

Integrated devices based on networks of nanotubes and nanowires

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Although advanced devices based on nanotubes (NTs) and nanowires (NWs) are drawing much attention, devices based on a single NT or NW are not suitable for general manufacturing purposes, as it is still extremely difficult to control the electronic properties, growth and alignment of individual NTs or NWs on an industrially reliable scale. An alternative strategy for implementing NTs or NWs in real-world devices is the use of NT- or NW-network-based structures containing a number of NTs or NWs. Herein, we review the recent progress in NT/NW-network-based integrated devices. The technology for NW/NT-network-based devices is supported by massive integration methods, such as directed assembly, printing and directed growth, and devices based on NW/NT networks display several unique properties, such as percolating conduction and scaling behaviors, that differentiate them from individual NT/NW-based devices. A variety of applications are possible for NT/NW networks, including transistors and sensors, all of which offer unique characteristics for use in integrated nanoelectronics.

Nanotubes (NTs) and nanowires (NWs) have a one-dimensional structure and nanometer-scale diameter, and show remarkable electrical, mechanical, optical and chemical properties [1]. However, as-produced NTs and NWs are usually in a powder form, and the integration of NTs and NWs into an integrated circuit requires assembly steps involving individual NTs and NWs. As-produced NTs and NWs also exhibit large variation in physical and chemical properties, making them inappropriate as components for large-scale integrated devices for practical applications. Networks of NTs and NWs have been reported as an alternative strategy for implementing NTs or NWs in real-world devices. Recent reports have shown that such networks can fill new roles and applications that could never be possible using individual NTs or NWs, including use as large-scale transparent conductors, in low-cost flexible electronics and as interfacing devices for living cells (Figure 1) [2,3].

In this review, we discuss the various fabrication methods for NT/NW-network-based integrated devices for practical applications. We then review several interesting properties of NT/NW-based percolative networks and the fundamental parameters that characterize such networks, and finally discuss some of the diverse device applications for NT/NW-network-based devices.

Massive integration of NT/NW network-based devices

Directed assembly

For massive integration of NT/NW-network-based devices, NTs or NWs, usually grown by chemical vapor deposition (CVD), should be assembled using some kind of force (*e.g.* directed assembly) or grown in a specific direction (*e.g.* directed growth) using predefined patterns of catalysts on a substrate. In the directed assembly method, NT/NWs

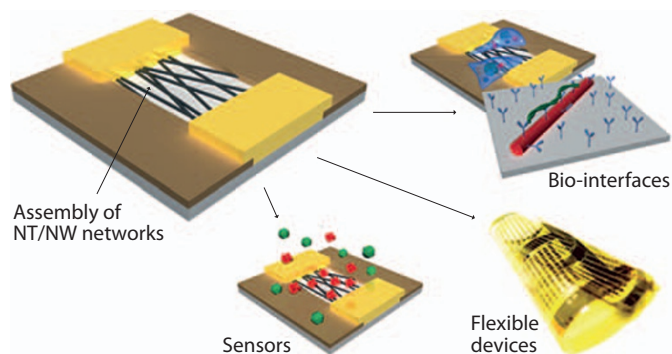


Figure 1. Examples of NT/NW-network-based devices. Networks of NTs or NWs can serve as platforms for many applications, including sensors, flexible devices and integrated circuits.

in powder form are assembled at the desired locations on a substrate. Various methods, such as dielectrophoresis, surface-programmed assembly and fluidic flow, have been developed by several groups to achieve directed assembly.

Duan *et al.* [4] fabricated electronic and optoelectronic devices by assembling indium phosphide NWs parallel and orthogonal on a substrate using a direct-current (DC) electric field (Figure 2(a)). They dropped a solution of NWs onto a substrate and applied a DC bias of 50–100 V, causing the assembly of NWs between the two electrodes. Under a very strong DC electric field, charged NTs or NWs are attracted to the electrodes via electrostatic force, allowing NTs and NWs to be selectively assembled in a specific region on a solid

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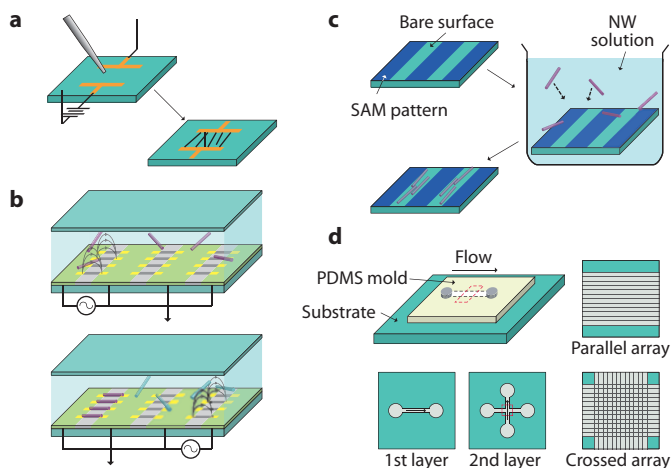


Figure 2. Directed assembly methods. (a) DC electric field assembly. Application of a 50–100V DC bias to electrodes across a NW solution causes the NWs to assemble between the electrodes. Figure modified after Ref. 4 (© 2001 C. M. Lieber). (b) AC electric field assembly. Suspensions of NWs coated with different DNA molecules are injected sequentially under an AC electric field to directly assemble and align the NWs. Figure modified after Ref. 8 (© 2009 AAAS). (c) Surface-programmed assembly. The self-assembled monolayer (SAM) molecular pattern guides the adsorption and alignment of NTs or NWs onto bare surface regions. Additional electric potential could be utilized to improve NT/NW adsorption. Figure modified after Ref. 9 (© 2006 S. Hong). (d) Fluidic flow assembly using fluidic channel structures using a polydimethylsiloxane (PDMS) mold. Figure modified after Ref. 12 (© 2001 AAAS). All figures reproduced with permission.

substrate. However, it is very difficult to control the alignment of NTs or NWs by this approach.

An alternative strategy is dielectrophoresis [5,6]. In this approach, an alternating-current (AC) bias voltage is applied to create a high electric field region between two electrodes. In a solution of NWs dropped onto the substrate surface, NWs are attracted toward the high electric field region between the two electrodes via dielectrophoretic forces. Once the electrodes are bridged by the conducting NWs, the bias voltage between the electrodes drops, preventing additional NW assembly. This self-limiting mechanism allows the assembly of individual NWs. For example, Vijayaraghavan *et al.* reported on the directed assembly of single-NT devices by applying a high-frequency AC electric field [7]. Morrow *et al.* assembled NWs functionalized with three different DNAs using sequential NW injection and application of a spatially restricted AC electric field in order to fabricate DNA-coated NW devices (Figure 2(b)) [8]. The main advantage of this dielectrophoretic strategy is that the NT/NW assembly can be activated or deactivated simply by controlling the bias voltage on the electrodes, making it possible to assemble different NTs/NWs on a single substrate using a rather simple procedure. However, as relatively large electrode structures are required in order to apply high bias voltages, it is difficult to apply this method for the fabrication of high-density electronics.

The surface-programmed assembly method, which utilizes self-assembled monolayers (SAMs) of molecules to guide the assembly of NTs and NWs, is effective for the assembly of large numbers of NT- or NW-based network arrays, as no external force, such as an electric field, is needed (Figure 2(c)). A SAM pattern can be formed simply using conventional processes such as photolithography, micro contact printing (μ CP), and dip-pen nanolithography (DPN). Lee *et al.* assembled single-walled carbon nanotubes (SWCNTs) and V_2O_5 NWs directly on various substrates, including silicon, SiO_2 , aluminum and gold, using this method [9]. In the method of Lee *et al.*, bare substrate surfaces served as polar regions and octadecyltrichlorosilane patterns as non-polar SAM regions. Due to van der Waals interaction between the carbon nanotubes (CNTs) and the polar SAM region, CNTs were assembled directly from dispersion in solution onto the desired

substrate location to form a CNT network with monolayer thickness. The electrical charge on NWs allows them to be assembled onto SAM regions with opposite charge. LeMieux *et al.* successfully controlled the chirality, alignment and density of CNTs on solid substrates through functionalization of the SiO_2 surface and spin coating of CNTs from solution. By this approach they produced thin-film transistors with high on/off ratios [10]. Functionalizing SWCNTs through a controlled cycloaddition reaction with fluorinated polyolefins similarly allowed Kanungo *et al.* to produce transistors with high mobility and on/off ratio [11]. This technique could be used to improve the efficiency of the directed-assembly method. Surface-programmed assembly using SAM patterns is a straightforward strategy that can be applied to virtually any type of NW or substrate. However, the method cannot be utilized for the assembly of larger NT/NWs with diameters greater than 100 nm because of the weaker interaction between NT/NWs and the SAM patterns in such systems.

External forces such as fluidic flow can also be utilized to exert sufficient force on larger NT/NWs to induce assembly into the desired device structure (Figure 2(d)). Lieber *et al.* used a fluidic channel structure of polydimethylsiloxane (PDMS) in contact with a substrate patterned with molecules with an affinity to the target NWs. The NWs passing through the fluidic channel were assembled onto the substrate in an alignment parallel to the flow direction [12]. Oh *et al.* reported a different fluidic flow method for the assembly of *N,N*-bis(2-phenylethyl)-perylene-3,4:9,10-tetracarboxylic diimide (BPE-PTCDDI) microwires on an SiO_2 substrate. In their method, BPE-PTCDDI microwires were first filtered through a PDMS mask to align the wires uniformly, and then transferred to an SiO_2 substrate [13]. Another useful fluidic flow method is the Langmuir–Blodgett technique, which involves floating the NWs on a water surface and compressing the wires to high density between two barriers to align the NWs in one direction. This aligned NW layer can then be transferred onto any substrate. Using nanometer-scale patterns as etching masks, Whang *et al.* generated well-aligned, dense NW films by this method [14]. Yu *et al.* reported an interesting and simple method to fabricate well-aligned NT/NW films by forming a bubble from a homogeneous suspension of NTs or NWs in an epoxy solution using a circular die [15]. The bubble film could be transferred onto a solid substrate or to an open frame structure.

Overall, the directed assembly strategy is versatile and can be applied to nearly all NWs and substrates. As this method can be conducted entirely at room temperature using conventional microelectronics facilities, it can be easily adopted by existing device industries for practical applications. However, large quantities of NT/NWs are required to prepare the NT/NW solutions that form the basis for this approach. In the case of certain NT/NWs, good dispersions in solution can only be achieved with the addition of surfactants, which can cause contamination and degrade device performance.

Direct printing

Another strategy for the assembly of NT and NW networks is direct printing, by which techniques such as μ CP, inkjet printing and DPN are used to transfer NTs and NWs directly onto a substrate surface. The dry transfer of SWCNT films to a substrate using a patterned PDMS stamp is a common method for direct printing to a substrate. Meitl *et al.* reported the generation of various CNT pattern shapes through the transfer of SWCNT films to planar and nonplanar substrates (Figure 3(a)) using PDMS stamps [16]. Ahn *et al.* used a patternless PDMS stamp to transfer nanostructures from a pre-patterned source substrate [17]. In their approach, a flat stamp was brought into contact with a patterned source (NTs, NWs, ribbons), and then the patterned semiconductor material structures were transferred to a substrate by stamping. Javey *et al.* transferred NW networks to a photoresist-patterned substrate by sliding the NW-bearing substrate against the photoresist-patterned substrate [18]. Removal of the photoresist spacers leaves the aligned NW patterns on the substrate surface (Figure 3(b)). Repeating this procedure, they generated

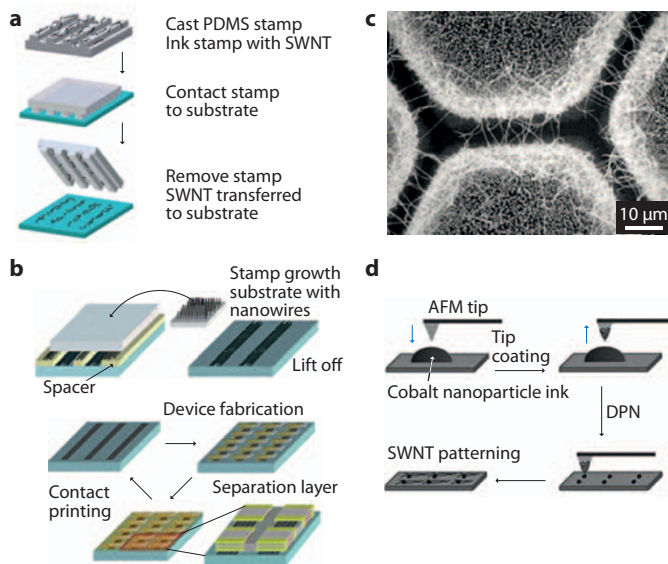


Figure 3. Printing and directed growth methods. (a) Stamping method for transferring single-walled nanotube (SWNT) patterns onto a substrate using the polydimethylsiloxane (PDMS) stamp method. Figure modified after Ref. 16 (© 2004 ACS). (b) Schematic diagram of three-dimensional NW circuit integration. NWs from the growth substrate are transferred to a photoresist-patterned substrate by sliding transfer. The photoresist patterns are then removed leaving aligned NWs. Figure modified after Ref. 18 (© 2007 ACS). (c) Scanning electron microscopy (SEM) image of ZnO NW networks grown from patterned gold catalysts prepared by gold deposition using a copper mask. Figure modified after Ref. 21 (© 2001 Wiley-VCH Verlag GmbH & Co. KGaA). (d) Cobalt nanoparticle dot array generation using an atomic force microscopy (AFM) tip for dip-pen nanolithography (DPN). Figure modified after Ref. 27 (© 2008 Wiley-VCH Verlag GmbH & Co. KGaA). All figures reproduced with permission.

three-dimensional NW-based integrated circuits in which several layers of NW patterns were stacked with alternating separation layers. Nanotube or NW networks have also been directly printed by inkjet printing. Ahn *et al.* generated various shapes of silver wires on a substrate using silver nanoparticle ink and an omni-directional inkjet printer with a cylindrical nozzle on a three-axis motion-controlled stage [19].

The main advantage of the direct printing strategy is the ability to print NT/NW patterns directly over large areas without requiring complex processing. This approach usually requires much smaller quantities of NT/NWs than the directed assembly strategy. However, it is still at present difficult to control and print NT/NWs with nanometer resolution. This strategy has therefore mainly been utilized to generate large NT/NW devices over large surface areas.

Directed growth

In directed growth strategies, NT/NWs are grown from catalytic patterns formed using various nano-patterning techniques. One of the pioneering works in this field is that of Dai *et al.*, who grew individual SWCNTs by CVD from micrometer-sized catalyst islands on a silicon substrate [20]. Huang *et al.* grew ZnO NW networks from a patterned gold catalyst by depositing gold through a copper grid mask (Figure 3(c)) [21]. Various patterning techniques such as photolithography can also be used to fabricate catalyst patterns [22,23]. The direction of NT/NW growth is often controlled by external forces. Joselevich *et al.* utilized electric fields to control the growth direction of CNTs [24]. By growing CNTs between two electrodes while applying a bias voltage, CNT growth could be directed along the electric field lines. Inkjet printing and DPN have also been used to produce the catalyst patterns needed for directed NT/NW growth. For example, Park *et al.* used an electro-hydrodynamic jet printer to produce catalytic patterns containing ferritin [25]. The use of an electric field provides fine control of ink flows with improved resolution compared with printers based on thermal or acoustic effects. Dip-pen nanolithography is performed

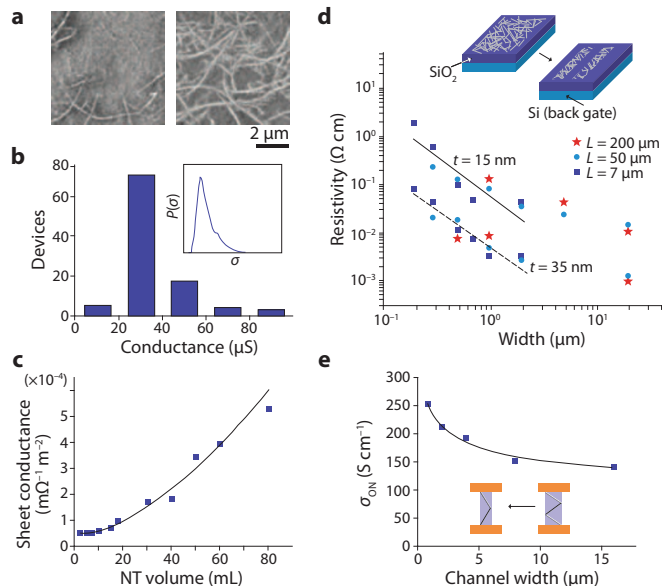


Figure 4. Unique properties of NT-based networks. (a) Scanning electron microscopy images of NT networks on alumina substrates, (left) near and (right) above the percolation threshold. Figures modified after Ref. 30 (© 2004 ACS). (b) Log-normal conductance distribution of SWNT junctions (100 devices) from Ref. 9 (© 2006 S. Hong). Inset shows earlier data reported in Ref. 31 (© 1985 APS). (c) Sheet conductance versus volume of NT solution for the deposition of CNT films. Figure modified after Ref. 30 (© 2004 ACS). (d) Resistivity of NT networks as a function of channel width, length (L) and thickness (t). Figure modified after Ref. 32 (© 2006 AIP). (e) Scaling behavior of conductivity with respect to channel width. Inset is schematic diagram illustrating the effect of reduced channel width. Figure modified after Ref. 33 (© 2009 Wiley-VCH Verlag GmbH & Co. KGaA). All figures reproduced with permission.

using an atomic force microscope, where the tip acts as a pen to transfer molecules to a substrate [26]. The DPN method has several advantages, including high resolution and registration. Li *et al.* prepared catalytic cobalt nanoparticle patterns on a substrate by DPN, on which SWCNTs could be grown by CVD (Figure 3(d)) [27].

Directed growth of NTs and NWs on a substrate allows devices to be fabricated without complicated chemical or printing procedures, and also makes it easier to achieve pristine device structures. However, the directed growth strategy requires high temperatures, making it incompatible for integration with microelectronics processing.

Unique properties of NT- and NW-based networks

Percolating conduction in random networks

The channel formed by a random network of NTs or NWs can be modeled as a two-dimensional percolative mesh comprised of a multiple interconnected NT/NWs. Classical percolation theory states that the probability of a channel forming between two spatially separated electrodes is not simply proportional to the areal density of NT/NWs. Alam and co-workers developed a generalized stick-percolation model for finite random networks of NTs and NWs [28,29]. In their model, NT/NWs are regarded as percolating sticks making up a network. Below a certain areal density of sticks ($\rho < \rho_c$), no conducting path exists. Above a threshold density ($\rho > \rho_c$), however, one or more conducting paths can be formed and carriers can flow from the source to the drain electrode. Figure 4(a) shows images of examples of CNT networks below and above the percolation threshold [30]. The resistivity of such random networks is known to follow a log-normal distribution [31]. Such a log-normal distribution has been demonstrated in a set of 100 channels of SWCNT network monolayers (Figure 4(b)) [9]. The conductance distribution exhibits a sharp peak at a certain value, indicating that one can utilize percolating NT or NW networks to produce devices with uniform characteristics.

The percolating NT or NW networks have drawn considerable attention for use in various practical applications because this approach allows the variable properties of individual NTs or NWs to be averaged, resulting in devices with uniform characteristics. However, NT/NW networks also suffer from certain fundamental limitations. Unlike individual NTs or NWs, charge transport in the random network is influenced by inter-tube or -wire contact resistance. This degrades the conductivity and mobility of random networks compared with single-NT or -NW channels. In addition, the log-normal distribution of resistivity has a rather long tail to larger values (Figure 4(b)), which implies that large networks are required in order to properly average the non-uniform characteristics of individual NTs or NWs and achieve NT/NW network-based devices with uniform characteristics.

Scaling behavior of random network-based devices

Percolative networks of NTs and NWs exhibit interesting scaling behaviors, where the electronic parameters are not simply proportional to the physical dimensions of the channel, unlike bulk materials, where, for example, the conductance is linearly proportional to the channel thickness. The SWCNT-based networks fabricated by Gruner and co-workers were found to display a relationship between sheet conductance and NT network density (Figure 4(c)) [30]. The relationship followed a power law with an exponent of around 1.5, which is a typical value for percolative systems. Behnam *et al.* also fabricated NT-based random networks with controlled channel width and length by shaping the channels with ion etching [32]. Figure 4(d) shows the resistivity of their networks as a function of network width for channel thicknesses (t) of 15 and 35 nm. For channels with width (W) less than $2\ \mu\text{m}$, the data can be fitted by a $\rho \propto W^{-1.53}$ relation for $t = 15\ \text{nm}$ and $\rho \propto W^{-1.43}$ for $t = 35\ \text{nm}$, indicating an increase in resistivity with decreasing channel width, whereas the resistivity of bulk materials does not change appreciably with changes in device shape. This scaling behavior originates from the unique property of percolated networks based on randomly oriented one-dimensional structures such as NTs and NWs. As the channel width decreases, the number of NTs or NWs in the channel also decreases. In addition, the NTs or NWs at the channel boundary lose connectivity, further reducing the number of current paths in the channel. Thus, the resistivity of the channel increases with decreasing channel width. This result indicates that random networks are not suitable for small devices with micro- or nanometer-wide channels.

Effect of NT/NW alignment in networks

Some groups have reported that the electronic properties of NT/NW networks change if the NT/NWs in the network are aligned in a certain direction. Hong and co-workers modified the structure of an NT network by aligning the NTs parallel to the channel direction using a surface-programmed assembly method [33], while Rogers and co-workers fabricated perfectly aligned NT-based networks by growing NTs on temperature-stable cut quartz wafers via CVD [34]. Hong and co-workers showed that the electrical properties of the textured NT-network devices are enhanced compared with random networks. Figure 4(e) shows the scaling behavior of on-current conductivity with respect to channel width for textured networks. Here, the conductivity increases with decreasing channel width because the NT/NWs tend to align more uniformly in the narrower channel by surface-programmed assembly (Figure 4(e), inset). This is in contrast to the behavior seen in random NT networks prepared by ion etching (Figure 4(d)). This difference in conductivity scaling behavior can be explained by noting the alignment effect of NTs in the channel direction [33]. When the NTs are well aligned, the conduction paths are connected by a smaller number of NTs. In this case, a narrower channel has a shorter effective current path with fewer NT–NT junctions, which results in enhanced conductivity in the channel. In addition, the mobility increases with decreasing channel width [33], which can also be explained by the NT alignment effect. This result indicates that alignment of networks could overcome the poor scaling behavior problem of random networks and allow the fabrication of small-scale network-based devices with high performance.

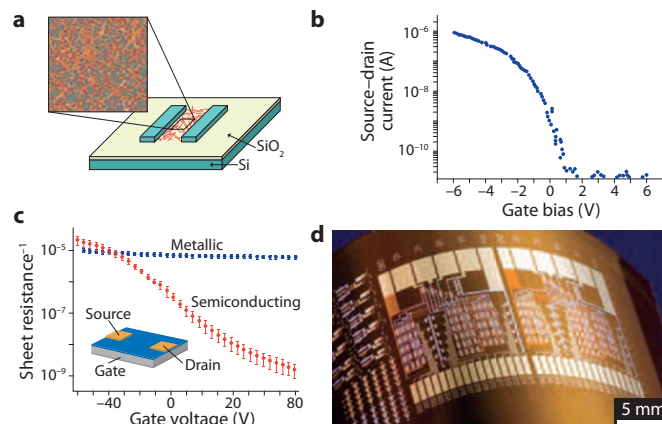


Figure 5. Electronic and electromechanical devices based on NT/NW networks. (a) Schematic of a back-gated NT network-based transistor. Figure modified after Ref. 35 (© 2003 AIP). (b) Gate effect of a random SWCNT network-based transistor after metallic SWCNT breakdown. Figure modified after Ref. 9 (© 2006 S. Hong). (c) Gate effect of SWCNT network transistors comprised of metallic and semiconducting SWCNTs. Inset shows the transistor structure. Figure modified after Ref. 36 (© 2006 M. C. Hersam). (d) A SWCNT-network-based flexible integrated circuit. Figure modified after Ref. 52 (© 2008 J. A. Rogers). All figures reproduced with permission.

Advanced devices based on NT/NW networks

Transistors and other solid-state electronics

Random networks of CNTs have been explored extensively as a practical electronics material [35]. For example, in simple transistor devices, source and drain electrodes have been connected by CNT network channels on an oxide layer with a backgate to modulate the carrier density (Figure 5(a)). Such transistors usually exhibit p-type behavior in air because of oxygen adsorbed on the CNTs. However, the device typically has a low on–off ratio due to the incorporation of metallic CNTs in the networks. Lee *et al.* showed that large-scale integrated devices based on such CNT-network transistors can be fabricated by surface-programmed assembly. The on–off ratio of such devices can be enhanced to the order of 10^6 by selective burning of the metallic CNTs in the channel (Figure 5(b)) [9]. The on–off ratio can also be increased if more semiconducting paths are formed than metallic paths. Several groups have reported methods to separate metallic and semiconducting CNTs. For example, Hersam and co-workers successfully separated metallic and semiconducting SWCNTs and fabricated network-based transistors comprised mainly of semiconducting CNTs [36]. Figure 5(c) compares the gate effect of metallic and semiconducting SWCNT-network transistors. Similarly, efforts have been made to dope the CNTs to obtain p- or n-type channels for use in logical devices. For example, Zhou *et al.* fabricated logical devices consisting of polymer gates with p- and n-type SWCNT-network-based transistors [37].

Nanowire-network-based devices have also been used in large-scale electronic devices. Unalan *et al.* fabricated thin film transistors (TFTs) comprised of ZnO network channels as the basis for complementary inverter devices [38], and Heo *et al.* produced large-scale arrays of transistors using Si-NW networks [39]. In the latter case, the thin SiO₂ layer on the Si-NWs significantly increases the contact resistance between individual Si-NWs, necessitating the introduction of intermediate contact electrodes for the fabrication of large-scale devices.

The various applications of NT/NW-based networks also include optoelectronic devices, photovoltaics devices and high-frequency circuits. For example, Fan *et al.* reported the large-scale, heterogeneous integration of photosensitive CdSe NWs and high-mobility Ge/Si NWs for image sensors [40]. In optoelectronics and photovoltaics, networks of CNTs are expected to complement or perhaps replace the role of certain inorganic layers such as ZnO and indium tin oxide (ITO). A 50 nm-thick layer of networked CNTs on a transparent sapphire

substrate was reported by Wu *et al.* to have a sheet resistance of less than $100\ \Omega\text{m}^{-2}$ and a transmittance greater than 70% in the visible region [41]. Hatton *et al.* replaced ITO films in organic light-emitting diodes and organic photovoltaic devices with layers of multi-walled CNTs (MWCNTs) [42]. Using the same technique as Hersam's group [36], Green *et al.* fabricated an improved transparent conductor by using sorted metallic SWCNTs [43].

As a replacement for ITO, layers of networked CNTs are still inferior in many respects, in particular their higher resistance at inter-tube junctions and the random orientations of CNTs in the network. However, the high mechanical stability and flexibility of CNTs are evidently advantageous over brittle ITO for future flexible display applications. A number of challenging developments, including post-treatment of CNT film to enhance conductivity [44], are being attempted to produce CNT networks with performance superior to that of ITO.

Although most of the studies so far have concentrated on the DC or near-DC properties of CNT networks, increasing numbers of high-frequency applications for CNT networks are being reported. Due to the low impedance of CNT networks, direct scattering-parameter measurement of SWCNT TFTs is possible. Kocabas *et al.* implemented a fully functional radio using SWCNT TFTs for all active components [45]. Carbon nanotube transistors display relatively high cutoff frequencies; Le Louarn *et al.* have reported an intrinsic current gain cutoff frequency of 30 GHz [46], while Chimot *et al.* implemented a high-frequency transistor with a current gain cutoff frequency of 1 GHz [47].

Electromechanical and flexible devices

Physical strain or buckling causes changes in the electrical properties of CNTs, and these changes manifest as a current change under a voltage bias. Such a property can be utilized to implement strain or pressure sensors based on networks of NTs. For example, Dharap *et al.* reported a simple stress sensor that utilizes a CNT network film to detect tensile and compressive stress using a four-probe measurement setup [48]. In that study, the researchers obtained a linear relationship between induced voltage and strain. Similarly, Maune *et al.* assembled CNTs on elastomeric substrates for use as local strain sensors [49], and Lee *et al.* demonstrated a pressure sensor based on vertically suspended CNT network channel, where an initially stretched-out CNT channel undergoes mechanical bending via external pressure resulting in a change in the conductance of the CNT channel [50].

Carbon nanotubes can be easily incorporated into metals as laminates to form composite structures with enhanced mechanical properties. In the incorporation of CNTs into aluminum suspending nanobridges, Bak *et al.* showed that the Young's modulus of the composite was higher than that of the individual components, and that the natural frequency of the bridge was increased, features that are advantageous for high-frequency mechanical actuators [51].

In the field of flexible circuits, Rogers and co-workers have performed a series of pioneering studies in implementing flexible integrated circuits using CNT networks (Figure 5(d)) [52]. They have been able to transfer CVD-grown SWCNT networks onto flexible polyimide substrates for the fabrication of digital logic circuits comprised of up to 100 SWCNT network transistors. The transistors thus fabricated had top-gated structures and were based on high- k dielectric materials. The performance of the circuit could be modeled using a commercial circuit simulator, and the researchers demonstrated a four-bit decoder operating at a clock frequency of 1 kHz using this logic circuit.

Chemical sensors and biosensors

Nanotubes and NWs display remarkable properties when used in devices sensing external target molecules. The high surface-to-volume ratio of these one-dimensional nanostructures makes NT/NWs particularly suitable for use in sensor transducers. Pristine SWCNTs, for example, consist entirely of surface atoms, and are therefore very sensitive to the adsorption of molecules on their surfaces. For example, NT- and NW-based networks have been used as sensitive gas sensors. The general

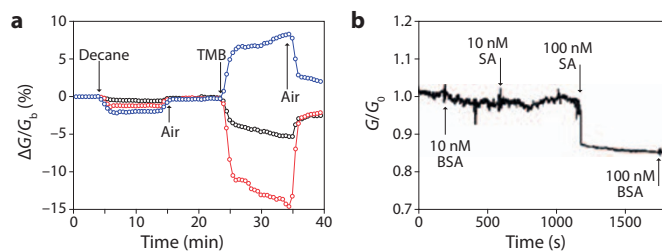


Figure 6. Use of NT/NW networks for gas and biomolecular sensing applications. (a) Normalized conductance change $\Delta G/G_0$ with respect to base conductance G_0 for the detection of nonpolar molecules (decane) and polar molecules trimethylbenzene (TMB) using random CNT-network device. Figure modified after Ref. 55 (© 2009 ACS). (b) Normalized conductance change due to specific streptavidin (SA) binding to functionalized CNTs. Sensor shows no response to bovine serum albumin (BSA). Figure modified after Ref. 59 (© 2003 NAS USA). All figures reproduced with permission.

form of such sensors is the field-effect transistor (FET) structure, in which two electrodes and a channel form the basic device. During operation, adsorption of target molecules results in chemical gating, which modulates the carrier density in the channel. Early studies in gas sensing involved the use of single-CNT FET structures to detect NH_3 and NO_2 gases [53]. Networks of NTs were soon used to sense gases. Novak *et al.* constructed a simple chem-resistor structure consisting of a CNT film between metal electrodes to detect sub-parts-per-billion concentrations of dimethyl methylphosphonate, a simulant for the nerve agent sarin [54]. Recently, Peng *et al.* detected nonpolar molecules in gas phase using random CNT networks by functionalizing the SWCNTs with organic molecules to enhance carrier scattering (Figure 6(a)) [55]. They were able to detect nonpolar molecules (decane) and polar molecules such as trimethylbenzene using random NT-network devices. Lim *et al.* patterned SWCNT networks on flexible PDMS substrates for the detection of NH_3 and NO_2 based on the conductance change in a simple two-probe device [56]. Snow *et al.* have proposed a capacitor structure in which changes in the capacitance between the CNT network and the gate provide for the detection of various gas species, including acetone and dimethylmethylphosphonate [57]. Sun *et al.* fabricated high-performance hydrogen sensors with mechanical flexibility by decorating networks of SWCNTs on thin plastic substrates with palladium nanoparticles via electrochemical deposition [58].

Another sensor application of NT/NW-network-based devices is in the field of biosensors. Nanotubes and NWs have diameters comparable to those of various biomolecules, including DNA and proteins, suggesting the possibility of direct interaction with single biomolecules. One of the seminal works in this field was performed by Chen *et al.*, who used CNT-network devices to detect antibodies and proteins such as streptavidin (Figure 6(b)) [59]. They reported the detection of protein binding by the noncovalent functionalization of CNTs for enhancement of specificity toward target proteins.

DNA detection using NTs functionalized with single-stranded DNA for the detection of complementary strands has been reported by Star *et al.* [60]. Those researchers successfully monitored DNA hybridization in a label-free manner using a FET structure of NT networks set between interdigitated electrodes. Gui *et al.* studied the effect of NT/electrode contact metals by monitoring the gate profile changes in response to DNA hybridization [61], reporting that the sensing mechanism originates from both the charge transfer and Schottky barrier modulation related to DNA hybridization. Tang *et al.* used NT-network devices to investigate the effect of DNA hybridization on the work function of the electrodes [62]. They showed that a change in the work function of the electrodes results in a change in the height of the Schottky barrier at the interface of the electrode and the NTs. Using CNT FETs with passivated electrodes, Kim *et al.* determined that biorecognition events at the CNT channel are responsible for the current change, and that the Debye length around the NTs plays an important role in biorecognition by the NTs [63].

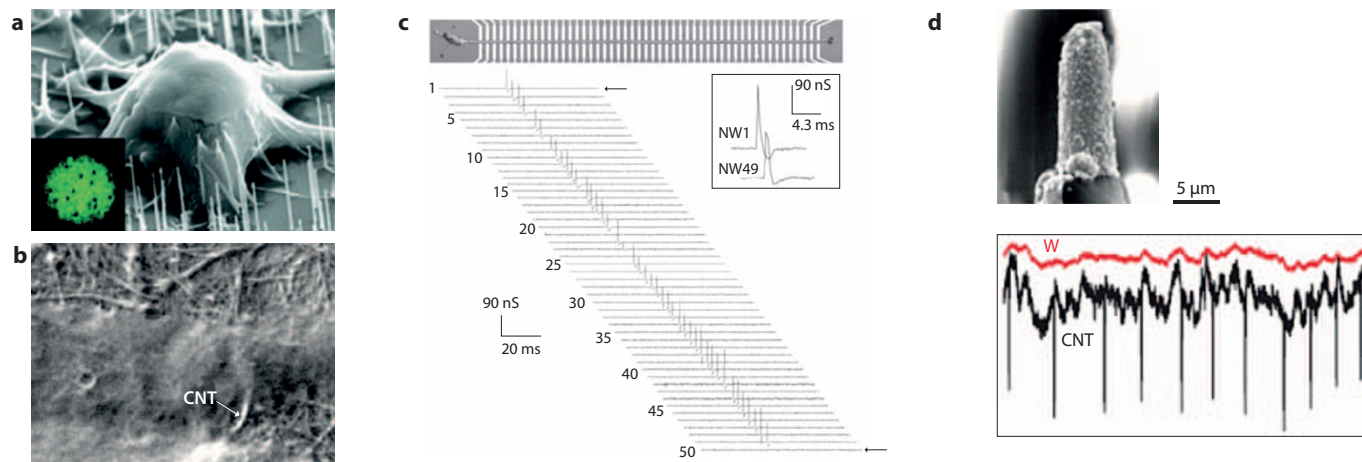


Figure 7. Interfacing NT- and NW-based networks with living cells and monitoring cellular signals using NTs and NWs. (a) SEM image of mouse embryonic stem (mES) cells on a NW array substrate. The insert shows a confocal microscopy image of mES cells penetrated with silicon NWs. Figure modified after Ref. 64 (© 2009 ACS). (b) SEM image at higher magnification of the neurons grown on SWCNTs. Figure modified after Ref. 66 (© 2007 Society for Neuroscience). (c) Monitoring neuronal signal propagation using highly integrated Si-NW arrays. Figure modified after Ref. 73 (© 2006 AAAS). (d) CNT-coated tungsten electrode (upper) and recorded data for a bare tungsten (lower, red) and CNT/gold-coated (lower, black) stereotrode tips. Figure modified after Ref. 75 (© 2008 E. W. Keefer). All figures reproduced with permission.

Substrates for cell and tissue engineering

Considering the compatibility of NT/NW-based networks with biological molecules and their versatile mechanical and electrical properties, it is natural to study the response of living cells in nano-textured environments or to use NT- or NW-based devices for the monitoring of complex cellular/sub-cellular functions or signals.

The functions of living cells, such as adhesion, motility and differentiation, are affected by various microenvironmental factors (*e.g.* chemicals, biological molecules, surface structure, topography). Due to the high aspect (or surface-to-volume) ratio of CNTs, a nanoscale CNT structure can provide mechanical support and a favorable extracellular environment for cell growth compared with that provided by a smooth surface. The use of NTs or NWs in the study of living cells could elicit a wide range of cellular responses depending on structure and surface functionalization. Kim *et al.* demonstrated that the use of vertically aligned Si-NWs (Figure 7(a)) was not fatal to cells for a short time, even when a single cell was penetrated by 20–30 NWs [64]. They were also able to transfer genetic material into the cells using DNA-bearing Si-NWs. It may also be possible for an NT- or NW-based network structure to affect not only cell morphology and cytoskeleton organization, but also intracellular signaling pathways. It has been reported that CNT networks or matrixes make close contact with the cell membrane, an effect that has been confirmed by scanning electron microscopy (Figure 7(b)) and transmission electron microscopy [65,66].

Due to membrane deformation by the nanoscale surface roughness of CNTs, the motion of vesicles in the near-membrane region, as observed by total internal reflection fluorescence microscopy, may be hindered [67]. In that study, the areal coverage of vesicle motion was reduced in cells grown on a CNT layer, demonstrating clearly the nanotopographical effects of CNTs on intracellular characteristics.

With regard to the effect of NT- or NW-alignment on living cells, an aligned nanofiber surface has been shown to significantly enhance cell migration compared to randomly oriented nanofibers [68]. This result indicates that aligned NT- or NW-based networks affect cell migration significantly.

Nanotube- and NW-based networks functionalized with chemicals or biological molecules can affect cell growth behaviors, which could prove useful in tissue engineering. For example, MWCNTs coated with 4-hydroxynonenal have been shown to enhance the growth of multiple neurites and branches of embryonic rat hippocampal neurons [69].

In recent years, NTs and NWs have even been utilized in stem-cell engineering. Several NW/NTs (*e.g.* CNTs, ZnO NWs, V_2O_5 NWs) have been tested for human mesenchymal stem cell (hMSC)

growth [70]. The results show that assembled CNT monolayer patterns induce selective hMSC growth and cell alignment. The selective cell growth can be mainly attributed to the higher affinity of cell adhesion proteins to CNTs than to other surface regions. In common cell adhesion processes on solid substrates, cell-adhesion proteins, such as fibronectins and laminins, adhere to certain regions on the substrates, and then cells bind to the proteins. In our group's experiments, we have found that the high affinity of CNTs toward cell-adhesion proteins results in preferred cell adhesion onto CNT patterns. An immunofluorescence study of fibronectin adsorption confirmed the higher adsorption of fibronectin to CNT regions compared with other surface regions [70]. This affinity is presumed to be due to the strong non-covalent interactions between CNTs and proteins, including hydrophobic interactions and π - π interactions. This type of affinity has also been exploited by Jan and Kotov, who used a layer-by-layer assembly method to prepare multilayer thin films of SWCNTs and a polyelectrolyte as a substrate for the differentiation of mouse neural stem cells into neurons, astrocytes and oligodendrocytes [71].

As the number of studies on cell responses to CNTs has increased, issues regarding the cytotoxicity of CNTs have also been raised. However, a general agreement on the issue has yet to be reached. Early research on the topic often resulted in contradictory results due to inter-study dissimilarities in experimental conditions such as cell lines, assessment methods, doses, forms of CNTs, purification methods of metal catalysts and CNT functionalization methods. However, it could be concluded from reported data that large agglomerations of CNTs and residual metal catalysts might have higher apparent cytotoxicity than an individual CNT. A few researchers have also reported that CNTs display pulmonary toxicity *in vivo*, although there have so far been only a few case studies [72].

Devices for monitoring cellular signals

In parallel with the interfacing of NTs and NWs with living cells, recent research has demonstrated that both NTs and NWs could be utilized to monitor spatially resolved cellular signals or to enhance sensor sensitivity for cellular signals. Patolsky *et al.* monitored neuronal signal propagation using Si-NW arrays (Figure 7(c)) [73]. In that study, the researchers functionalized the surface of the device to guide axon growth across individual Si-NW FETs. After stimulation, neuronal signals transmitted through the ion channels in the axon and the ion currents through the ion channels changed the effective potential surrounding the individual Si-NW FETs. The altered effective potential was monitored by measuring the source–drain current of the Si-NW FETs in the highly integrated

Si-NW arrays, which allowed the neuronal signals to be read at 50 sites along an axon.

In other research, Heller *et al.* succeeded in monitoring activities inside macrophage cells by applying an interesting strategy to insert CNTs into the cells [74]. The CNTs were coated in advance with antibodies, which caused the macrophage cells to recognize the coated CNTs as an external pathogen, triggering phagocytosis to ingest the coated CNTs. They also deposited platinum nanoparticles on the CNTs to enhance the detection of reactive oxygen and nitrogen species, which are released during phagocytosis. The detection of spatially resolved vesicular release events was possible due to the nanoscale dimensions of the CNTs.

An alternative approach to the use of NTs or NWs as the current channel in FETs was demonstrated by Keefer *et al.*, who used CNTs as an interface between cells and metal electrodes [75]. In that work, CNT-coated tips were shown to have lower impedances and higher charge transfer compared with uncoated tips, attributed to the high surface-to-volume ratio and high conductivity of the CNTs. The CNT-coated tips provided greatly enhanced stimulation efficacy and sensitivity in the recording of neuronal signals (Figure 7(d)). Successful electrical stimulation of neuronal cells using CNT electrodes has also been demonstrated [76]. Similarly, Cellot *et al.* obtained improved neuronal responsiveness through the use of SWCNTs to directly stimulate neural circuit activity [65]. The intimate contact between cultured neurons and SWCNTs was shown in that study to provide close electrical coupling of the neurons and SWCNTs.

Sub-cellular components have also been hybridized with NTs and NWs in order to investigate sub-cellular functions in a controlled manner. In such experiments, specific sub-cellular components, such as membranes or membrane proteins, are separated from other complex cellular systems to allow highly controlled experiments to be performed. For example, Zhou *et al.* used CNTs to study the mobility of lipids and bound proteins in a series of lipid bilayers [77]. They reported that the synthetic lipid bilayers on the CNTs mimicked cell membranes and that the CNTs could be used to monitor events in the membrane. Using a similar strategy, Misra *et al.* deposited ion channel-bearing membranes on a Si-NW FET [78]. Using this hybrid system, they succeeded in monitoring voltage- and ligand-gated ion transport through membrane pores. As the function of membrane proteins in cells depends on the structural equilibrium with the surrounding membrane and is restricted by two-dimensional diffusion, these model systems should provide a useful framework for studying events at the cell surface. Kim *et al.* used a CNT-based device containing human olfactory receptor protein embedded in a lipid membrane as a sensing component for a 'bioelectronic nose' [79]. The olfactory receptors provide unmatched selectivity and sensitivity for the detection of odorants, and the binding of odorants to olfactory receptors induces structural changes in the receptor proteins that cause a concomitant lowering of CNT conductance. In this approach of Kim *et al.*, the natural function of the human olfactory receptors combined with CNT devices made it possible to discriminate odorants at the single-molecule level. The selectivity of CNT sensors was also greatly enhanced by incorporating the olfactory receptors into the lipid membrane, which should provide an efficient block to non-specific binding of other molecules similar to the membrane's protective role in the cell system. The combination of highly selective olfactory receptors and protection of the lipid membrane could therefore be a plausible explanation for remarkable performance of Kim *et al.*'s CNT-network-based bioelectronic nose.

Hybrid nano-bio-mechanical system

As another application of the hybridization of NTs or NWs with sub-cellular components, NTs and NWs have been assembled with motor proteins to build highly efficient transport systems. This approach could broaden the application of NTs and NWs from electrical devices to nanoscale bio-mechanical devices. Motor proteins are essentially molecular machines that are responsible for many mechanical processes

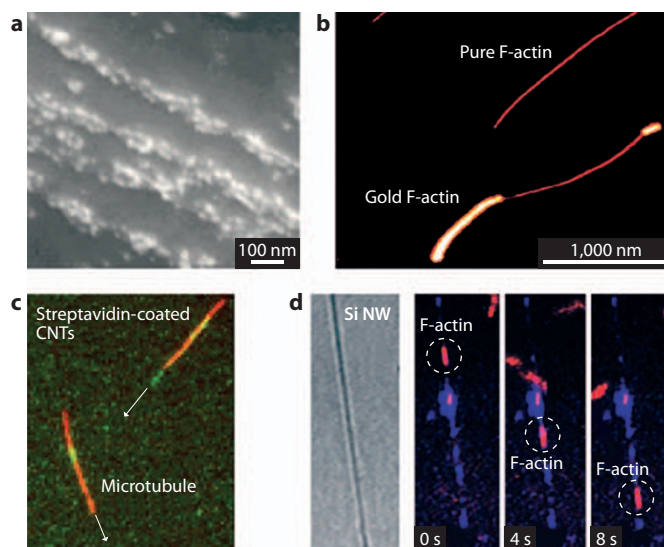


Figure 8. Hybrid nano-bio-mechanical structures based on motor proteins and NTs/NWs. (a) SEM image of the gold NW array formed on microtubule patterns. Figure modified after Ref. 80 (© 2008 Wiley-VCH Verlag GmbH & Co. KGaA). (b) Fabrication of gold-NW-actin filament hybrid structure. Figure modified after Ref. 81 (© 2004 I. Willner). (c) Transportation of CNTs by microtubules. Figure modified after Ref. 82 (© 2008 Wiley-VCH Verlag GmbH & Co. KGaA). (d) Motility of actin filament along a myosin-functionalized Si-NW. Figures modified after Ref. 83 (© 2009 Wiley-VCH Verlag GmbH & Co. KGaA). All figures reproduced with permission.

in biological systems. For example, myosin interacts with actin filaments to generate force in muscles, while kinesin cooperates with microtubules to transport chromosomes, proteins and vesicles in cells. These nanoscale molecular machines also perform their original function outside of the biological system, and interact with other materials such as NTs and NWs.

The high aspect ratio of actin filaments and microtubules makes cytoskeleton components good candidates for assembling NT and NW-based motor protein complexes. Gold NWs have been fabricated by attaching gold nanoparticles to both microtubules and actin filaments as templates [80]. The nanoparticles adsorbed on the microtubules and actin filaments were enlarged catalytically and connected to form highly conductive NWs. Alignment of the microtubules to an array using tau protein and subsequent metalization then gave gold NW arrays (Figure 8(a)) [80]. Actin filaments have also been utilized to fabricate gold NWs [81], where part of the filaments were left unmodified to provide a site for interaction with myosin (Figure 8(b)). This hybrid gold NW-actin filament structure was able to move on a myosin-coated surface. MWCNTs have also been transported by motor proteins (Figure 8(c)) [82]. To achieve this effect, MWCNTs were functionalized with streptavidin, dispersed in a buffer and then loaded onto biotinylated microtubules via streptavidin-biotin linkage. The microtubules transported the MWCNTs on a kinesin-coated surface.

Conversely, NWs have also been used to manipulate the movement of motor proteins. As motor proteins display controlled movement in biological systems, many researchers have tried to build narrow motor protein patterns or guiding channels to imitate the one-dimensional movement of *in vivo* systems. In the work of Byun *et al.*, the myosin-functionalized Si-NW acted as a track for actomyosin motility (Figure 8(d)) [83]. In that study, the actin filaments were confined to the Si-NW and underwent one-dimensional transport along the wire. These results demonstrate the possibility of combining various inorganic nanostructures with motor proteins while retaining their functionality. This method should provide considerable flexibility and could prove to be a key technology for building nanomechanical systems based on motor proteins.

Summary

The ease of manufacture makes NT- or NW-network-based devices promising for a range of practical applications where single NTs or NWs would be applicable. Many approaches to the large-scale fabrication of NT/NW-network-based devices have been reported, and much theoretical effort has been devoted to understanding the unique properties of percolating network structures. There is also extensive research on applying these networks in advanced applications such as high-performance electronics, flexible devices, sensors, tissue engineering and even cell-signal monitoring. Many of these applications are only possible due to the combination of unique properties of NT/NW networks, such as flexibility, high mobility and sensitivity. Networks of NT/NWs could therefore form the basis for a completely new device industry in the future.

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