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REVIEW

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Carbon nanotube-graphene hybrids for soft electronics, sensors, and actuators

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Abstract

Soft devices that are mechanically flexible and stretchable are considered as the building blocks for various applications ranging from wearable devices to robotics. Among the many candidate materials for constructing soft devices, carbon nanomaterials such as carbon nanotubes (CNTs) and graphene have been actively investigated owing to their outstanding characteristics, including their intrinsic flexibility, tunable conductivity, and potential for large-area processing. In particular, hybrids of CNTs and graphene can improve the performance of soft devices and provide them with novel capabilities. In this review, the advances in CNT-graphene hybrid-based soft electrodes, transistors, pressure and strain sensors, and actuators are discussed, highlighting the performance improvements of these devices originating from the synergistic effects of the hybrids of CNT and graphene. The integration of multidimensional heterogeneous carbon nanomaterials is expected to be a promising approach for accelerating the development of high-performance soft devices. Finally, current challenges and future opportunities are summarized, from the processing of hybrid materials to the system-level integration of multiple components.

Keywords: Carbon nanomaterials, Heterogeneous materials, Stretchable electronics, Wearable devices, Electronic skins, Soft robotics, Artificial muscles, Human–machine interfaces

Introduction

Carbon nanomaterials composed of a hexagonal lattice of sp^2-hybridized carbon atoms have been actively investigated for scalable production in various fields of applications [1, 2]. They are classified into one-dimensional (1D) carbon nanotubes (CNTs) and two-dimensional (2D) graphene according to their dimensional shape. CNTs and graphene exhibit excellent and unique properties, making them attractive candidate materials for electronics, displays, transducers, and other devices. In particular, the high elastic modulus, strength, and aspect ratio

of CNTs, along with their high electrical conductivity, allow for mechanically strong and electrically conductive composites with various potential applications [3, 4]. These characteristics of CNTs have also been adopted for microelectromechanical devices by integrating aligned CNTs at specific locations in microstructures [5–10]. The incorporation of CNTs has led to improved performance and reliability in structures and devices. In view of the properties of the CNTs, it was expected and eventually discovered that graphene also exhibits extraordinary mechanical, electrical, thermal, and optical properties [11, 12]. Among the many interesting characteristics of graphene, its high carrier mobility outperforms those of other materials, which has accelerated the development of carbon nanoelectronics [13]. Furthermore, owing to their atomic diameter or thickness, both CNTs and graphene can be key components of transparent devices in the form of a monolayer while providing high mechanical compliance [14, 15]. These features have advanced the development of soft and transparent electronics, sensors,

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and actuators, based on 1D CNTs or 2D graphene as building blocks.

Manufacturing processes have been continuously developed to scale up the excellent properties of individual CNTs and graphene in functional devices. With progress in synthesis, integration, and transfer processes, carbon nanomaterial-based devices have achieved a higher degree of design freedom, including in the selection of substrate materials. Thus, conventional rigid substrates can be replaced by elastomeric substrates, such as rubber and many polymers, leading to the active development of wearable electronics and sensors that can be conformally mounted on curved surfaces based on the low flexural rigidity of thin and soft substrates [16–19]. In addition, the incorporation of CNTs and graphene into polymeric materials has enabled the realization of soft actuators requiring large deformations by electrical and chemical stimuli [20]. Soft sensors and actuators composed of carbon nanomaterials are expected to be applied in future wearable, implantable devices, and human–machine interfaces. The performance of soft devices can be further improved by heterogeneously integrating multidimensional nanomaterials, as previously demonstrated by combining 1D CNTs and 2D graphene for electronics, optoelectronics, and electromechanical and electrochemical devices [21–25]. CNTs and graphene have intimate lattice structures beneficial for minimizing contact resistance and reinforcing mechanical strength when forming heterostructures [26]. As such, in addition to their own properties, hybrids of 1D CNTs and 2D graphene could synergistically reach improvements in the properties and performance of target devices otherwise unachievable by homogeneous materials. Therefore, CNT-graphene hybrids are an attractive choice for realizing flexible, stretchable, and transparent devices, thereby providing new functionalities and leading to better performance and reliability relative to conventional devices.
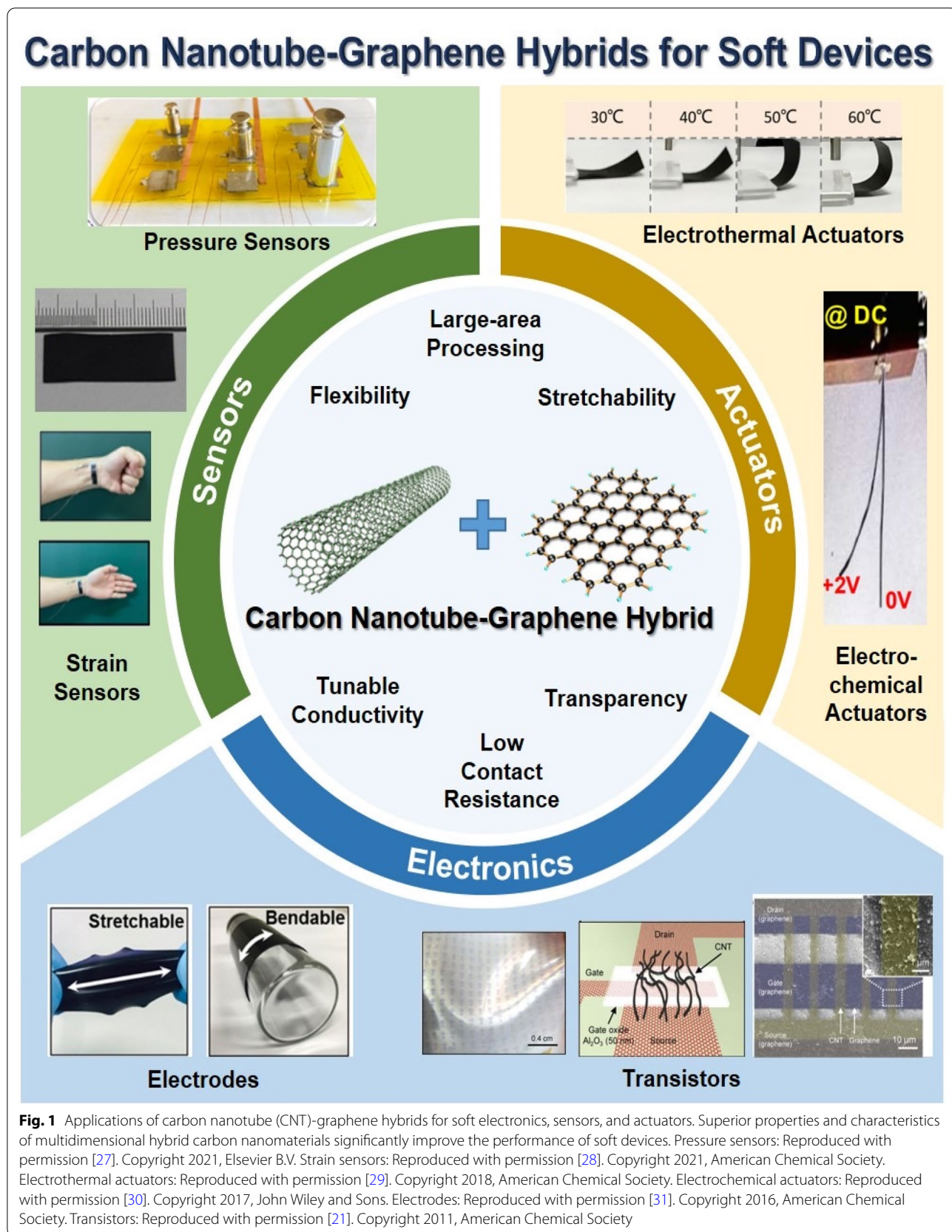
This review covers the progress in CNT-graphene hybrid-based soft electronics, sensors, and actuators, highlighting the merits of the heterogeneous integration of carbon nanomaterials in terms of device performance (Fig. 1). As the performance improvement depends on the characteristics of the CNT-graphene hybrid, the properties of a CNT-graphene hybrid film are discussed in addition to their synergistic effects as binary reinforcing fillers for elastomeric composites. Further, we review flexible and stretchable electronic devices composed of CNT-graphene, which can be integrated to operate and control soft sensors and actuators. In addition, CNT-graphene hybrids for soft physical sensors for detecting changes in pressure and strain are discussed, focusing on improvements in the sensitivity and sensing range. Finally, applications for CNT-graphene composites and networks as core components in electrochemical and electrothermal soft actuators are discussed in detail. We conclude the review with current challenges and future perspectives in the field of soft devices potentially consisting of other combinations of nanomaterials, as well as carbon nanomaterial hybrids.

Carbon nanotube (CNT)-graphene-reinforced soft structures

The superior material properties of CNTs and graphene, such as their high Young's modulus and tensile strength, make them favorable building blocks for various electromechanical applications. Their broad structural applications include thin films on soft substrates and reinforced polymer composites for various types of soft electronics, sensors, and actuators. The mechanical properties of CNT-based structures have been experimentally measured in many studies. The experimentally measured Young's modulus of CNT-family materials ranges from 4.7 GPa to 1 TPa, depending on the size scale of the structured material and measurement methods [32, 33]. The tensile strengths of these CNT-based structured materials range from 71 MPa to 200 GPa. Extraordinary mechanical properties are generally obtained by using pristine individual nanoscale specimens. However, for scaled-up macrostructures allowing for practical and manufacturable usage of the CNTs, the material properties are generally one to two orders of magnitude smaller. This is mainly because the characteristics of the interconnections and microstructures between the materials dominate the superior intrinsic properties as the material is scaled up into a macro form, resulting in deteriorated mechanical properties in the overall structure on a macro scale. Efforts have been made to overcome this issue, and certain techniques have been developed to enhance material properties on a macro scale [34, 35]. Graphene has attracted significant interest for various applications, and its material properties have been widely tested in many structural forms. An experimentally measured Young's modulus of 1 TPa as measured by nanoindentation in an atomic force microscopy setup was reported for pristine monolayer graphene [36]. Nonetheless, the Young's modulus of graphene can decrease to as low as 31.7 GPa for practical macro-scale bulk material configurations, such as in paper forms [37]. Experimental reports have shown that the tensile strength of graphene can range between 223 and 130 GPa [36, 38].

To date, there have been many academic explorations and technical efforts concerning CNT and graphene reinforcements, aiming to find superior structures for soft devices (electronics, sensors, and actuators). These attempts can be classified into many nano and microstructures, but CNT-graphene hybrid thin films and

Fig. 1 Applications of carbon nanotube (CNT)-graphene hybrids for soft electronics, sensors, and actuators. Superior properties and characteristics of multidimensional hybrid carbon nanomaterials significantly improve the performance of soft devices. Pressure sensors: Reproduced with permission [27]. Copyright 2021, Elsevier B.V. Strain sensors: Reproduced with permission [28]. Copyright 2021, American Chemical Society. Electrothermal actuators: Reproduced with permission [29]. Copyright 2018, American Chemical Society. Electrochemical actuators: Reproduced with permission [30]. Copyright 2017, John Wiley and Sons. Electrodes: Reproduced with permission [31]. Copyright 2016, American Chemical Society. Transistors: Reproduced with permission [21]. Copyright 2011, American Chemical Society

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reinforced composite matrices with CNT-graphene fillers [39] are the two most widely explored structural materials. CNT-graphene hybrid films have been widely tested to explore, verify, and utilize their synergetic size and geometrical effects. The Young's modulus of a CNT-graphene hybrid film has been reported as much higher than that of a super-aligned CNT film. For example, it was reported that a Young's modulus achieved by a CNT-graphene hybrid film was larger by an order of magnitude (2.34 GPa) than that of a super-aligned CNT film (0.35 GPa) [40]. Graphene fillers generally offer superior mechanical strength to a host material relative to CNT nanofillers [41]. The synergetic effects of CNT-graphene hybrid nanofillers on the mechanical characteristics of composite materials have recently been explored [42]. Efforts have been made to enhance mechanical properties by combining CNTs and graphene nanoplatelets and optimizing the mixing ratio [43]. Others have reported that adding CNT-graphene hybrids as fillers to natural rubber almost doubles its tensile strength [44].
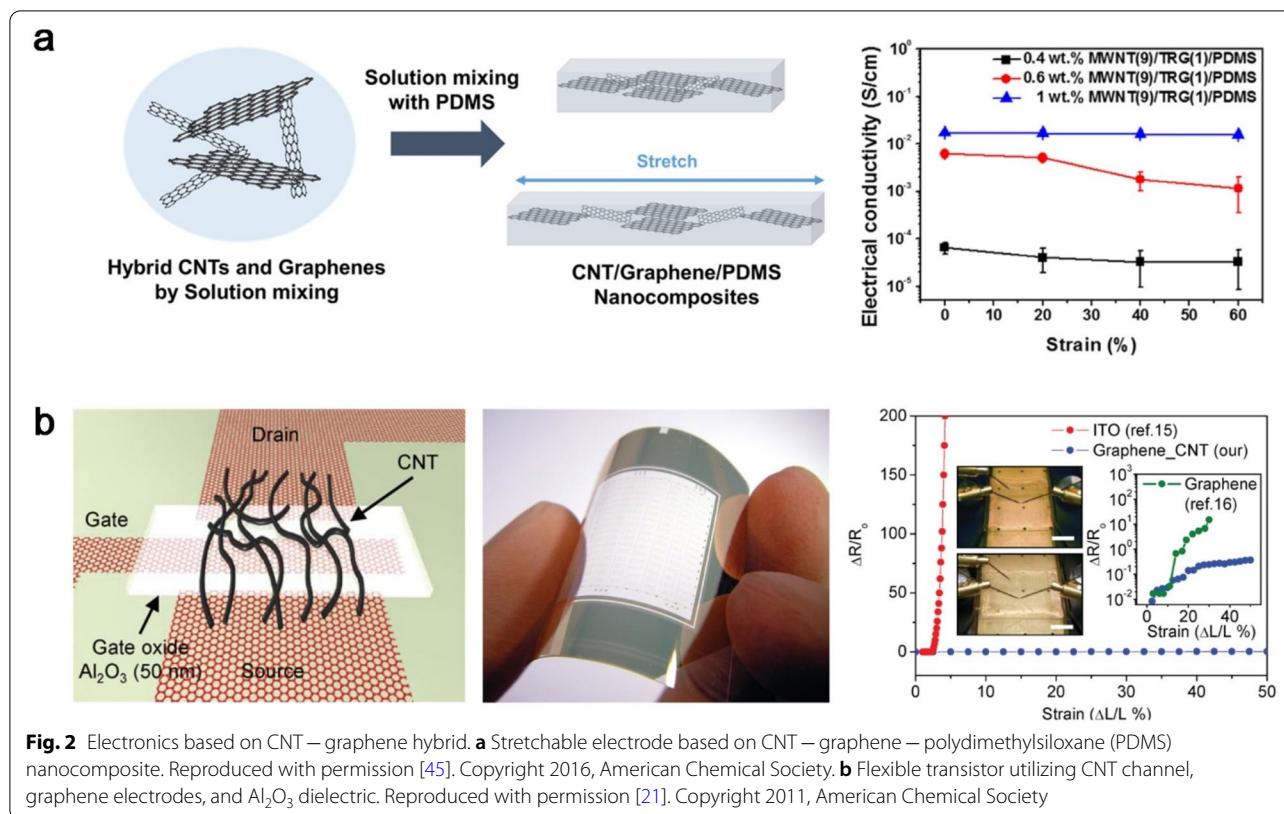
Soft electronics

Electronic components are key parts for the operation of transducers, such as those for signal transmission, power supply, and control. To realize soft sensors and actuators, the mechanical flexibility/stretchability of the electronic materials should be considered along with their electrical performance. Metal and doped Si thin films (the most widely used electronic materials) are unsuitable for such devices, owing to their insufficient flexibility. Thus, the development of flexible and stretchable electronic materials has become an important research topic in recent decades. With recent advances in production techniques, many studies have been performed with the goal of utilizing CNTs and graphene as conductors or semiconducting channels for soft electronics, owing to their remarkable electronic and mechanical properties. Stretchable electrodes based on monolayer graphene with a high fracture strain resistance and low sheet resistance have been proposed [14]. Likewise, the high mobility and on–off ratio of semiconducting CNTs make them an excellent active-channel material for soft electronics [46]. More recently, flexible and stretchable electronic materials utilizing CNT−graphene hybrids have been developed, as the 1D−2D heterostructure provides novel characteristics such as van der Waals interaction, reinforced mechanical strength, and formation of an efficient percolating network enabling significant improvements in the electrical and mechanical properties [23, 24, 47].

One promising application of the CNT−graphene hybrid is as a flexible and stretchable electrode. Flexible and stretchable electrodes must retain conductivity at a substantial strain level without significant changes in their electrical transport properties. The combination of 1D metallic CNTs and 2D graphene is ideal for achieving these attributes; thus, extensive research has been conducted on developing flexible and stretchable electrodes based on CNT–graphene hybrids [45, 48–52]. Placing both CNTs and graphene on the surfaces of plastics or polymer substrates is one strategy for exploiting hybrid structures as electrodes. Nguyen et al. developed a CNT–graphene hybrid thin film for flexible electrodes [49]. The graphene film was grown via chemical vapor deposition (CVD), and thin networks of CNTs were synthesized on the graphene surface using the same CVD approach. The CNT−graphene hybrid film exhibited a much lower sheet resistance (420 Ω sq^{-1}) than a graphene film (2.15 kΩ sq^{-1}). The considerable improvement in electrical conductivity was attributed to the formation of a CNT network for connecting the gaps between the graphene sheets and the low contact resistance via the π−π interactions between them. The mechano-electrical properties were also investigated. The results showed that the deviation in the resistance of the hybrid film on the polymer substrate was only 7.2% after 100 bending cycles at a bending angle of 150°, owing to the CNTs extending in a bent state without cracking. Another type of CNT-graphene hybrid electrode is a CNT-graphene-elastomer nanocomposite. As shown in Fig. 2a, stretchable electrodes based on CNT−graphene−polydimethylsiloxane (PDMS) nanocomposites were developed via the solution mixing of CNT−graphene fillers with a PDMS matrix, and then the electrical conductivity and stretchability were investigated with respect to the fractions of the filler [45]. The synergistic effects of the 1D−2D interconnections prevented CNT aggregation and graphene restacking and reduced the contact resistance between them, resulting in the higher electrical conductivity of the nanocomposite with a filler fraction of 0.6% (6.17×10^{-3} S cm^{-1}) compared to CNT-PDMS or graphene−PDMS composites ($< 1.85 \times 10^{-3}$ S cm^{-1}). In addition, the nanocomposite retained its electrical conductivity when strained up to 60%, owing to the high aspect ratio of the CNTs and their homogeneous dispersion with the graphene.

Flexible and stretchable transistors based on CNT−graphene hybrids have also been actively studied [21, 53–58]. The transistors operating in signal processing and control circuits are an essential part of electronic devices, and all components of transistors must be flexible or stretchable to be fully functional for soft electronics. A widely used type of transistor is the field-effect transistor (FET), which comprises active channels, electrodes, and gate dielectrics. To date, flexible and stretchable FETs employing various semiconducting and metallic nanomaterials have been explored. FETs

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Fig. 2 Electronics based on CNT − graphene hybrid. **a** Stretchable electrode based on CNT − graphene − polydimethylsiloxane (PDMS) nanocomposite. Reproduced with permission [45]. Copyright 2016, American Chemical Society. **b** Flexible transistor utilizing CNT channel, graphene electrodes, and Al_2O_3 dielectric. Reproduced with permission [21]. Copyright 2011, American Chemical Society

employing CNT–graphene hybrids have several unique advantages over other nanomaterial-based FETs. Yu et al. developed nanocarbon-based transistors on flexible substrates and reported their outstanding characteristics [21]. The device was fabricated using a CNT network channel, monolayer graphene electrodes, and an Al_2O_3 dielectric (Fig. 2b). The authors revealed that the contact resistance of CNT-graphene is 100 times lower than that of CNT − Au. This was owing to the smaller difference in the work function of CNT-graphene, which allowed the CNT-graphene transistor to exhibit 20-times-higher mobility (81 cm^2 V^{-1} s^{-1}). No appreciable degradation was observed in the conductivity during the bending test, and the change in resistance during stretching (50% strain) was 36%. The change was considerably smaller than that of indium tin oxide (2000% at 5% strain) and graphene (200% at 30% strain). A few years later, stretchable CNT − graphene transistors were developed by the same group [54]. In that study, a geometrically wrinkled Al_2O_3 layer containing effective built-in air gaps was used to achieve stretchability. A wrinkled structure that can be stretched provides stability to the dielectric under tensile strain, i.e., different from a flat structure. Therefore, the transistors were successfully operated under a high strain (20%) without noticeable leakage current or physical degradation. To validate the potential utility

of the transistors, they were mounted on various media such as human skin, aluminum foil, and a plastic heart. These results imply that the CNT-graphene transistor is a promising candidate for the operation of soft sensors and actuators.

Soft sensors

Wearable electronics, including artificial electronic skins, have attracted considerable attention for various applications, such as in human motion detection and physiological monitoring [62–67]. Flexible and stretchable physical sensors are key components for such applications, and mechanical flexibility/stretchability, high sensitivity, fast response time, and repeatability are considered as important requirements [68]. However, conventional physical sensors based on thin films and nanowires exhibit insufficient performance for practical applications, such as by having a narrow sensing range and/or low sensitivity [69–72]. In recent years, CNTs and graphene have been introduced as sensing materials to fabricate flexible physical sensors with a wide sensing range and high sensitivity [73–84]. CNT-based physical sensors have high flexibility but suffer from poor sensitivity. In contrast, flexible physical sensors using graphene exhibit strength in sensitivity, although the sensing range is limited. Thus, a CNT-graphene hybrid structure can provide

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a synergistic electrical network induced by the interconnections between the graphene and CNTs, resulting in superior stretchability and high sensitivity [85–90].

Flexible physical sensors based on CNT-graphene hybrids are classified as pressure and strain sensors. Jian et al. demonstrated a flexible pressure sensor using aligned CNTs and graphene hybrid materials [59]. In their study, aligned CNTs were applied to form networks with graphene, resulting in enhanced electrical

conductivity (Fig. 3a). The aligned CNTs were prepared from vertically synthesized CNT bundles using a conventional CVD process, and then graphene was grown from a copper (Cu) foil on the prepared aligned CNT films. The fabricated aligned CNT-graphene films showed a good optical transmittance of 81.4% (550 nm) and high sensitivity of 19.8 kPa^{-1} (< 0.3 kPa) owing to increased conductance from the alignment in the same orientation, providing a very low detection pressure level (0.6 Pa)
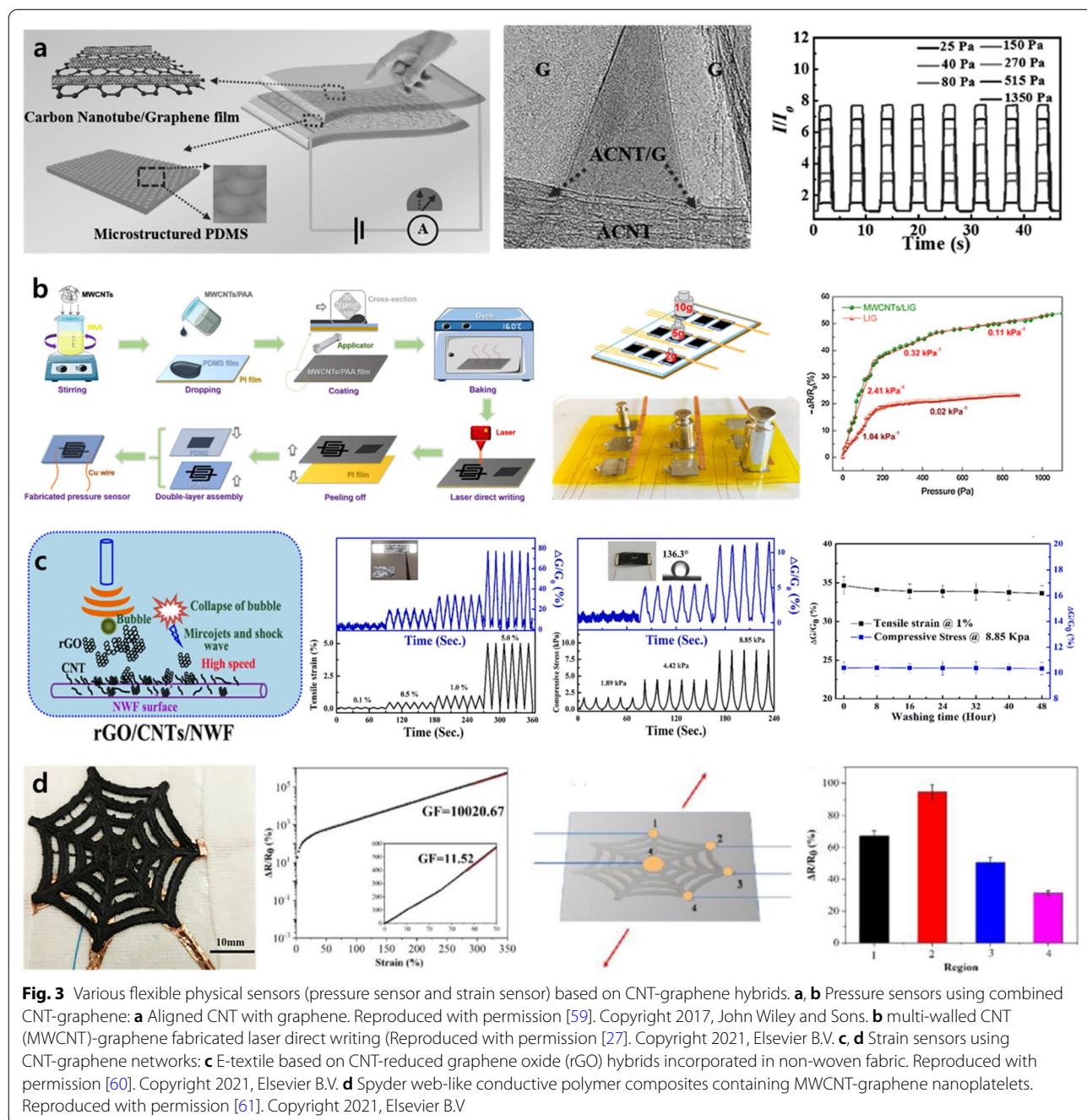
Fig. 3 Various flexible physical sensors (pressure sensor and strain sensor) based on CNT-graphene hybrids. **a, b** Pressure sensors using combined CNT-graphene: **a** Aligned CNT with graphene. Reproduced with permission [59]. Copyright 2017, John Wiley and Sons. **b** multi-walled CNT (MWCNT)-graphene fabricated laser direct writing (Reproduced with permission [27]. Copyright 2021, Elsevier B.V. **c, d** Strain sensors using CNT-graphene networks: **c** E-textile based on CNT-reduced graphene oxide (rGO) hybrids incorporated in non-woven fabric. Reproduced with permission [60]. Copyright 2021, Elsevier B.V. **d** Spyder web-like conductive polymer composites containing MWCNT-graphene nanoplatelets. Reproduced with permission [61]. Copyright 2021, Elsevier B.V

and superior stability over 35,000 cycles. The CNT-graphene hybrid structure has emerged as a very attractive material for pressure sensors; nevertheless, providing a cost-effective and simple method for fabricating such materials remains a challenging issue. To overcome this difficulty, Zhao et al. reported asymmetric pressure sensors based on multiwalled CNT (MWCNT)-graphene fabricated using direct laser writing [27]. As shown in Fig. 3b, a combined structure of MWCNTs and graphene was formed using a laser direct-writing technique, providing convenience, high reliability, good efficiency, and the ability for mass production. A MWCNT-embedded polyimide film was graphitized into MWCNT-embedded laser-induced graphene using the laser direct-writing method. The fabricated sensor exhibited a high sensitivity of 2.41 kPa^{-1}, low detection limit of 1.2 Pa, and fast response recovery time of 2 ms. Tran et al. introduced a quantum resistive pressure sensor comprising 3D-sprayed CNT-graphene, and showed a very large pressure sensitivity range (0–8 MPa) [91]. Thus, these flexible pressure sensors have shown good potential for applications such as artificial skins.

In addition to flexible pressure sensors, CNT-graphene hybrid materials have been widely used as flexible wearable strain sensors. Yao et al. reported an electronic textile-based wearable and washable strain sensor composed of a CNT-reduced graphene oxide (rGO) hybrid incorporated in a non-woven fabric [60]. Many studies combining CNT-graphene and textiles have been proposed for electronic textile sensors, but these methods have several drawbacks (e.g., requiring additional harmful treatments or specific experimental conditions, and/or showing weak adhesion between the CNT-graphene and textiles). Therefore, a washable, durable, and wearable e-textile sensor based on CNT-rGO was fabricated using an ultrasonic nano-soldering method, resulting in strong adhesion between the rGO-CNT and non-woven fabric (Fig. 3c). Previous flexible strain sensors have generally been used to measure unidirectional strain, thus showing limitations in application. Flexible strain sensors usable for various applications require multidirectional sensing, high sensitivity, and a wide detection range. Chen et al. demonstrated a spider web-like 3D-printed flexible strain sensor constructed using conductive polymer composites, including MWCNT-graphene nanoplatelets [61]. This sensor exhibited a large strain range (0%–300%), good linearity, a gauge factor of over 1000, and good stability (Fig. 3d). Li et al. reported graphene nanoplatelets and CNTs forming hierarchical hybrid networks fabricated by spray coating. These were demonstrated in flexible wearable strain sensors by applying them to a human finger and front neck area [92]. In general, the enhanced sensitivity of the graphene from introducing the CNTs

allows for the detection of subtle motions. There are other strain sensors that use porous PDMS with CNT-graphene as a hybrid filler to control the sensitivity and stretchability using the Soxhlet extraction method [28]. Thus, strain sensors based on CNT-graphene-PDMS show promise for wearable smart electronics.

Soft actuators

Soft actuators play a critical role in soft robotics and bionics, with broader impacts in wearable and flexible electronics, robotic exoskeletons and prosthetics, artificial organs, and implantable medical devices. Along with advances in actuation mechanisms, innovative materials with unique physical and chemical characteristics have been vigorously explored. Carbon-based nanomaterials are known for their outstanding electrical and mechanical properties, high surface-to-volume ratios, and light specific weights; these aspects are critical for building high-performance soft actuators. CNT/graphene hybrid composites have been reported to offer the synergistic enhancement of such material properties for even larger actuation strains, faster response times, and greater stability and durability.

Ion polymer metal composite (IPMC) actuators are a type of electro-chemo-mechanical actuator that consist of an ionic polymer layer coated with conductive layers on both sides (Fig. 4a). Ion migration and redistribution occur when a potential is applied to the conductive layers. This induces an asymmetric volume expansion of the conductive layers and consequent structural deformation. Because CNT/graphene composites have advantages over metals when used as conductive layers, different structural designs and fabrication processes have been reported. Yang et al. fabricated ionic polymer actuators using an electrospray-coated MWCNT/graphene mixture with carboxymethyl cellulose on a Nafion membrane [95]. Compared to conventional IPMC actuators with platinum electrodes, the MWCNT/graphene actuators achieved a larger actuation displacement of ± 2.33 mm. The increasing ratio of graphene in the MWCNT/graphene composites led to decreasing in the actuation displacement and increasing in the fundamental natural frequency owing to the increased electrical resistance and mechanical stiffness, respectively. Lu et al. revealed that an rGO/MWCNT hybrid had a porous structure and exhibited more efficient electrochemical charging and discharging characteristics (Fig. 4b) [93]. An electrochemical actuator fabricated using the rGO/MWCNT hybrid in the electrodes demonstrated a larger bending displacement than those with only rGO or MWCNTs at frequencies ranging from ~ 0.1 to 1 Hz, with no signs of significant performance degradation even after 10^6 cycles. To
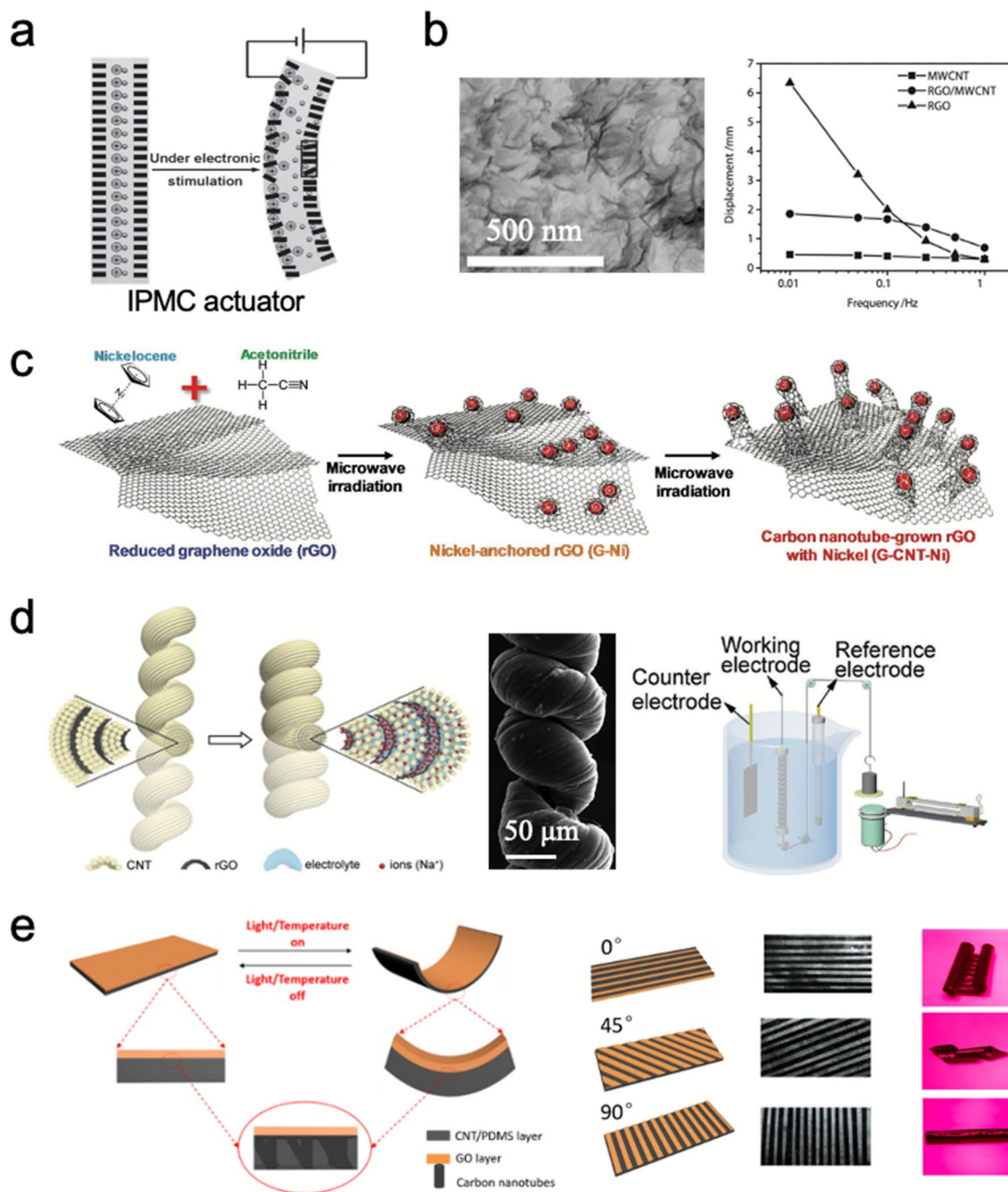
Fig. 4 a Illustration of the working mechanism of an ion polymer metal composite (IPMC) actuator. Reproduced with permission [20]. Copyright 2014, John Wiley and Sons. **b** Scanning electron microscope (SEM) image of rGO/MWCNT hybrid layer showing porous surface structures and displacement of bending actuation with different conductive materials over the frequency range from 0.01 to 1 Hz. Reproduced with permission [93]. Copyright 2012, John Wiley and Sons. **c** Fabrication process of graphene-carbon nanotube-nickel (G-CNT-Ni) heterostructures. Reproduced with permission [30]. Copyright 2017, John Wiley and Sons. **d** Coiled CNT/rGO hybrid yarns: a schematic diagram of the working mechanism, a SEM image showing the structure of the yarn, and an illustration of the testing setup. Reproduced with permission [94]. Copyright 2018, John Wiley and Sons. **e** Illustration of the bending and recovery actuation of the graphene oxide (GO)-CNT/PDMS bilayer membrane in response to light and temperature stimulations and the controlled bending behaviors with the aligned GO patterns in different directions. Reproduced with permission [29]. Copyright 2018, American Chemical Society

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further increase the specific surface areas of the conductive layers to promote faster ion diffusion and higher electrochemical responses for improved actuation performance, three-dimensional rGO/MWCNT-based hierarchical nanostructures have been synthesized and adapted for ionic actuators. rGO/MWCNT hybrid films with vertically aligned NiO nanowalls on the surface have achieved large displacements and fast responses (18.4 mm bending peak-to-peak deformation at 10 Hz), with long-term stability over 5×10^5 cycles [96]. rGO sheets with vertically grown MWCNTs with Ni dots at the tips (Fig. 4c) have also shown significant performance enhancements, with a bending strain, blocking force, and cyclic duration of 6.59 mm, 4.53 mN, and 4 h, respectively [30].

CNT artificial muscles are another type of electrochemical actuator able to generate torsional or tensile motions with high energy conversion efficiency and controllability. CNT yarns, which are the main building blocks of CNT artificial muscles, can be fabricated by twist-spinning electrolyte-filled CNTs. Similar to IPMC actuators, ions migrate and fill the electrical double layer at the applied voltages (Fig. 4d). This causes a volumetric expansion of the yarns, resulting in untwisting and contraction. The capacitance of CNT yarns is one of the critical factors determining the actuation performance, and it can be increased by the addition of graphene. Qiao et al. fabricated coiled CNT/rGO hybrid yarns using a bioscrolling method. The yarns were driven in an aqueous inorganic electrolyte with a maximum tensile contraction of 8.1%, contractile stress of 16 MPa, and work capacity of 236 J/kg [94]. Similarly, Hyeon et al. fabricated coiled graphene/CNT yarns containing 80 wt% graphene and recorded an order-of-magnitude larger tensile stroke and work capacity of ~19% and 2.6 J/g, respectively [97].

Carbonaceous nanomaterials and their composites are promising candidates for flexible heating elements, owing to their excellent electrical, thermal, and mechanical properties. Wang et al. developed simple electrothermal actuators by integrating CNTs and graphene oxides (GOs) into a polyvinyl alcohol matrix to spin composite fibers [98]. When a voltage was applied to the fiber, the single-walled CNT/GO networks generated heat, and the fibers underwent thermal expansion. Another type of thermal actuator consisting of bilayer membranes with different thermal expansion coefficients can generate more sophisticated motions (Fig. 4e) [29]. A GO-CNT/PDMS bilayer structure was obtained by spin coating a CNT/PDMS mixture onto a GO layer. When exposed to light or heat, the membrane bent toward the GO layer and was also responsive to moisture changes. Bending was controlled by patterning the GO layer into ordered strips in different directions, and various soft robotic applications, such as smart tweezers and tendrils, were demonstrated.

Conclusion

Soft electronics, sensors, and actuators are expected to be essential components in robotics, wearable devices, and implantable devices, as these devices can be integrated with arbitrary structures and surfaces based on their flexibility and stretchability. Among the many materials and structures developed so far, hybrids of multidimensional carbon nanomaterials have shown great potential, given their compelling properties suitable for soft devices. The integration of carbon nanomaterials into elastomers would also provide new functionalities compared to conventional metals or semiconductors, such as transparency. Furthermore, hybrids of CNT and graphene could complement each other, resulting in improved device performance, e.g., in regards to sensitivity, sensing range, and actuation displacement. Thus, devices based on CNT-graphene hybrids can outperform others comprising homogeneous materials. Hybrids of nanomaterials are not limited to CNT and graphene; other combinations of multidimensional materials such as 1D metallic nanowires and 2D semiconducting transition metal dichalcogenides can be designed for new device architectures and functionalities.

In addition to the considerable achievements in hybrid carbon nanomaterial-based soft devices, several challenges and opportunities remain for future work. From a materials perspective, controlling defects and the number of walls or layers of CNTs and graphene during scalable and low-cost production may expand the applicability of carbon nanomaterials. Moreover, providing effective and simple purification between metallic and semiconducting CNTs would enhance the performance of carbon-based transistors and be useful for other applications, such as in transparent conductors and electromagnetic wave shielding. In terms of the performance of soft devices, improved structural and electronic design would enable a high-density sensor array for measuring and processing multiple stimuli without crosstalk, and an actuator for generating large displacements and forces with high resolution. To realize a standalone soft device, the development of manufacturing and packaging processes for integrating all components (such as electronics, sensors, and actuators) into a single system is another important challenge. Flexible and stretchable energy storage devices and wireless communication modules can also be integrated with such soft systems. Overall, high-performance, reliable, and biocompatible soft devices composed of carbon nanomaterial hybrids will be available in many fields through interdisciplinary efforts to address these challenges.

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Abbreviations
1D: One-dimensional; 2D: Two-dimensional; CNT: Carbon nanotube; CVD: Chemical vapor deposition; PDMS: Polydimethylsiloxane; FET: Field-effect transistor; MWCNT: Multi-walled carbon nanotube; GO: Graphene oxide; rGO: Reduced graphene oxide; IPMC: Ion polymer metal composite.

Declarations

Competing interests
The authors declare that they have no competing interests.

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