoluminescence imaging of the carbon nanotube. (a) A photograph of enrichment of (12, 1) and (11, 3) chirality-sorted SWCNTs after a density gradient ultracentrifugation process. (b) The optical absorption spectra of the SWCNTs. (c) The PLE mapping (vs. excitation) of (12, 1) and (11, 3) SWCNTs. (d) Raman spectra. (e) NIR-II fluorescence image and (f) principal component analysis image of the inner organs of mice after the injection of chirality-sorted SWCNTs. Reproduced with permission from Ref. [206], © American Chemical Society 2012.

In the biomolecular sensing and biomedical imaging of biological tissues, the optical systems involve molecular labeling, photoluminescence, and fluorescence. Conventional labeling agents, such as dyes and fluorescent proteins, suffer from photobleaching. Ultraviolet light damages living tissues; the imaging of cells is invasive and causes the death of cells or tissues after irradiation. In the visible light range, tissues exhibit high absorption and scattering, leading to low probing depth. The near-infrared range, particularly the 750–1,400-nm range [207], is capable of high-penetration depth due to the negligible resistance to absorption of the tissues.

As a fluorescent material, SWCNTs show resonant emission in the near-infrared range, e.g., 980 nm for the (6, 5) tube [208]. Moreover, they can be used to fabricate non-photobleaching materials that remain robust when irradiated with near-infrared light [209]. The wrapping of SWCNTs with DNA ensures the successful individualization of CNTs. This separation between two neighboring tubes boosts the quantum yield of photoluminescence [210]. A single-stranded DNA exhibits sequence-determining interaction with CNTs. The DNA adsorption on SWCNTs depends on the pH level and oxygen concentration in an aqueous solution [211]. Furthermore, the

deformation of DNA could lead to a different degree of dispersion during the separation of SWCNTs [212].

When a s-SWCNT is irradiated by laser with a wavelength matching its optical transition energy band, it emits a photon due to the exciton recombination at the band edge; this is termed photoluminescence. Moreover, the diameter and chirality of the SWCNT determine the energy of transition (termed E_{11}). It can function as a powerful tool for imaging and sensing in the near-infrared II range, which has a non-invasive influence on biological tissue.

The SWCNTs have excellent biocompatibility and can noncovalently interact with organic molecules, such as DNA. Moreover, they can be targeted when the structure remains unchanged after the combination of aptamer and CNTs [213]. The emission of SWCNTs at the near-infrared range is quenched by the surface of heme, whereas the specificity demonstrates that they do not react with other proteins. Therefore, the specific binding of the receptor and target material leads to the shift in the wavelength or photoluminescence intensity in the CNT (Fig. 15).

Figure 15 Scheme of DNA detection by photoluminescence. The wavelength shift or extinction caused by the combination of target detection substance and aptamer. (a) Reproduced with permission from Ref. [214], © The Royal Society of Chemistry 2013. (b) Reproduced with permission from Ref. [213], © The Royal Society of Chemistry 2017. (c) Reproduced with permission from Ref. [215], © Pan, J. et al. 2017.

The emerging trend lies in the utilization of the NIR-II therapeutic window [216]. The near-infrared range could be divided into two sections [217], i.e., NIR-I (700–900 nm) and NIR-II (1300–1700 nm). In particular, the NIR-IIa light at the range from 1,300 to 1,400 nm has considerable potential as an irradiation source in early diagnosis, chemotherapy [218], and photothermal therapy [219]. Therefore, SWCNTs can be conjugated with light-sensitive agents, such as green fluorescent protein [220], exploiting both large specific area and targeting capability.

The transparent conduction of electrodes is important to photoelectronic devices, such as solar cells and photodetectors. Commercial transparent conducting oxides, such as indium-tin oxide [221] and fluorine-doped tin oxide [222], are predominantly n-type doping due to oxygen vacancies. The limitation for transporting electrons does not satisfy the hole transport requirement for collecting generated solar absorber materials.

The CNTs can be easily tuned as a p-type material through boron doping [223]. The CNT films can afford an efficient alternative for the hole transport layer in perovskite solar cells [224]. Moreover, it may provide an innovative solution to the problem of conventional p-i-n microcrystalline-amorphous silicon thin film-based [225] solar cells with a p-type absorber directly in contact with the top electrode, i.e., indium-tin oxide over glass. Such a top electrode has boosted the SWCNT/a-Si:H solar cell efficiency to 8.8% [226]. It may be utilized as a general top electrode to match the p-type solar absorber materials.

The s-SWCNTs and their heterostructures have been employed for improving the performance of photonic and optoelectronic

devices [227]. The transition energy can be regulated with the dielectric environment for immobilizing CNTs. Upon the generation of a strongly bound exciton [227], the excited state generates strong dissociation energy of approximately 0.5 eV through illumination, flow of hot carriers, and electro hole recombination. The CNTs have photonic applications, such as light source, light wave guide, laser, and nonlinear saturable absorber [228].

Rapid photoluminescence results from the radiative decay of excitons [229]. The band of photoluminescence could be tuned with a gate voltage on the CNT FET. Moreover, the energy levels of SWCNTs have been regulated via the surface functionalization of single-stranded DNA [230]. High-intensity electroluminescence originates from the hot carriers under unipolar conditions. In addition, the illumination of one SWCNT using polarized light has been developed for determining complex optical susceptibility [231]. High-intensity electroluminescence originates from the hot carriers under unipolar conditions. In addition, the illumination of one SWCNT using polarized light has been developed for determining complex optical susceptibility.

Photonic quantum science has extended applications, such as quantum computation [232] and quantum cryptography. The photonic circuits include single-photon light source, linear optical components, and detectors. Moreover, the use of a single-photon light source has become the major focus in the concept of utilizing a photon as an information carrier [233]. Electrically driven CNTs emit highly compatible and intense single photons [234]. Hence, the coupling with the nanophotonic cavity could boost proton emission [235]. The single-photon purity of the CNT light emitter has been observed in the telecom wavelength with defect sites [236], solitary dopant [237], or covalently functionalization [238] for photon emission. The band of emitted light can be narrowed down with the aryl edge functionalization of zigzag CNTs [239].

The (9, 8) SWCNT device with a 10-nm channel length has excellent electroluminescence performance in the telecommunication band. Here, luminescence from excitonic and trionic recombination can be turned on and off with the gate voltage. Moreover, the Sc/CNT/Pd contact has led to the fabrication of light-emitting diodes [240]. The magnetophotoluminescence of CNT can possibly be applied to quantum computing and spintronics [241].

The interfaces of SWCNTs-Si can form a heterostructure junction for solar cells. The excitons at a higher energy level generate photovoltage and photocurrent that can be applied to solar cells and photodetectors. Pristine SWCNT films have demonstrated superior photodetector performance in the visible and near-infrared range [242]. The CNT optoelectronic devices also have considerable potential in neural interfaces [243]. With three cascading configurations, SWCNT film photodetectors linearly amplify the output photovoltage [244]. The Schottky junction formed in SWCNT-separated graphene has also exhibited excellent photoresponse [245].

The p-n diode of SWCNT/MoS₂ ensures the tuning of conductivity from insulation to rectification depending on the gate voltage [246]. Moreover, the perovskite-CNT-based phototransistor has elevated photodetection at low-illumination intensity (i.e., nanowatt per square millimeter) [247].

Two photodetectors based on CNTs have been used to form an NOR logic gate [248] as well as AND gate circuit [249] through the integration of a silicon waveguide in a single chip. In addition, the CNT-Si junction [250] has demonstrated the use of the AND and ADDER/OR gates as well as a four-bit digital-to-analog converter. In the CNT-Si junction, the (7, 5) and (7, 6) tubes have contributed to photocurrent generation [251]. The heterostructure

of CNT/h-BN has resulted in bright photoluminescence with a narrow peak width [252]. Moreover, the W/CNT junction has played a role in thermophotovoltaics [253] for converting heat into electricity.

4.5 Pressure and strain sensors for electronic skin

Pressure sensors for healthcare and tactile applications have been fabricated using CNT-polymer composites [254]. The fabric, modified with tactile CNT-polydimethylsiloxane, is transformed into wearable electronic skin [255]. The pressing motion and friction could be recognized based on the difference in electrical resistance, i.e., high and low resistance values for tangential and normal forces, respectively. The MWCNT-gelatin hydrogel, whose deformation is correlated to the electrical resistance, has ideal electronic skin performance [256]. The strain sensors of CNTs have been incorporated into a drone system for human-machine interaction [257].

Piezoresistive electronic skin (E-skin) is an essential part of human-machine interaction and requires high spatial resolution. To resolve this, a simple approach is to fabricate a patterned piezoresistive layer. A composite of polymethyl methacrylate/CNTs (Figs. 16(a) and 16(b)) has been blended and prepared. Such a composite features a holey honeycomb array in the course of molding with ZnO nanorods as a template [258].

The piezoresistive device features ultrahigh sensitivity to a pressure value of 88 kPa (Fig. 16(c)) and a fast response time of 110 ms (Fig. 16(e)). The current-pressure curve of the film can be divided into three stages. In the first stage, the sensor displays linear sensing behavior in the 10-kPa range with high sensitivity to 88 kPa⁻¹. The nanopore channel structure begins to deform and disappear, and resistance rapidly diminishes with increasing contact area. The second stage ends with the 115-kPa pressure, which is an ideal range for daily motion sensors. Such sensors exhibit average sensitivities of 6.25 and 0.38 kPa⁻¹ in the pressure ranges 10–45 and 45–115 kPa, respectively. At this stage, film deformation becomes difficult; hence, its resistance gradually decreases. In the third stage, the film has reached its deformation

limit; the resistance barely changes with the increase in pressure. The stability of the film was examined by loading and unloading the material with 10-kPa pressure in a 1-s cycle. After 800 cycles, the sensor was highly steady (Fig. 16(f)). This demonstrates the considerable potential of CNTs as a conductivity-enhancing additive material for electronic skin applications.

In addition to the above, CNTs have exhibited success in humidity sensing. The fabrication of electrochemical sensors and breath sensors have been demonstrated using sericin-CNT materials printed on flexible substrates [259]. Printed electronics have become facile using a silk protein ink-CNT composite. The silver nanowire-CNT composite has been employed as a heating material for thermotherapy pad [260] integrated with a thermochromic indicator for controlling overheating.

Triboelectric nanogenerators based on CNT composites have been demonstrated as highly efficient high-power energy harvesters [261]. These nanogenerators can be integrated with tactile sensors as a self-powered device [262]. Moreover, CNT yarns have been adopted in inertial sensors to detect human motions, such as walking, jumping, and squatting [263]. Inertial sensors are self-powered by collecting mechanical energy from coiled CNT yarns.

4.6 Thermoelectric power generators

Thermoelectric materials have demonstrated their capacity to generate power in the IoT, wearable heating devices [264], and biomedical implants [265]. The CNT yarns sewed into textiles were observed to generate thermoelectric power with high output power density and satisfactory specific power [266].

The thermoelectric effect ensures the conversion of thermal energy into electricity, affording a strategy for electricity production or refrigeration [267]. This highlights the necessity of identifying alternative materials that can be employed to achieve excellent thermoelectric performance through waste heat utilization [268]. Moreover, surface functionalization may influence phonon transport and charge carrier transport of nanomaterials.

Figure 16 Piezoelectric characteristics of carbon nanotubes films. (a) Schematic diagram of the pressure sensor and (b) the measuring system of mechanical force. (c) The variety of current depends on the changing pressure within 0 to 115 kPa. (d) Corresponding stress distribution calculated in simulation software COMSOL. (e) Pressure response diagram of the CNTs sensor with the top section pressure of 25 kPa. (f) The operational steadiness of the CNTs sensor gauged for ca. 800 cycles with the pressure of 10 kPa. Reproduced with permission from Ref. [258], © American Chemical Society 2020.

Low-dimensional materials have demonstrated considerable potential with their high thermoelectric figure of merit [269], high electrical conductivity [270], and low in-plane thermal conductivity [271]. Indeed, CNTs have exhibited performance (i.e., figure of merit) comparable to that of state-of-the-art thermoelectric materials, such as organic polymers [272]. Two other parameters are necessary for evaluating thermoelectric performance: the Seebeck coefficient and power factor [273]. To collect more heat and increase the conversion efficiency, low work temperature is necessary [274]. For the integration of flexible and stretchable electronics, flexibility is also significant [274].

An integrated thermoelectric device requires a p-n junction to generate voltage and power outputs [273]. In this regard, the homostructured junction between p-doped and n-doped SWCNTs exhibits high power factor and charge carrier mobility. Early-stage doping has been achieved through the physical or chemical adsorption of functional radicals or molecules [268]. However, functionalization is not sustained at high work temperatures. Hence, sustainable doping is required and extremely important.

Efficient n-type doping of SWCNTs can be achieved via ammonia plasma treatment; SWCNT films have been tuned from p-type doping to n-type doping using this facile strategy [275]. Here, different treatment times have been investigated according to their optical absorption and thermoelectric performance (Fig. 17).

An efficient p-doping of SWCNTs has been realized through boron incorporation. Argon-protected annealing, which is implemented by mixing B_2O_3 , has been achieved as a result of successful boron-doping [276]. The power factor reached $226 \mu W \cdot m^{-1} \cdot K^{-2}$ for this excellent material (Fig. 18).

Seebeck coefficients of 125 and $133 \mu V \cdot K^{-1}$ have been observed in a thermoelectric device for p-doped and n-doped SWCNTs, respectively [275]. Superior power factors of 95 and $55 \mu W \cdot m^{-1} \cdot K^{-2}$ have also been obtained for these SWCNTs, respectively. These performance levels exhibited sustainability at a temperature range

of 25–100 °C. Further work is necessary to explore thermoelectric power efficiency and output voltage.

The open-circuit voltage and output power of thermoelectric power generators fabricated using p-doped SWCNTs are 20 mV and 400 nW, respectively [276]. High efficiency and voltage are expected upon the integration of n-doped SWCNTs with the p-n component. The further doping of CNTs with boron nitride can modulate the thermal conductivity and boost the power factor of the thermoelectric material [277]. The edge terminations of graphene nanoribbons [278] can also improve the Seebeck coefficient [279]. The production of other puckered two-dimensional materials has been predicted using numerical calculation approaches [280]. The thermoelectric generator has been integrated into the electrocatalyst device to utilize the heat generated during electrocatalysis for water splitting [281].

Future opportunities remain by improving the Seebeck coefficient and electrical conductivity. Pristine CNTs exhibit thermoelectric power factors in the range of 700–1,000 $\mu W \cdot m^{-1} \cdot K^{-2}$ for both n-type [282] and p-type doped nanotubes [272]. The values are comparable to those of conventional thermoelectric materials [283], such as PbTe and BiTe₂. Accordingly, numerous opportunities await the development of SWCNT-based thermoelectric generators in terms of conversion efficiency and power.

Highly aligned CNTs have been employed in thermoelectric generators without metal contact [284]. This reduction in the contact resistance between CNTs and electrode has boosted the output power of thermoelectric generators. Moreover, by incorporating a thin layer of poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS), the contribution of metallic CNTs has been suppressed [285]. Here, PEDOT:PSS functions as an organic electrochemical transistor and creates an energy barrier to abate the thermal power from metallic CNTs. Furthermore, the composite of CNT and poly(dimethylsiloxane) as a thermoelectric material exhibits flexible conformal features [286].

5 Future opportunities in the IoT and artificial intelligence

Great opportunities remain for the applications of carbon

Figure 17 Thermoelectric performances with single-walled carbon nanotubes. (a) Scheme of the heterostructured thermoelectric junction. The p-type doping of SWCNTs has been achieved through the adsorption of water and oxygen. The n-type doping has been obtained with N substitution by ammonia plasma treatment. (b) Optical absorption spectra and photographs of SWCNT films after different plasma treatment duration. (c) Seebeck coefficient and electrical conductivity of SWCNT materials with ammonia plasma treatment for the different time duration. The unsorted SWCNTs mean the as-delivered SWCNT samples from the supplier, which contain some metallic tubes. The sorted s-SWCNTs stand for the enriched semiconducting SWCNTs after sorting. (d) Seebeck coefficient and electrical conductivity of n-type doped SWCNT films measured at different ambient conditions and temperatures. Reproduced with permission from Ref. [275], © American Chemical Society 2019.

Figure 18 Thermoelectric power generators with p-doped single-walled carbon nanotubes. (a) Photograph of a homemade testing platform for measuring the thermoelectric component. (b) Scheme of the flexible thermoelectric device. (c) Demonstration of temperature difference for providing a work condition. (d) The powers (left) and dispersion (right) of p-doped single-walled carbon nanotubes. (e) Photograph of a thermoelectric generator with the patterning of p-doped SWCNT ink over Ag electrodes. (f) Illustration of the architecture of SWCNTs based thermoelectric generator. (g) The performances of the thermoelectric generator with output voltage and power. Reproduced with permission from Ref. [276], © American Chemical Society 2020.

nanotubes in the IoT based sensors, high frequency transistors for 5G communications, and the perception-action for artificial intelligence [287]. To start with, the progress in CNT based sensory is summarized as follows for the five sensing types (Fig. 19).

Figure 19 The prospect of carbon nanotubes in system integration for IoT sensors and artificial intelligence.

First, CNT based E-eye, i.e., artificial retina, could detect the light intensity by photocurrent conversion. The sight sensory may require the optical [286] and photoelectronic devices [288, 289]. Furthermore, the image computing circuitry has been demonstrated with thousands of CNT transistors [290]. Another emerging trend for imaging is based on CNT terahertz camera [291–293]. Also, CNT transistor based artificial synapses serve as good neuromorphic device by generating photocurrent under light-stimulation [294].

Second, the electronic ear can be mimicked by the CNT based eardrum from the piezoresistance device [295]. The sensing mechanism is to convert the sound waves into the voltage signals based on the sound-vibration-caused-change in contact resistance of two stacking CNT/PDMS pyramids. Furthermore, device arrays could be assembled for pixel resolution recognition of speech pattern [296].

Third, the E-nose depicted the sensors for the recognition of flavors by chemoresistive devices [297, 298]. Individual gases could be recognized by CNT based sensors, including ammonia [299], NO₂ [300, 301], carbonyl chloride [302], and toluene [303]. In the CNT transistors functionalized by different ionic liquid, volatile compounds in the breath [304] can be distinguished by the types of decorating materials [305]. The various gases could be detected and classified, i.e., sensing and computing, with an integrated system [170] including the CNT transistor-based gas sensors, CNT logic gate circuits, resistive random-access memory, and interconnect. Indeed, clove essential oil could be recognized with CNT based electronic nose [306].

Fourth, the E-tongue could be achieved by the fusion of chemical sensing of several substances in aqueous solution, e.g., the glucose [307] and tea taste [308]. The chemical sensors can be fabricated for flexible integrated circuits [95] and customized healthcare [309]. Then, the production cost could be reduced by printed electronics processing [310].

Fifth, the E-skins are fabricated with the tactile sensors with extraordinary pixel resolutions. Indeed, the CNTs [20, 311] and

related composites [312] guarantee the high sensitivity of electronic skins [313] and fast response rate [314]. In addition, the fluorescence of CNTs leads to the detection of biomarker molecules [315–317]. Besides, the acceleration rate could be transduced into output power, which may provide feedback to the autopilot [318]. After the perception of surrounding environment, we humans are able to communicate with the world by speaking and body language. Indeed, the loudspeakers have been demonstrated based on the thermoacoustic chip of carbon nanotube devices [319–322]. Also, the actuation based on CNT/elastic polymer-based composite has been reported [323, 324]. Besides, neural interfacing [325–327] bridges the brains and the artificial sensory devices through microelectrode arrays [328–330] and neuronal signaling [331] and neural recording [332, 333].

Future opportunities exist in the highly integration of devices and components for these sensory, i.e., fusion of several sensory [334, 335], distributed networking [336, 337] and response upon stimulus [338] as well as wireless communication modules.

Besides, the light-weight distributed sensory can be guaranteed by various types of power sources, such as stretchable energy storage devices [339, 340], passive powering by RFID tags [341, 342], and self-powering by nanogenerators [343–346]. In addition, the data processing brings the opportunities for CNTs based high frequency transistors, logic gate circuits, and central processors. Moreover, the commercialization [347] are being testified based on these IoT oriented devices.

The thermodynamics and kinetics still require the first-principle calculation [29] and experimental extraction [348, 349], and growth rate [350] during the CVD growth. The investigation of the intrinsic properties is still ongoing, such as superlong and super-durable mechanical performances, small diameter [351], electronic mobility, and electrical conductivity.

Precisely controllable synthesis of the carbon nanotubes with demanded optical band gap [28, 203, 352, 353], e.g., (*n*, *n*–1) chirality, is still intensively investigated, which are suitable for the NIR biomedical imaging as well as next-generation electronics [354]. To this end, various strategies are being continuously updated, e.g., catalyst anchoring [355–357] and engineering [109, 358, 359], and CNT arrays alignment [36, 105, 360]. The enrichment of semiconducting CNTs remains important goals through metal carbide catalyst [361, 362], chirality cloning [363–365], and selective suppression of metallic CNTs [366]. The high-resolution microscopy has been combined with heating stage and gas ambience for *in-situ* observation of the growth mechanism [85, 123, 367] and etching process [368].

New trends emerge for the synthesis of carbon nanotubes based 1D van der Waals heterostructures [369, 370]. Indeed, the CNTs/h-BN heterostructure [31, 371–373] facilitates the exploration of the exciton dynamics [374] as well as theoretical limit of the ballistic transport. Also, the charge transfer mechanism remains unclear at the CNTs/zero-dimensional materials interface [375]. Indeed, the radical stacking with inorganic nanotubes [376–378] and nanocrystals [379, 380] and sub-nanometric materials [381–383] could lead to heterostructures that may exhibit interesting physical phenomena.

6 Concluding remarks

In this review, the electronic devices applications of CNTs were summarized, e.g., field-effect transistors, DNA sequencing, gas and organic molecule sensors, and metal ion detection. To start with, the properties and preparation methods are briefly delivered. Then, the IoT have brought up the gorgeous synthesis approaches aimed at the chirality and diameter control of CNTs. Subsequently, the

energy conversion by CNTs was briefly introduced with the piezo-electronics based electronic skin, and thermoelectric power generation. Eventually, the potentials and promises of CNTs have been proposed for the IoT and artificial intelligence.

Acknowledgements

The authors acknowledge the financial funds of the National Key Research and Development Program of China (No. 2017YFB0405400) and the Project of “20 items of University” of Jinan (No. 2018GXRC031). W. J. Z thanks the National Natural Science Foundation of China (No. 52022037) and Taishan Scholars Project Special Funds (No. tsqn201812083). J. B. P. shows his gratitude to the National Natural Science Foundation of China (No. 51802116) and the Natural Science Foundation of Shandong Province, China (No. ZR2019BEM040). F. Y. was supported by the National Natural Science Foundation of China (Nos. 52002165, 92161124, and 21631002), the National Key Research and Development Program of China (No. 2021YFA0717400), Shenzhen Basic Research Project (Nos. JCYJ20210324104808022 and JCYJ20170817113121505), Beijing National Laboratory for Molecular Sciences (No. BNLM202013), Guangdong Provincial Natural Science Foundation (No. 2021A1515010229), Innovation Project for Guangdong Provincial Department of Education (No. 2019KTSCX155), and Guangdong Provincial Key Laboratory of Catalysis (No. 2020B121201002). M. H. R. thanks the National Science Foundation China (No. 52071225), the National Science Center and the Czech Republic under the ERDF program “Institute of Environmental Technology-Excellent Research” (No. CZ.02.1.01/0.0/0.0/16_019/0000853) and the Sino-German Research Institute for support (Project No. GZ 1400).

Funding note: Open Access funding enabled and organized by Projekt DEAL.

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