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Carbon Nanotube Electroactive Polymer Materials: Opportunities and Challenges

Liangti Qu, Qiang Peng, Liming Dai, Geoffrey M. Spinks, Gordon G. Wallace, and Ray H. Baughman

Abstract

Carbon nanotubes (CNTs) with macroscopically ordered structures (e.g., aligned or patterned mats, fibers, and sheets) and associated large surface areas have proven promising as new CNT electroactive polymer materials (CNT-EAPs) for the development of advanced chemical and biological sensors. The functionalization of CNTs with many biological species to gain specific surface characteristics and to facilitate electron transfer to and from them for chemical- and bio-sensing applications is an area of intense research activity.

Mechanical actuation generated by CNT-EAPs is another exciting electroactive function provided by these versatile materials. Controlled mechanical deformation for actuation has been demonstrated in CNT mats, fibers, sheets, and individual nanotubes. This article summarizes the current status and technological challenges for the development of electrochemical sensors and electromechanical actuators based on carbon nanotube electroactive materials.

Introduction

Carbon has long been known to exist in three forms: amorphous carbon, graphite, and diamond.1 The Nobel-Prize-winning discovery of buckminsterfullerene C_{60} in 1985 created an entirely new branch of carbon chemistry.^{2,3} The subsequent discovery of carbon nanotubes (CNTs, Figure 1a) in 1991 opened a new era in materials science and nanotechnology.⁴ As can be seen in Figure 1b, a CNT may be conceptually viewed as a graphite sheet that is rolled into a nanoscale tube form as a singlewalled carbon nanotube (SWNT), or as a multiwalled carbon nanotube (MWNT) with additional graphene coaxial tubes around the SWNT core.5

Depending on their diameter and the helicity of the orientation of graphite

rings along the nanotube length, CNTs can exhibit semiconducting or metallic behavior to allow them to be used for many potential applications,⁵ including as new electroactive polymers (CNT-EAPs) in advanced composite systems, single-molecular transistors, molecular computing elements, field-emitting flat-panel displays, sensors, and artificial muscles. In this article, we summarize the current status and the remaining technological challenges for the development of advanced CNT-EAP materials, with a focus on their use as sensors and actuators.

Carbon Nanotube EAPs

Conjugated polymers with alternating single and double carbon-carbon bonds

have been demonstrated to possess interesting optoelectronic properties through a continuous overlapping of π -orbitals along the polymer backbone.⁶⁷ The overlap of π -orbitals opens a channel of charge transport along the polymer backbone to provide tunable electronic properties. These properties make conjugated polymers attractive for use as electroactive polymers (π -EAPs) in various sensing and actuation devices. Having a well-defined conjugated all-carbon one-dimensional structure, CNTs are polymeric in essence and also show interesting electronic, photonic, mechanical, and thermal properties.5 Due to their peculiar nanoscale hollow molecular symmetry, however, CNTs also show unique diameter- and helicity-dependent physiochemical properties useful in various advanced composite materials and devices.8-11

For most of the device-related applications, an aligned or patterned form of CNTs is highly desirable.^{12,13} Various growth and fabrication methods have been developed to produce vertically aligned MWNTs (VA-MWNTs), in a patterned or nonpatterned form, on a large variety of substrates.^{14–22} Figures 2a and 2b show scanning electron microscopy (SEM) images for some of the VA-MWNT micropatterns reported previously.14,21 Although the formation of aligned/micropatterned MWNTs has been known for some years, the syntheses of vertically aligned SWNTs (VA-SWNTs) have only recently been reported, 23-32 as exemplified in Figure 2c.23 Multicomponent micropatterns of VA-MWNTs interposed with nanoparticles, self-assembled non-aligned CNTs, or VA-SWNTs have also been produced (Figure 2d).33-35 Along with the aligned/micropatterned nanotube growth, significant progress has also been achieved in the enrichment of semiconducting or metallic carbon nanotubes by diameter- and helicity-controlled nanotube production³⁶⁻³⁸ or post-production separation.39-50

With the availability of both VA-SWNTs and VA-MWNTs in relatively large quantities, the recent production of CNT yarns and sheets from VA-MWNT forests in scalable quantities by continuous high-rate spinning accomplished by Zhang and others⁵¹⁻⁵⁷ has led to an upsurge of worldwide research on incorporating CNTs into functional composites and devices. One meter of yarn can typically be made in a few minutes. The CNT yarns and sheets (Figure 3) are potentially useful for making transparent and highly conducting electrodes, planar sources of polarized broadband radiation, flexible organic light-emitting diodes, advanced sensors and actuators, and microwave bonding of plastics, to mention



Figure 1. (a) Model of a [10, 5] armchair single-walled carbon nanotube (SWNT). (b) Schematic representation of single-walled/multiwalled nanotube formation by rolling graphene sheet(s). The roll-up chiral vector $C_{\rm h} = na_1 + ma_2$, and the chiral angle is θ . In Figure 1a, n = 10 and m = 5. $C_{\rm h}$ forms the circumference of the nanotube, with its ends meeting each other. If $n - m \neq 3q$ (n, m, and q are integers), the SWNT is semiconducting; if n - m = 3q, the SWNT is metallic.



Figure 2. Scanning electron microscopy (SEM) images of vertically aligned multiwalled nanotube (VA-MWNT) micropatterns prepared by (a) pyrolysis of iron phthalocyanine onto a photopatterned substrate (adapted from Reference 21) and (b) simultaneous growth onto patterned silica surfaces with different tilt angles (adapted from Reference 14); (c) vertically aligned single-walled nanotube (VA-SWNT) forests prepared by the water-assist growth method (adapted from Reference 23); (d), (left) schematic representation of the growth procedure for an interposed VA-SWNT/VA-MWNT pattern with its top view (right, top) and cross-section view (right, bottom). Scale bars, 50 µm. (Adapted from Reference 34.)

but a few applications.⁵² Recent developments of electrochemical sensors and electromechanical actuators based on these tailor-made CNT arrays and associated yarns or sheets are summarized in the subsequent sections.

Carbon Nanotube Sensors

The high surface-to-volume ratio and good electronic properties provided by CNTs are attractive features in the advancement of chemical or biosensors. The tiny size of nanotube sensors implies that they are very energy-efficient, highly sensitive, and could move through the cell membrane without obvious harmful effect on cell function.58-64 The ability of SWNTs to operate as gas sensors was first demonstrated by Kong and co-workers65 based on the change in electrical properties induced by charge transfer with the gas molecules (e.g., NH_3 , NO_2). Subsequently, CNTs have been extensively studied as a new class of sensing element for the development of various chemical and biological sensors.5,58-64 The fundamental principles and recent developments in CNT sensors are discussed with an emphasis on the electrochemical transduction.

CNT Gas Sensors

In 2000, Kong et al.65,66 also reported that semiconducting SWNTs can act as rapid and sensitive chemical sensors at ambient temperature. The conductivity of a single semiconducting SWNT changed rapidly, over several orders of magnitude, upon exposure to nitrogen dioxide or ammonia. While the former interaction increased the hole carriers in the p-type semiconducting SWNT through the charge transfer to the electronaccepting NO₂, enhancing conductance, the latter interaction had an opposite effect. By modifying the device design, the CNT sensors could be applied to many other analytes of electron donating or accepting capabilities (e.g., I_2 , AsF₅, CO) and even to some inert gases (e.g., He, N2, Ar, O2, CO2).65-70

By partially coating VA-MWNT arrays with an appropriate polymer thin film along their tube length,⁷¹ sensors of a high sensitivity, good selectivity, and excellent environmental stability for the detection of a broad class of chemical vapors with low power consumption have been developed recently.72 As schematically shown in Figure 4, the absorption and desorption of chemical vapors by the polymer matrix cause changes in the intertube distance and, hence, the surface resistance across the nanotube film. Simple measurements of the resistance change, therefore, produce the sensor response. With appropriate surface modification for gaining specific interactions with analytes of biological significance,73 the CNT devices could be used



b

Figure 3. SEM images showing (a) small carbon nanotube (CNT) bundles simultaneously pulled and twisted from the CNT forest to form CNT yarns. (b), (left) Photograph of a transparent self-supporting MWNT sheet, and (right) SEM images showing the conversion of a VA-MWNT forest into a single MWNT sheet and a two-dimensionally reinforced structure fabricated by overlaying four nanotube sheets with a 45° shift in orientation between successive sheets (adapted from References 51 and 52).

to detect biological systems in solution. These flexible composite films are also being developed for thermal, optical, and mechanical sensors^{72,74,75} and even as dry adhesives that mimic the gecko foot.^{76–78}

CNT Electrochemical Biosensors

In most of the gas-sensing studies just discussed, the detection of the gases is accomplished by measuring the change in electrical properties of individual CNTs or their assemblies. Consequently, the number of analytes that can be determined using a CNT-based sensor is limited by the transduction mechanisms employed, which involve charge transfer with specific gas molecules. However, CNTs have been demonstrated to enhance the electrochemical reactivity of biomolecules79 and promote the electron-transfer reactions of redox proteins such as myoglobin,80 cytochrome c,81 and microperoxidase MP-11.82 Having been functionalized with various transduction materials, therefore, both nonaligned and aligned CNT electrodes have been studied extensively as electrochemical sensors.

produced from iron VA-MWNTs phthalocyanine (FePc) have been used to make conducting polymer-carbon nanotube (CP-CNT) coaxial nanowires by electrochemically depositing a concentric layer of polypyrrole onto each of the constituent aligned nanotubes⁸³ to immobilize glucose oxidase for monitoring glucose concentration.84 The CP-NT nanowire sensors were demonstrated to be highly sensitive and selective even in the presence of some interfering species.84,85 Other CNT-based glucose sensors, including aligned nanotubes supported by micro-sized carbon fibers,86 have also been reported.87-93

While direct electrochemical detection of DNA hybridization has been demonstrated,⁹⁴ Wang and co-workers⁹⁵ developed a CNT-based dual amplification route for ultrasensitive electrical bioassays of proteins and DNA. CNTs play a dual amplification role in both the recognition and transduction events, namely as carriers for numerous enzyme tags and for accumulating the product of the enzymatic reaction. The use of CNT amplifiers, loaded with numerous alkaline phosphatase tags, has been combined with the reconcentration feature of CNT transducers to yield a dramatic enhancement of the sensitivity. Selectively grafting singlestrand DNA (ssDNA) chains onto aligned CNTs produced sensors that are highly sensitive and selective for probing complementary and even non-complementary DNA chains in the presence of target DNAs.⁹⁶ Aligned and micropatterned CNT electrodes have also been used for more advanced micropatterned ultrasensitive DNA biosensors (Figure 5).97 Aligned CNT membranes may also be developed for sequence-dependent separation and sensing of certain biological species, including nucleic acids.98-101

CNT Mechanical Sensors

Owing to their exceptional electrical and mechanical properties, CNTs have



Figure 4. Schematic illustrations of the processes for (a) fabricating and (b) characterizing the aligned CNT–polymer composite chemical vapor sensor. SEM images of the VA-MWNTs (c) before and (d) after being partially coated with a polymer poly(vinyl acetate) film on top and turned upside-down (scale bars, 5 µm). (Adapted from Reference 72.)

also been used as mechanical sensors. For example, CNTs (both SWNTs and MWNTs) were used as scanning probe microscope tips with significantly better lateral resolution than commercial Si and SiN AFM tips.102-104 Apart from the high-resolution imaging, Lieber and coworkers^{105,106} have demonstrated the possibility of chemical force mapping at molecular resolution by covalently modifying the ends of these sharp nanotube tips with chemically and/or biologically active functionalities. Subsequently, Grow et al.¹⁰⁷ further demonstrated piezoresistive pressure sensors by growing SWNTs on suspended silicon nitride membranes. In this case, a change in resistance in the SWNTs was observed, corresponding to the air pressure applied on the membrane. Furthermore, Wagner and co-workers^{108,109} have found that the Raman peak of CNTs embedded in polymer matrices showed a highly reversible shift with pressure and temperature, demonstrating the usefulness of CNTs as molecular sensors for pressureand temperature-sensing applications. More references on CNT electromechanical sensors can be found in a recent review.¹¹⁰

CNT Flow and Mass Sensors

Král and Shapiro¹¹¹ showed theoretically that the generation of an electric voltage in a metallic CNT is possible by immersing it in a flowing liquid, a phenomenon which was confirmed by an experimental observation on SWNT bundles in the flow of a polar liquid.¹¹² Recently, the flow-induced voltage generation was also observed for MWNTs with a nonlinear voltage dependence on flow rate.¹¹³ The dominant mechanism responsible for the highly nonlinear response was proposed to involve a direct "pushing" of the free charge carriers in the nanotubes by the fluctuating coulombic field of the liquid passing through the nanotubes.^{111,112} By applying an alternating voltage with a tunable frequency on a CNT under vacuum, the static charge generation was also observed to cause resonance.114,115 In analogy to a pendulum, any object attached to the nanotube could significantly affect the resonance frequency. The mass of any trapped object, down to the femtogram (10^{-15} g) level, can thus be measured simply by monitoring changes in the resonance frequency. Further research in this area could open possibilities for the use of CNTs as flow-induced nanotube voltage generators, flow sensors, mass sensors, and/or electromechanical actuators.

CNT Actuators

The exciting prospect of using CNTs as mechanical actuators was first proposed in 1996.¹¹⁶ Shortly thereafter, the first devices were demonstrated.¹¹⁷ Three types of CNT actuators will be described: electrostatic, electrochemical, and chemical. CNTs have also been used for enhancing actuation achieved by other actuator materials. The basic mechanisms and performance of nanotube actuators and hybrid actuators are reviewed in this section.

Electrostatic Actuation

Bending displacements and corresponding oscillations can be induced in CNTs by electrically charging the nanotube in close proximity to an oppositely charged electrode.^{114,115} Tweezers consisting of two MWNTs can repeatedly firmly grab, lift, and release submicron-sized objects,^{118–120} and feedback systems have been designed for process control.¹²¹ Nanotubes fixed at both ends and suspended over a counter electrode have also been studied for possible use in electromechanical data storage122 and force measurement systems.^{123,124} MWNTs have also been extensively used as torsional springs in prototype nanoscopic electric motors. As shown in Figure 6, a metal rotor was attached to a suspended MWNT in close proximity to several stator electrodes.125-127 Stator electrodes are located around the metal rotor and provide additional voltage control elements for rotor rotation. Application of voltages between the stator and rotor could produce limited or full rotation for the rotor, depending on the electric field strength applied and whether outer MWNT walls were separated from inner ones using novel processes. Torsional oscillators of resonant frequencies in the megahertz range (potentially useful in high-frequency electronics as sensors or clocks) have been built using similar designs.¹²⁸ Electrostatically driven SWNT torsion pendulums have been fabricated that enabled 180° elastic rotation.129

Other reported successes with electrostatic actuation devices are the batch fabrication of MWNT bearings126,127 and the design of tube-direction linear actuators that use displacement between layers in MWNTs for servo motors and stable ON/OFF switches for electromechanical electronic circuits.^{130–132} In particular, Zettl and co-workers first demonstrated that it was possible to telescopically pull out the inner layers from a MWNT, and van der Waals forces would pull the core back inside.131 This telescopic action has also recently been used to develop stable ON/OFF switches.¹³³ To prepare the switch, a MWNT is first suspended between two electrical contacts. A high voltage causes successive damage to the outer layers of the MWNT, eventually causing complete rupture of the nanotube (producing the OFF state). A bias voltage applied between the ruptured ends causes telescopic expansion of the nanotubes, making contact and producing the ON state. Adhesion forces are sufficient to maintain the ON state.

A similar sliding-nanotube linear servo motor has been fabricated, where the inner core of an opened MWNT can be made to telescopically move in response to a bias voltage applied to a nearby electrode.¹³² A sliding nanotube linear servo motor is a single-nanotube device, where the inner tube shell of a MWNT can be moved reversibly under applied (e.g., electrostatic) force. The movement of the nanotube shell is determined by applied electrostatic force, van der Waals interaction, and the total intershell sliding resist-



Figure 5. (a) SEM image of an individually addressable 3×3 VA-MWNT microcontact array (scale bar, 200 µm). (b) Gel electrophoresis. The lanes (migration lines of DNA samples through agarose gel) from left to right are DNA molecular-weight standard (Φ X174RFDNA-HaeIII digest), a specific polymerase chain reaction (PCR) amplicon target with ~300 bases, and a control sample with an unrelated PCR amplicon with ~400 bases, respectively. PCR enables researchers to produce millions of copies of a specific DNA sequence in approximately two hours. Gel electrophoresis is a DNA separation method that uses electricity to separate DNA fragments by size as they migrate through a gel matrix. The bright lights indicate the separated DNA fragments. (c) Schematic of the mechanism to detect DNA hybridization using a MWNT nanoelectrode array. The long single-stranded DNA PCR amplicons are hybridized to the short oligonucleotide probes, which are functionalized at the very end of the MWNTs. Ru(bpy)₃²⁺ mediators are used to transfer electrons from the guanine oxidation amplified with Ru(bpy)₃²⁺ mediators. (d) The schematic mechanism for the guanine oxidation amplified with Ru(bpy)₃²⁺ mediators. (Adapted from Reference 97.)



Figure 6. CNT nanobearing using a single MWNT as the electrical connection and mechanical support for an electrically driven metal rotor. (a) Schematic illustration. The inset shows an SEM image of the fabricated array at a tilt of 40° with the scale bar representing 50 μ m. (b) SEM image of an example device, with inset showing degrees of freedom of the nanotube. (Adapted from Reference 126.)

ance force. A bias voltage on a MWNT generating an actuation force larger than the sum of the interlayer friction and van der Waals forces on the core of the MWNT will produce a sliding nanotube servo motor. In one demonstration, a bias voltage of 30 V applied between a MWNT and a nearby electrode caused the inner CNT cores to extend by more than 1500 nm. Position sensing was also incorporated into the device to achieve feedback control.

Electrochemical Actuation

Actuation of assemblies of nanotubes can also be achieved by electrochemical charging, where the CNTs are in contact with an electrolyte. Electrochemical actuation in CNTs involves non-faradaic double-layer charging that changes carbon-carbon bond lengths.117,134 The new actuators use carbon SWNT sheets as electrolyte-filled electrodes of a supercapacitor. The actuation mechanism (quantum-chemical-based expansion due to electrochemical double-layer charging) does not require ion intercalation for device operation. Changing the applied voltage injects electronic charge into a SWNT electrode, which is compensated at the nanotube-electrolyte interface by electrolyte ions (forming the so-called double layer). The actuators use dimensional changes in covalently bonded directions caused by this charge injection. Other possible contributions to the actuation can be from electrostatic (coulombic) repulsion between neighboring CNTs.135,136 At extreme potentials, other processes including dopant intercalation and volume expansion due to gas generation (electropneumatic actuation) can also occur.

Electrochemical double-layer charge injection was initially used to produce actuation for either a nonaligned SWNT or MWNT sheet strip laminated with a second flexible sheet strip and immersed in an aqueous electrolyte containing a counter electrode (Figure 7).117,137,138 Applying a potential between the CNT sheet and the electrochemical counter electrode induced nanotube length changes that provided cantilever-based bending (Figure 7c). In aqueous electrolyte, maximum linear actuator strains of ~0.1% and ~0.2% were observed for SWNT and MWNT sheets, respectively, with the degree of actuation varying approximately parabolically with applied potential.117,138,139 Since the CNTs were randomly oriented within the sheet, these strains represent an average of the individual CNTs in both the radial and axial directions. The use of organic electrolytes¹⁴⁰ and ionic liquids¹⁴¹ having large electrochemical stability windows enabled the application of higher potentials to the CNT sheets, resulting in correspondingly increased charge injection and actuation (0.7% and 0.5% for SWNT and MWNT sheets, respectively, in acetonitrile-based electrolytes).140,142 Fully solid-state bendingtype trilayer actuators have also been demonstrated using solid electrolytes.143 A thickness strain of ~1% was also observed by actuating a SWNT buckypaper under a



Figure 7. Schematic illustration of charge injection in a nanotube-based electromechanical actuator. (a) An applied potential injects charge of opposite sign in the two pictured nanotube electrodes, which are in a liquid or solid electrolyte (blue background). (b) Charge injection at the surface of a nanotube bundle, which is balanced by the pictured surface layer of electrolyte cations. (c) Schematic edge-view of a cantilever-based actuator operated in aqueous NaCl at ± 1 V. (Adapted from Reference 117.)

small compressive load using double-layer charge injection.¹⁴⁴ A different faradaic actuation mechanism, which is current-induced or electrochemical-reaction–induced, has been reported for SWNT mats operated at high potentials in NaCl aqueous electrolytes.¹⁴⁰ In the latter case, gas (O_2 and Cl_2) is produced within the porous SWNT structure and expands to generate very large thickness direction strains (300%) and in-plane contractions of ~3%. The gas can be removed electrochemically (by reversing the potential), thereby providing reversible electropneumatic actuation.

Oriented MWNT yarns are considerably stronger than unoriented MWNT sheets and can produce actuation strains similar to those of the CNT sheets at much higher applied loads (~30 MPa).139,142 Oriented MWNT yarns generate isometric stresses (up to 26 MPa, based on a geometric cross-sectional area of the yarn) that are about a hundred times greater than natural muscle and 26 times those for unoriented CNT sheets.8,145,146 Fast actuation was achieved by using "resistance compensation" techniques to give actuation rates of 0.6%/s (for constant mechanical load)147 and 180 MPa/s (for constant length).146 Direct measurements of changes in the tube direction height of highly aligned nanotube arrays during electrochemical actuation provided actuator strains of up to 0.15%, 148 which if constrained to constant length would generate a stress of ~1 GPa, compared with the 0.3 MPa stress-generation capability typical of natural muscle.

Predictions based on the experimentally measured actuator strains (typically 0.5%) and the Young's modulus ($\overline{Y} = 640$ GPa) of prototypical SWNTs gives a potential stress generation capability of 3.2 TPa and a work density per cycle $1/2\epsilon^2 Y$ of 8 J/cm³, which are much higher than the values obtained for other actuator materials. Electrochemical actuation in conducting polymer (π -EAP) artificial muscles involves intercalation and deintercalation of electrolyte ions into the bulk material, which is often slow (due to the required solid-state diffusion) and limits cycle life (due to partially irreversible phase changes). In contrast, electrochemical actuation in CNTs results from formation of the electrochemical double layer at CNT surfaces. This process should be fast, since ion diffusion occurs within the electrolyte and diffusion distances are small. Therefore, CNT-EAPs offer several advantages over conventional low-voltage actuator materials, including π -EAPs, and continued development of aligned CNT assemblies14-22,149 and even single-CNT devices150 will likely provide continued improvement in actuator performance.

Chemically Powered Artificial Muscles

Electrically driven actuators have a major disadvantage when used in

autonomous robots and prosthetic systems because of the relatively low energy storage density of batteries and long recharge times. As a result, athletically active autonomous humanoid robots typically perform for less than an hour before recharge is needed. To help solve this problem, Ebron and co-workers^{151, 152} take advantage of the more than 30 times higher gravimetric energy storage capability obtainable for fuels than for batteries by converting chemical energy to mechanical energy locally at the artificial muscles, as done in natural muscle. In particular, actuators that convert chemical energy to mechanical energy have been developed. In the nanotube-based fuel-powered actuator, a SWNT sheet or fiber electrode simultaneously functions as a fuel cell electrode for converting chemical energy to electrical energy, as a supercapacitor electrode for storing this electrical energy as injected charge, and as an artificial muscle for using changes in stored charge for mechanical energy. Actuation occurred when positive charges (holes) were injected by chemical reaction into a SWNT sheet strip (Figure 8a). Actuation was reversed by discharging the fuel cell actuator by closing the inter-electrode circuit to allow the recombination of electrons from the hydrogen electrode with holes on an oxygen electrode (Figure 8b). The actuation strain from the nanotube-based fuelpowered actuator is within a factor of 3 of the typically 0.1% maximum strain for electrically operated high-modulus ferroelectrics.¹⁵¹ The work capability per cycle of the fuel-powered actuators can be a hundred times greater than for skeletal muscle. As such, they could serve as artificial muscles in various autonomous systems, especially where electrically driven actuators are impractical, as in many lifelike robots able to operate autonomously for long periods, active medical devices, and certain micro- or nanoactuators.

CNT Hybrid Actuators

CNTs have also been used to enhance the actuation of other actuator materials. Various examples have been published highlighting how CNTs can augment the performance of other actuator materials by making use of their excellent mechanical properties, and their high electrical and thermal conductivities to improve the mechanical properties^{153–156} and to reduce the power consumption of hybrid actuators.^{157–159} With conducting polymer electrochemical actuators, for example, the addition of small amounts of SWNTs (<1%) significantly increased the conductivity (from 500 S/cm to 700 S/cm) and mechanical strength (from 170



Figure 8. (a) Oxygen dissolved in water at right transfers charge to Pt-coated SWNT sheet (black), causing it to bend by expanding relative to the platinum/carbon/ionomer layer (dark gray). (b) The bending is reversed by closing the circuit to allow partial recombination of electrons on the hydrogen electrode with the holes on the oxygen electrode (adapted from Reference 152).

MPa to 255 MPa). As a consequence, actuation could be achieved isotonically at much higher applied stresses (up to 125 MPa) and with significantly less creep.¹⁵³ Composites of polyaniline and CNTs have also been prepared by dip-coating the CNT mat into a polyaniline solution.154 The resultant hybrid material contained 75 wt% CNTs. Even at such high loadings, the actuation was dominated by the conducting polymer component, giving strains of up to 0.5%, as compared with ~0.1% expected from the CNTs alone and $\sim 2\%$ for the conducting polymer alone. Significantly, however, it was found that the hybrid material produced higher strains than the pure polyaniline when higher isotonic loads were applied.

CNT additions have also been used with other ionic actuator materials, specifically to improve their mechanical properties. Actuators based on Nafion^{®155,156} or hydrogels¹⁵⁷⁻¹⁵⁹ have shown increased Young's modulus by addition of CNTs such that the force generation was increased. An enhanced actuation was also observed in dielectric elastomer-type actuators with the addition of ~0.1% CNTs to a nematic elastomer.¹⁶⁰

Several bending-type actuators using CNTs as the surface electrode material have also been demonstrated. In these applications, a highly conductive and flexible electrode is necessary to impart the electric field responsible for the actuation. Examples include bending actuators based on cellulose paper,¹⁶¹ piezoelectric ceramic,162 and piezoelectric polymers.163 The excellent thermal conductivity of the CNTs was useful in reducing capacitive losses due to self-heating in piezoceramic actuators, leading to a 25% reduction in power consumption.162 It is expected that similar benefits can be achieved using CNTs as flexible electrodes in dielectric elastomer and conducting polymer bending-type actuators.

Conclusion

Owing to their nano-dimensions, unique electronic properties, large surface area, high mechanical strength, and excellent chemical and thermal stability, carbon nanotubes have been shown to be useful for a wide range of potential applications. These features are attractive in the advancement of chemical/biosensors as well as electromechanical actuation. Compared with more conventional electroactive polymer actuation systems, the high elastic modulus of carbon nanotubes could lead to potentially high stress generation and high work per cycle under significantly less demanding stimulation conditions (e.g., voltage).

Recent advances have led to several different actuation mechanisms for CNTs, including electrostatic, electrochemical, and chemical actuations. Even this brief account has revealed the versatility of carbon nanotubes for making sensors with high sensitivity and selectivity and actuators with rapid, strong strain/stress responses. However, the functionalization of carbon nanotubes with macroscopically ordered structures to gain specific surface characteristics required for particular sensing applications, and to allow manipulation of carbon nanotubes into practical sensor/actuator devices, remains a challenge. Overcoming this challenge will assist in the development of sensors and actuators with enhanced performance for a number of practical applications.

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