

Formation of Highly Dense Aligned Ribbons and Transparent Films of Single-Walled Carbon Nanotubes Directly from Carpets

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owing to the attractive physical properties of carbon nanotubes, materials and applications involving carbon nanotubes have been sought since their discovery. Recently, the ability to grow both multiwalled^{1,2} and single-walled^{3–6} carbon nanotubes in aligned arrays perpendicular to a substrate from which they grow has brought new ideas toward highly scalable applications involving carbon nanotubes. One major advantage of vertically aligned arrays of carbon nanotubes (carpets) is that the carbon nanotube alignment is sustained during growth, leaving the arduous processing to achieve alignment by wet chemical methods unneeded.⁷

Recently, carpets have been receiving increased attention from the standpoint of applications. Ranging from self-cleaning “gecko” adhesives^{8–10} to foundation materials for the spinning of aligned fibers of MWNT,^{11,12} aligned nanotube arrays are promising for a large range of applications, spanning many disciplines and fields. Because of a homogeneous nucleation density of nanotubes in carpets in addition to the inherent nanotube alignment that is present from growth, these materials surely have promise in the field of thin films. In the past, the ability to form thin, transparent films composed of carbon nanotubes has opened the door for a multitude of important applications involving single-walled carbon nanotubes. One of the most attractive of these is the use of carbon nanotube films as flexible, lightweight, low-cost alternatives for indium tin oxide (ITO) coatings which are notorious for being fragile and expensive to produce.¹³ Currently, ITO coat-

ABSTRACT Utilizing aligned carbon nanotube arrays grown from chemical vapor deposition, we present a highly scalable route toward the formation of ribbons and ultrathin transparent films directly from vertically aligned single-walled carbon nanotube arrays (SWNT carpets). To “lay-over” the aligned nanotubes to form a film, we use a roller which acts to compress the film and preserve the alignment of nanotubes within the film. As we demonstrate, we can control the nanotube-catalyst interaction, leading to highly efficient transfer of the film to virtually any host substrate by following growth with a controlled H₂O vapor etch. In addition, we demonstrate our ability to grow carpets on patterned substrates leading to upright carpet lines, which can be rolled over to form transparent films composed of ultralong carbon nanotubes. This work demonstrates a highly scalable technique to form homogeneous, transparent films of aligned SWNTs that can be ultralong with absolutely no need for liquid phase SWNT processing.

KEYWORDS: transparent films · carbon nanotubes · chemical vapor deposition

ings are utilized in a wide range of applications, including touch-screens, flat panel displays, electronic ink in digital displays, anode terminals in organic light emitting diodes, among many others. The numerous applications described here emphasize the technological importance of producing highly scalable, cheap alternative materials composed of carbon nanotubes for these applications.

In this spirit, recent pioneering work by Rinzler¹³ has established a method to make transparent SWNT films based on vacuum filtration of surfactant suspended SWNTs. Following filtration, the membrane can be dissolved away to leave a random network of SWNTs whose transparent properties can be varied based upon the thickness of the network. This work has paved the way for numerous follow up studies characterizing the stability and properties of these films,^{14–17} including the most recent work where this method is utilized along with density gradient centrifugation to produce films with an enriched metallic SWNT

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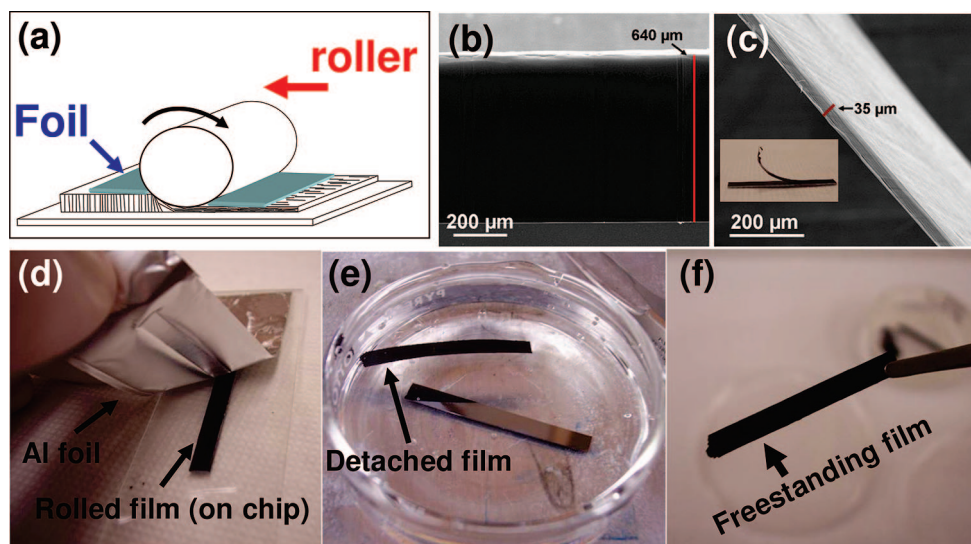


Figure 1. General illustration of the roll-over process and one possible route toward detachment by catalyst etching with HCl. (a) Cartoon of the rolling process to form films of aligned single-walled carbon nanotubes. The translational direction and rotational direction of the roller are each labeled with arrows (red and black, respectively). The film is compressed by a downward applied force during the rolling process. Before (b) and after (c) SEM images of rolled-over carpets. Inset in panel c is a photograph of the carpet where the film is peeled over a carpet to image the thickness. (d) Image showing the result from rolling over a carpet, as the aluminum foil is detached from the carpet. Notice the lack of any transfer to the Al foil. (e,f) Wet chemical process for detaching films, including (e) detachment by catalyst etching in 1 M HCl solution and (f) an image of a freestanding film after being soaked in H₂O (being held by tweezers).

content.^{18,19} However, from an industrial perspective, this technique involves multistep liquid processing that combines (i) long periods of sonication to suspend SWNT in surfactant solution, (ii) centrifugation to remove impurities from the suspension, (iii) vacuum filtration to form the film followed by another liquid treatment to dissolve the filtration membrane, and (iv) vigorous washing to remove excess residual surfactant from the film. In addition, it has been observed that even a light exposure to air, among other liquids and solvents, can result in dramatic changes to the physical properties of SWNT films.¹⁹ At the current time, the roles of excess ionic surfactant and/or residual membrane species in the SWNT films have not yet been established.

Other recent pioneering work by Baughman²⁰ has utilized the alignment in multiwalled carbon nanotube arrays to produce sheets of aligned nanotubes that can be drawn directly from the side of the carpet. This process involves no liquid treatment to produce MWNT conducting films, even though it does heavily depend upon the internal structure of the carpet (*i.e.*, alignment, density, bundle size, *etc.*). Because of the known structural differences between MWNT and SWNT forests,^{21,22} our experiments to date to make SWNT sheets in this way have been unsuccessful. More recently, Murakami and Maruyama²³ have developed a method for removal of intact arrays of vertically aligned SWNT from the growth substrate by placing a cold carpet in hot water. They found that this method is effective at removing pieces of the initial carpet based on the thermocapillary effect. The drawback of this method is that the ma-

jority of those who grow carpets cannot afford such a wet processing step, since water will penetrate into the array, leaving capillary forces during the drying process to form cellular or monolithic-like structures in the carpet.^{24–26} This not only disrupts the alignment of the nanotubes, but also affects their physical properties.

Therefore, in this article we have developed a simple, scalable technique to form ribbons and ultrathin films of aligned single-walled carbon nanotubes of various thicknesses directly from carpets. As we demonstrate, by manipulation of the catalyst-film interface, we determine a mechanism by which film

removal and transfer can take place with no liquid treatments or intermediate steps. We find that transfer to nearly any host surface is possible, and we observe that the transfer process preserves the majority of alignment that is present in the initial carpet. Combined with the highly controllable and simplistic nature of a chemical vapor deposition reactor for SWNT growth, we believe this method could be a highly efficient route toward large-scale production of ribbons and aligned transparent films of SWNTs.

RESULTS AND DISCUSSION

Figure 1 shows the typical process that is employed for compression of the vertically aligned carbon nanotube arrays, as well as a film during subsequent stages of a wet transfer process that can be utilized as one route to etch the catalyst away and detach the SWNT film. In Figure 1a, a diagram of the general rolling method of the carpet is illustrated. In this method, the roller undergoes rotation and translation as shown in Figure 1a. In our experiments, the compression takes place because of an applied downward force by the hands during the rolling process. For the case where the catalyst-film interaction mandates that the film remain anchored to the surface, a thin sheet of foil between the roller and the carpet keeps the nanotube arrays from sticking to the roller surface. It should be also noted that prior to the rolling process, the foil is sheared along the direction of rolling in order to retain alignment in the film. This is found to be of greater importance in the longer carpets, where it is easier to smash the carpet instead of “laying over” the carpet as

intended. When no postgrowth catalyst treatment is utilized, the carbon nanotubes are well-anchored to the substrate resulting in absolutely no transfer to the foil regardless of the material utilized. This was performed with Cu, Al, Ta, and stainless steel foils, amidst polymer films such as polyethylene. The result from this method is a highly compressed film which retains alignment, as verified by scanning electron microscopy (SEM) and Raman spectroscopy. To have a good measure to the degree to which this process compresses the carpet, we studied a carpet grown at elevated pressure (25 Torr instead of 1.4 Torr), where the growth averaged 613.6 μm tall in 30 min. After compression, the resulting film was partially peeled from the substrate in order to measure its thickness. SEM measurements indicate that the average thickness of the film is 30.3 μm , which is over a 20 \times compression from the initial carpet. SEM images of the before and after, as well as an inset of a photograph of the carpet, are shown in Figure 1(b–c). The measured density of the carpet shown in Figure 1b is 20.6 mg/cm³, which after a 20 \times compression is 416 mg/cm³.

Further shown in Figure 1(d–f) are the processes for wet removal of the film. Later in this article, we will introduce a process for dry removal of the film, but we utilize the wet transfer process to emphasize that catalyst etching, that is, breaking C–Fe bonds, results in film detachment from the surface. In this particular case (for Figure 1), the carpet is grown by high temperature exposure to C₂H₂, H₂O, and H₂, followed by rapid cooling in this same gas flow environment. We find that this process results in a film with nanotubes that stay tightly bound to the growth substrate. In fact, the only way we find to remove such films is with adhesive tape after compression. Thus, by compression, we form a dense film that is adherent to the catalyst that resides at the base of the nanotubes. To remove the film, we etch the catalyst with 1 M HCl solution, which subsequently releases the freestanding film from the substrate in a matter of several seconds. Images of the compression and wet etch step are shown in Figure 1d,e, and a demonstration of a freestanding film after removal from the growth substrate is shown in Figure 1f, where the film is shown supporting not only its own weight, but also the weight of water from which it was taken.

After establishing the process by which films can be made, it is essential to understand whether the alignment in the film is retained after compression. One common way to characterize film alignment is

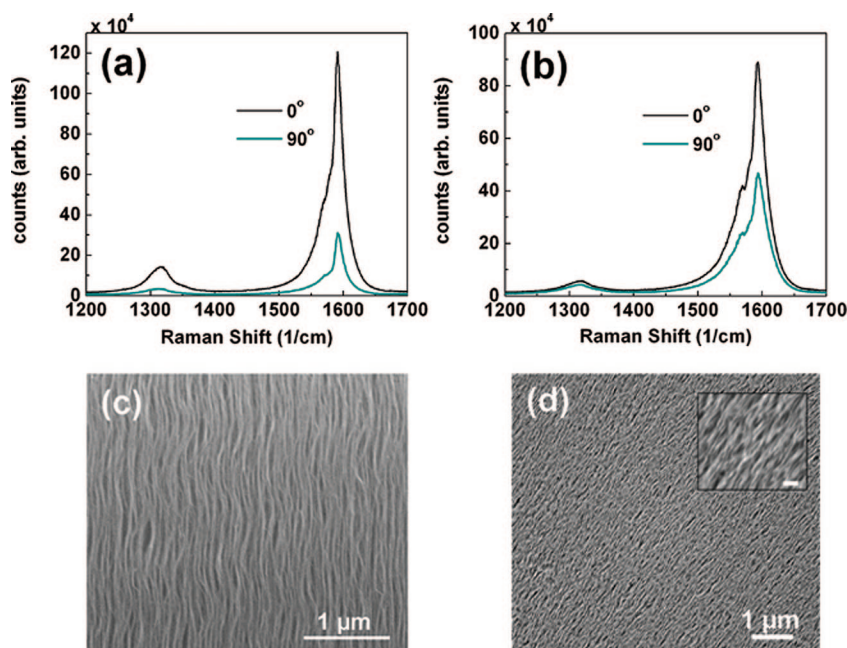


Figure 2. (a,b) Polarized Raman spectra (633 nm) of the D and G bands from films formed by the process illustrated in Figure 1; 0° corresponds to a configuration where the polarization direction of the laser light is parallel to the alignment in the carpet, while 90° corresponds to a configuration where the alignment is perpendicular to the direction of laser light polarization. The laser spot is focused on (a) the side of an as-grown carpet, and (b) the top of a rolled-over SWNT film. (c,d) SEM images of carbon nanotube alignment in (c) the side of an as-grown carpet and (d) the top of a rolled carpet (same carpet as shown in panel c). Scale bar in the inset figure is 100 nm.

through polarized Raman spectroscopy, which is shown in Figure 2 for a carpet before and after compression. In Figure 2a, a 633 nm laser spot is focused on the side of the carpet, with the laser light polarization both parallel (0°) and perpendicular (90°) to the carbon nanotube alignment direction. Since the absorption of laser light by a SWNT should decrease as the angle between the laser light polarization and the nanotube axis approaches 90°,^{27,28} this is a good measure of the overall alignment of the carbon nanotubes in the array. As shown in Figure 2a, the ratio between the intensity of the G-band in the parallel (0°) configuration is approximately 3.9 \times that of the perpendicular configuration, typical of a carpet. Following the shear/compression step, the ratio of the G-band at 0° and 90° is about 2, as shown in Figure 2b, suggesting that some of the alignment is lost in the process of forming the film. It should be noted that the compression process in this work is performed by hand, and an industrially optimized process would fare significantly better in retaining the initial alignment of the carpet.

In addition to Raman spectroscopy, SEM images shown in Figure 2c,d indicate that the alignment that is inherent in the carpet (Figure 2c) is retained in a top view image of the film (Figure 2d). In the film, the carpet is transferred onto a piece of carbon tape after the shear/compression technique, so the image shows the part of the array that initially is contacting the catalyst surface. Despite the observed loss of alignment, SEM

images suggest that the nanotube bundles appear to be well-aligned after compression. We note that alignment measurements with Raman spectroscopy could be influenced by the top layer of the film, which is sheared by the foil. We tested this by cutting the film in the longitudinal direction and testing if the alignment measurement was any different. We observe the same ratio in polarized Raman, even though the cutting process shears the film as well, and may disrupt the observed aligned structure. In any case, this supports that carbon nanotube alignment is retained in the process of laying over the carpet to form thin films.

In terms of scalability of the process of carpet and film removal, it is favorable in most cases to not utilize a wet chemical process, but to utilize a dry process that will detach the nanotubes from the catalyst surface so that the film can be transferred to a host substrate. This keeps the nanotubes in a pristine state with no further effect of capillary forces during drying or else residual HCl or solvent/surfactant remaining in the film. We find that such a technique is possible by allowing the carpet to rapidly cool in the presence of the acetylene, turning off the acetylene, and then allowing the carpet to undergo a $\text{H}_2\text{O}/\text{H}_2$ etch at the growth temperature (775°C) for up to 5 min. By doing so, we find that the carpet can now be easily attached to nearly any arbitrary surface by nothing more than contact with a host surface. This ability to transfer to any arbitrary surface is demonstrated in Figure 3 by showing a picture of a typical carpet that has been transferred to copper and stainless steel foils, as well as a thin piece of polyethylene. We find that the films are very sticky and form strongly bound flexible layers on all the host surfaces attempted. It should be noted that nearly any clean surface can be utilized for transfer. We were even successful at transferring to polished quartz and sapphire surfaces, even though a surface such as glass, with highly nonuniform surface roughness often resulted in partial film transfer.

In addition, we took the next step to utilize one of the major strengths of a low pressure CVD reactor, which is the precise control over short pulses of growth gas after rapid catalyst reduction with atomic hydrogen. To achieve this, a three-way valve system was specially designed to route the incoming $\text{C}_2\text{H}_2/\text{H}_2$ toward a mechanical pump or alternatively toward the furnace. Utilizing a capacitance monometer, we ensured that the inlet pressure to the furnace was identical to the pressure of gas when routed toward the vacuum pump. This gave us the ability to quickly deliver highly controllable, short pulses of carbon to the catalyst surface in order to grow well-defined thicknesses of films. Combined with the transfer process described above after a high temperature H_2O treatment, we find that it is a straightforward process to transfer a transparent thin film of aligned SWNTs to a polyethylene host substrate. Utilizing UV-vis-NIR transmittance measurements for

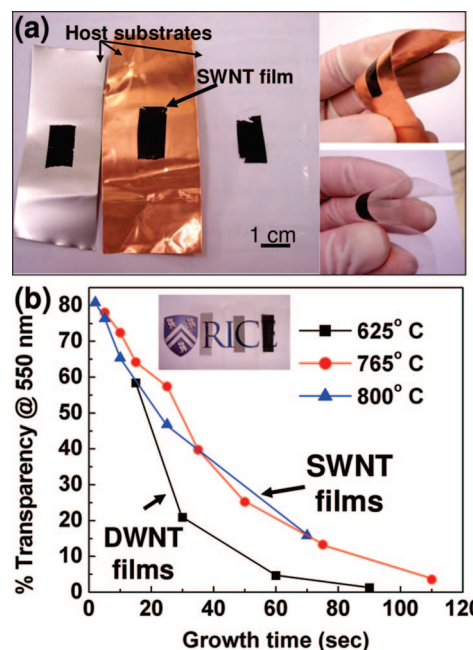


Figure 3. (a) Images of SWNT films attached to stainless, Cu, and polyethylene host substrates by H_2O etching after growth. The right-side panel shows the flexible and adherent nature of the films. (b) Percent transmittance at 550 nm as a function of growth time for films grown at three distinct temperatures. Two of the temperatures utilized (765 and 800°C) result in the growth of SWNTs, whereas growth at 625°C results in the growth of mostly double-walled carbon nanotubes (DWNT). Inset in the figure is a picture of three films of different transparencies sitting on a Rice logo.

the two films to determine film transparency, we have plotted the transparency at 550 nm as a function of growth time in Figure 3b. We did this for growths performed at three different temperatures. To produce high quality SWNT material, we grew at 765 and 800°C , where we find that our samples are composed of purely SWNTs. However, by lowering the growth temperature to 625°C , we are able to synthesize mostly double-walled carbon nanotubes (DWNT). The ability to vary the number of walls on the basis of a constant CVD carbon flux and decreasing temperature has been described by Puzos and Wood.^{29,30} As is evident from Figure 3b, the DWNT film is less transparent than both of the SWNT films for the same amount of deposited carbon. On the basis of our observations of growth rates in carpets and our estimation of the degree in which the film is compressed by the rolling method, the average thickness of a film can be estimated (at 765°C) to evolve at $\sim 0.3 \mu\text{m}/\text{min}$, or approximately $5 \text{ nm}/\text{sec}$. However, more important in judging the quality of the behavior of the film is the amount of carbon present (since the film is not a fully dense structure), for which transmittance data taken at 550 nm gives a standardized comparison of this quantity for SWNT films.

In addition to transparency, the DC electrical properties of these two films were tested after the growth took place. Typical sheet resistances along the alignment direction for as-grown films directly following growth

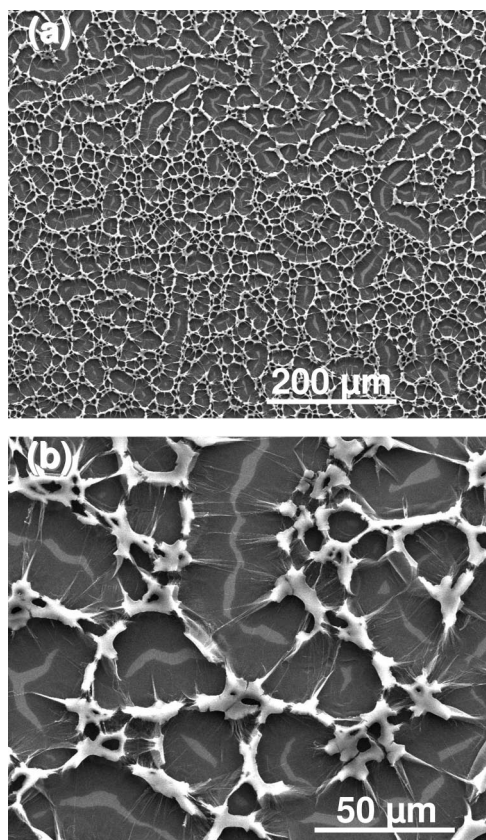


Figure 4. Scanning electron microscope (SEM) images of carpets after capillary force-induced drying occurs in a carpet that has not been subjected to a water vapor etch after growth. Panels a and b are two different magnifications of the same region.

(less than 1 h air exposure) were ~ 20 and ~ 2.8 $\text{k}\Omega/\text{sq}$ for a 60% and 20% transmitting film, respectively. After doping the films with vapors of HNO_3 overnight, we find that the sheet resistances of these films drop to 1.5 and 0.13 $\text{k}\Omega/\text{sq}$, respectively. The sheet resistance values for the as-grown carpet films are consistent with previous values for undoped SWNT films, and the 20-fold decrease in sheet resistance upon doping is consistent with studies which have investigated the effect of doping on vacuum annealed SWNT films.^{19,31} We note that a highly detailed characterization of the electrical and magneto properties of these films will be published elsewhere, as this is a topic too extensive in nature to include in this contribution.

After demonstrating these two methods for film removal from carpets, both a wet process where the catalyst must be etched away to remove the carpet, and a dry process by which a water vapor etch must be utilized, we focused on understanding the mechanism by which the films can be removed from the substrate by only an H_2O etch following growth. To study the effect of catalyst-film interaction, we grew two identical films (2 min, 750 °C), except one of them was treated with a H_2O etch for 1 min following growth, and one was rapidly cooled and removed from the reactor. Following growth, a droplet of water was placed on the top of

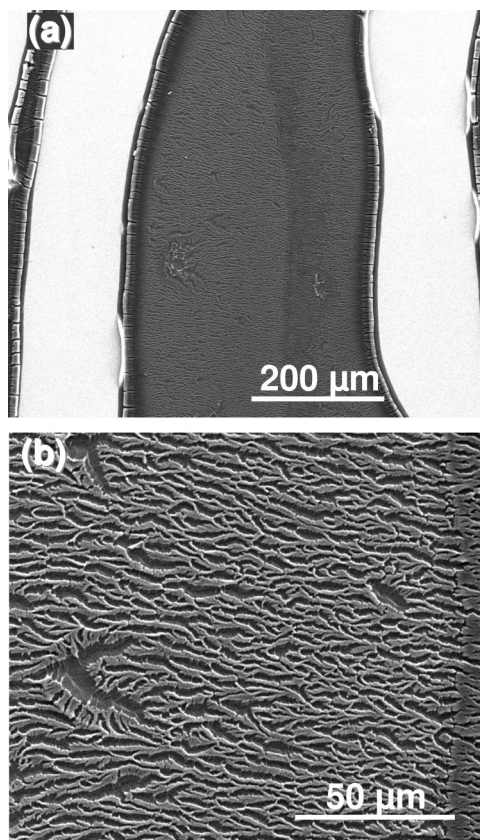


Figure 5. SEM images of an identical “collapsed” carpet to that shown in Figure 4, but with a water-vapor etch following growth. Again, panels a and b are two different magnifications of the same region.

each carpet and allowed to dry. Scanning electron microscope images of the two carpets after drying are shown in Figures 4 and 5. It is well-known that drying of a liquid after wetting an aligned carbon nanotube array results in the “collapse” of the carpet into highly dense mesas or cellular structures due to capillary forces between adjacent nanotube bundles as the liquid evaporates.^{24–26} With this in mind, the collapse of the carpet that was not etched with H_2O following growth (Figure 4) indicates the typical capillary force-induced drying effect that we are familiar with for our carpets. This is composed of small mesas of dense carbon nanotubes that have spider-web like features indicating that the collapse process occurs by “ripping” the nanotube bundles from the surface. The weblike features are indications of the strong surface interaction in the process of drying, emphasizing that the interaction between the catalyst and nanotubes is strong. However, this is not at all what is observed when a simple 1 min H_2O etch is performed following growth (Figure 5). In this case, the collapse occurs on a larger scale, with large voids forming between the collapsed regions. This is a characteristic of a weakly surface-bound film, isolating the H_2O vapor etch as being the process which is fundamentally responsible for our ability to transfer the films to any desired host substrate.

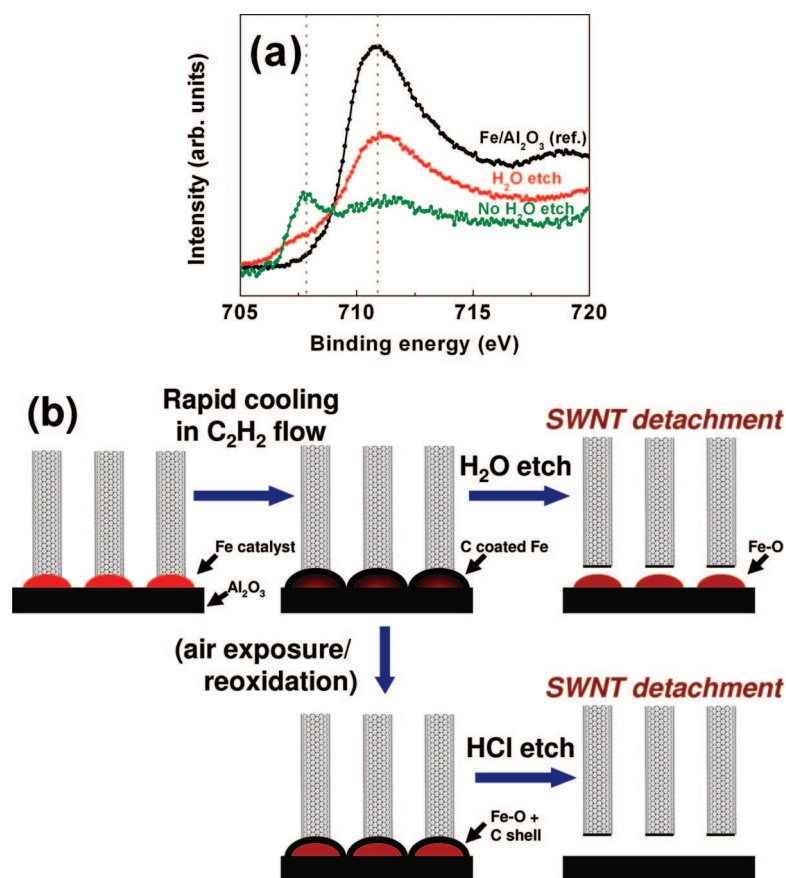


Figure 6. (a) Core-level X-ray photoelectron spectroscopy (XPS) results for Fe ($\text{Fe}2\text{P}_{3/2}$) in three different (labeled) cases, corresponding to (i) an as-deposited (and oxidized) $\text{Fe}/\text{Al}_2\text{O}_3$ reference, (ii) a catalyst layer, after growth and rapid cooling in C_2H_2 , followed by carpet removal prior to measurements, and (iii) a catalyst layer where a 60 s water etch is performed after rapid cool-down in C_2H_2 . Dotted lines mark peaks at 711.0 and 707.8 eV. (b) Diagram depicting the possible paths toward carpet removal, both with an H_2O etch after carpet growth (top panel) and an HCl catalyst etch after rapid cooling in C_2H_2 and air exposure.

After demonstrating the process by which transfer from the growth substrate can occur, we decided to investigate the mechanism behind the observed difference. Based upon X-ray photoelectron spectroscopy (XPS) data, shown in Figure 6a, a picture emerges for the mechanism of this process, which is illustrated in Figure 6b. In most CVD reactor systems, the catalyst-coated chip is rapidly inserted in a hot furnace to grow, and then rapidly cooled by pulling it out of the furnace while the carbonaceous gas is still flowing. As the catalyst particle cools, it will form a Fe–C compound, which involves a surface-segregated carbon shell surrounding the catalyst due to the difference in surface energy between Fe and C. At this point, the nanotubes in the arrays are fixed to the catalyst particle by C–C bonds to the C shell, which has mixed Fe–C bonds to the catalyst. The end result is an array of nanotubes that are anchored to the catalyst-coated substrate and can only be removed in a dry process by peeling with a sticky adhesive material, such as scotch tape or carbon tape, or else by a brute force method of etching away the catalyst with an acid (as we demonstrated earlier).

This is why a compression step with a foil on a carpet that is directly pulled out of the CVD reactor will leave the intact film on the chip with absolutely no transfer of the film to the foil. We can then proceed to acid etch the Fe catalyst from the substrate, which will release the intact film, but will destroy the catalyst layer. However, in the case where the chip is (i) pulled out of the furnace, (ii) the C_2H_2 flow is turned off, and (iii) the catalyst is exposed to a high temperature ($775\text{ }^\circ\text{C}$) treatment involving a mixture of H_2O and H_2 , the carbon in the catalyst particle is precipitated out and etched away by the water while the catalyst particle is reoxidized. Mechanical stresses in the film are expected to aid in the “pop-off” mechanism of the nanotube from the catalyst, and the SWNT film can now be easily removed by contact with another surface. This picture is verified in XPS data, shown in Figure 6a, that shows three core-level Fe spectra lines. One is a reference $\text{Fe}/\text{Al}_2\text{O}_3/\text{Si}$ chip that has a typical 0.5 nm Fe coating on a 10 nm Al_2O_3 support layer. The other two cases are after growth and removal of the SWNT array. In one case, the carpet was exposed to a H_2O etch after growth and cooling in C_2H_2 flow, and in the other case, the carpet was cooled in a C_2H_2 flow with no etch step. In each of the latter two cases, the carpets were grown, removed, and placed in the XPS system with limited air exposure (less than 2–3 min). XPS results are presented in Figure 6a with binding energy values relative to the core level adventitious carbon peak, located at 285.0 eV. One can clearly observe that the $\text{Fe}2\text{P}_{3/2}$ core-level peak positions

for the case of Fe with no growth, as well as the Fe after growth, H_2O etch, and carpet removal are in the same vicinity of highly oxidized Fe (Fe_2O_3) with core-level peaks fit to Gaussians with centers near 711.0 eV. However, $\text{Fe}2\text{P}_{3/2}$ spectra from the carpet that is grown and cooled in C_2H_2 before removal is indicative of a spectrum we have observed numerous times in similar experiments, with a core-level binding energy peak fit to 707.8 eV. This is too high for metallic Fe, and best corresponds to the formation of a Fe–C compound, as the binding energy for Fe_3C is at 708.1 eV,³² and there are many possible Fe–C states. This supports the picture that is illustrated in Figure 6b showing the different states of the catalyst in the two cases of film removal presented here.

Finally, it is in our interest to retain full carpet alignment after the lay-over process, as well as retain the ultralong nanotubes that can be achieved through carpet growth, in an effort to make a thin film. This motivates us to consider carpets which are grown after utilizing optical lithography to form narrow 1 or 2 μm lines of $\text{Fe}/\text{Al}_2\text{O}_3$, that are separated by a pitch of

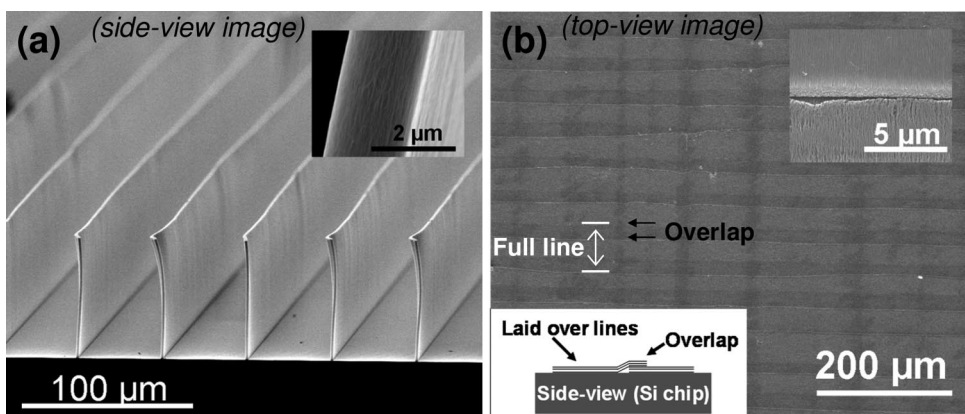


Figure 7. (a) SEM images looking down $2\ \mu\text{m}$ thick lines composed of vertically aligned SWNTs separated by spaces of $50\ \mu\text{m}$. The lines were grown in such a way as to support upright, vertical growth (see Supporting Information). Inset in panel a is a high magnification image of the edge of a single line, showing the aligned nanotube bundles inside. (b) SEM image of the top of a chip after the $2\ \mu\text{m}$ lines shown in panel a are rolled over by the process shown in Figure 1. Note there are two contrast regions: a thicker overlap region where the lines extend onto each other (illustrated in lower left inset), and a region between the overlap regions where there is only a compressed $2\ \mu\text{m}$ line. The vertical lines in the images are artifacts of the rolling process.

$50\ \mu\text{m}$. As shown in Figure 7, SEM images suggest that such lines ($2\ \mu\text{m}$ thick) can be grown to be self-supporting to heights near $70\ \mu\text{m}$. This is done by placing the direction of the lines perpendicular to the gas flow in the CVD apparatus (see Supporting Information). Since the height of the lines are greater than the spacing between each subsequent line, the lay-over process described in Figure 1 can be applied in a straightforward way. This methodology allows the resulting film to retain the full alignment of the initial carpet, be thin enough to appeal to transparent film technology, and still have nanotubes that are ultralong (50 or more micrometers). Figure 7b shows the lines in Figure 7a after the layover step (looking from top-down instead of a side view). One can notice that there are overlap regions that can be clearly observed corresponding to the fact that the lines are $20\ \mu\text{m}$ longer than the spacing between the lines. Also inset in Figure 7b is a close-up view of the overlapping region of the two lines— showing that one line is sitting on the other, and the nanotubes are highly aligned in both cases. Polarized Raman spectra (see Supporting Information) for the before and after lines indicate that 90.5% of the alignment is retained in this process, with about 5% uncertainty from the process of manually focusing the laser spot in achieving the G-band intensity. In addition, we find that the transfer process is just as straightforward as that previously described, and that highly transparent films of ultralong carbon nanotubes can be achieved in this way. In fact, we have achieved full transfer of arrays of lines to the following materials: KBr, NaCl, quartz (z-cut), sapphire, SiO_2 , MgO, Kapton, polyethylene, and glass. Therefore, this is a powerful method to achieve highly aligned, homogeneous films composed of ultralong SWNTs that are transparent and can be easily transferred to a host substrate of choice. This is important, as the future of sensor and device

technology based on aligned SWNTs will require one to not be constrained to a supporting substrate or device that is conducive to SWNT growth.

Finally, in the future we envision that aligned SWNTs will be grown in a continuous method, with carpets supported on meters of cheap metal foil substrates. In the transfer method presented here, the limitation of how much we can coat by a thin film is not dictated by the actual transfer process itself, but rather on our ability to grow on a substrate that will suffice high surface-area needs. This will surely be realized in the future when applications such as this introduce a demand for enhanced production of aligned SWNT. The work we present here provides a direct route to making thin and thick films of SWNTs that can be scaled in parallel to such a high productivity growth system. In addition, this method allows one to achieve an aligned, transparent film of as-grown SWNTs, free of metal or impurities, composed of ultralong nanotubes, and without any surfactant or solvent treatment.

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Supporting Information Available: Additional experimental details regarding growth of upright carpet lines and Raman spectra of films made from lines. This material is available free of charge via the Internet at <http://pubs.acs.org>.

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