Highly Organized Two and Three Dimensional Singlewalled Carbon Nanotubes-Polymer Hybrid Architectures

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ABSTRACT

Despite the remarkable progress in carbon nanotube technologies, the fabrication of scalable and tightly controlled integrated microscale functional flexible systems with singlewalled carbon nanotubes (SWCNTs) has remained largely elusive. Here, we present a new class of highly organized SWCNT network-polymer hybrid structures, which were never been demonstrated before, by incorporating aligned SWCNTs in desired locations, orientations, and dimensions on/inside flexible polymer matrices, representing an unprecedented control over growth, assembly, and transfer processes of SWCNTs. Several SWCNTs architectures have been built on polymer materials ranging from two-dimensional suspended SWCNTs micro-lines on PDMS micro-channels to three-dimensional "PDMS-vertically aligned SWCNTs-PDMS" sandwich structures. Also a combined lateral SWCNTs micro-lines and vertically aligned SWCNTs flexible device was demonstrated.

Keywords: hybrid architectures, flexible, transfer, singlewalled carbon nanotube

1 INTRODUCTION

Despite the remarkable progress in carbon nanotube technologies, the fabrication of scalable and tightly controlled integrated micro-scale functional flexible systems with organized singlewalled carbon nanotubes (SWCNTs) has remained largely elusive [1-9]. In order to build integrated SWCNT based flexible systems for diverse applications from electronics to functional membranes; it is required to have abilities to place highly organized and aligned two and three dimensional SWCNTs network architectures inside or on the flexible polymer substrates with controlled orientations, geometries and dimensions. To achieve this, three methods have been developed and employed: controlled growth [10-12], a well-defined fluidic assembly [13-16], and a precise transferring of SWCNTs [3, 5, 17]. To create complex two- and three-dimensional SWCNTs based flexible systems, a combination of both or all three of these techniques are required. We first designed and built horizontally and vertically organized SWCNTs network architectures at the micro-scale on SiO_2 substrates using a template guided fluidic assembly and chemical vapor deposition (CVD) methods, respectively. For horizontally organized SWCNTs network structures, a plasma treatment was used to enhance the hydrophilic nature of SiO_2 surfaces. Then, a photoresist film, hydrophobic to a SWCNT solution, was lithographically patterned to assemble SWCNTs into predesigned network structures following the hydrophilic surface patterns. To create millimeter scale long, organized, and vertically aligned SWCNTs structures, a highly effective ethanol CVD was conducted on micro-patterned Co catalyst films. The preference of SWCNTs to grow normal to, and selectively on, catalyst deposited surfaces forced SWCNTs to inherit the topography of the patterned Co catalyst film, resulting in vertically aligned and organized SWCNTs micro architectures. Then, these two- and three-dimensional SWCNTs micro-architectures were completely transferred onto the surface or inside of selected polymer matrices using wet-contact stamping and polymer casting transfer methods without disturbing the alignment, shape and dimension of the original SWCNTs network architectures.

2 RESULTS AND DISCUSSIONS

Fig. 1 shows striking examples of highly organized two-dimensional SWCNTs-polymer hybrid architectures. In Fig. 1a, arrays of aligned SWCNTs micro-lines are seen suspended orthogonally across polydimethylsiloxane (PDMS) micro-trenches. This unique structure was fabricated using a wet-contact stamping method: wet etching the SiO_2 layer underneath of assembled SWCNTs micro-lines in diluted HF solution followed by stamping with a patterned PDMS substrate. This wet-etching process can greatly attenuate the adhesion between SWCNT patterns and its original substrate and the stamp can effectively take away predesigned SWCNT patterns. SWCNTs micro-structures could also be transferred into other polymer films such as poly-methyl methacrylate (PMMA) using a polymer casting transfer method. For this, a layer of PMMA was casted on horizontally assembled SWCNTs micro-lines and cured such that SWCNTs were held by the PMMA polymer. By etching the underneath SiO_2 layer in a diluted HF solution, the PMMA layer with SWCNTs micro-structures could be peeled off from the original substrate. Fig. 1b shows the folded SWCNTs/PMMA film and vacancies along the with pattern (SWCNTs strips) which inherit the defects during the assembly indicate this transfer technique can achive almost 100% transfer. Together with SWCNTs micro-lines, we were also able to transfer gold micro contacts, deposited on the SiO_2 surface before the assembly of SWCNTs, forming the flexible device ready for electrical measurement (Fig. 1c).

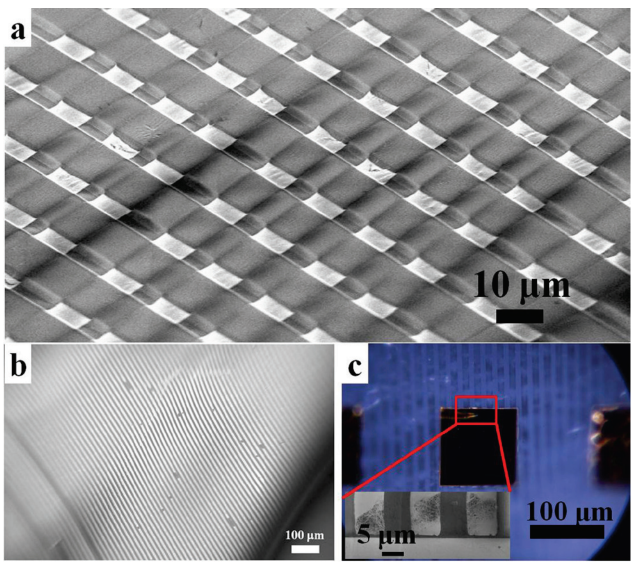

Figure 1: Horizontally organized SWCNTs-polymer hybrid structures. (a) Highly ordered arrays of one centimeter long SWCNTs micro-lines (9 µm in width) suspended across PDMS micro-trenches (6 µm in width, 3µm in depth and 9µm in space). (b) SWCNTs micro-lines (9 µm in width and 6 µm in space) incorporated into the thin PMMA film. (c) An optical image of SWCNTs micro-lines transferred on the thin PMMA film together with gold contact pads (100 µm by 100 µm, 150-nm-thickness). Scalar bars are (a) 10 µm, (b) 100 µm, and (c) 100 µm (inset 5 µm).

This nanotube transfer strategy could be extended to fabricate vertically aligned SWCNTs micro-structures on flexible polymer substrates as shown in Fig.2a. For this, arrays of vertically aligned SWCNTs (depicted in black columns in schematics) were grown on the SiO_2/Si substrate using an ethanol CVD method (step 1). Then, this SWCNTs/SiO_2/Si substrate was turned upside down and brought in contact with a very thin layer of uncured PDMS spin-coated on the SiO_2/Si substrate (step 2 and 3). After the PDMS was completely cured, the SiO_2/Si substrate on both sides can be detached leaving free standing SWCNTs directly planted on the surface of PDMS films (step 4). For examples, vertically aligned few millimeters long SWCNT arrays (Fig. 2b) or micro-patterned structures (Fig. 2c) could be planted on the top surface of the thin PDMS film. The length of SWCNTs planted into the surface of PDMS was controlled by changing the thickness of uncured PDMS film to obtain the effective transfer of vertically aligned SWCNTs with the maximized active surface area resulting in a macroscopic nanotube based gecko feet structure. It should be noted that the good wettability of PDMS on SWCNTs enables the casted thin PDMS films to anchor the top and bottom of vertically aligned SWCNTs structures effectively providing mechanically strong interfaces between SWCNTs and PDMS substrates [18].

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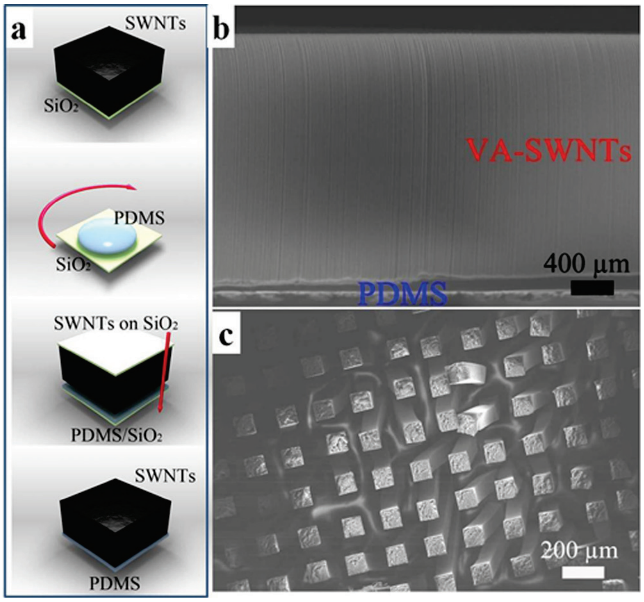

Figure 2: Vertically aligned and organized three-dimensional SWCNTs-polymer hybrid structures. (a) The schematic of contact polymer casting transfer method. (b) A cross-sectional SEM image of vertically aligned long SWCNTs film planted on the top surface of the thin PDMS film (300-µm-thickness). (c) Top view SEM image of square patterned vertically aligned SWNCT micro-pillars (100 µm by 100 µm) planted on the thin PDMS film.

Furthermore, we explored the potential for combining lateral and vertical SWCNT structures to construct three-dimensionally ordered electrical conduction networks inside of polymer materials. Fig. 3a shows micrographs of vertically aligned SWCNTs line patterns impregnated into a thick transparent PDMS matrix and interconnected to horizontally aligned SWCNTs micro-lines and gold contact pads placed on the top and bottom of the PDMS substrate. And the schematics was shown in Fig.3b. For this, firstly three columns of vertically aligned SWCNT line structures (Fig. 3a and Fig. 3b) were inserted in the bulk PDMS substrate using a polymer casting transfer method [7]. Then, the top and bottom surfaces of a PDMS substrate were mechanically polished to expose both ends of vertically aligned SWCNTs. This polished PDMS/vertically aligned SWCNTs substrate was used as the stamp and orthogonally contacted by two layers of optically transparent two dimensional ultra thin SWCNTs micro-lines (9 µm width) together with gold contact pads after removing SiO_2 underlayer using a diluted HF solution. Finally, the whole sample was sealed in the PDMS. The all-nanotube-polyer device inheriting the transparency and flexible nature of PDMS and thin lateral SWCNTs structure has great potential to be used in optical and eletrical applications .

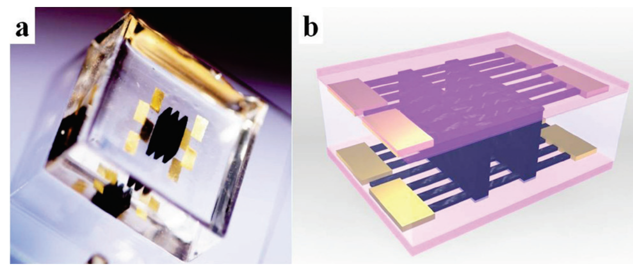

Figure 3: A combined lateral SWCNTs micro-lines and vertical aligned SWCNTs arrays inside of PDMS substrates. (a) An optical image showing horizontal and vertical SWCNT networks along with gold contact pads inside a centimeter thick PDMS matrix. Note that vertically aligned SWCNTs line structures (3 mm height, 7 mm length, and 700 µm width) are shown in black inside of a transparent PDMS substrate and physically contacted by arrays of SWCNTs micro-lines (9 µm in width and 6 µm in space, transparent in the optical image) with a 90° angle on the top and bottom of a PDMS substrate. (b) The schematic of combined three dimensional lateral SWCNTs micro-lines and vertical SWCNTs-polymer hybrid structures.

3 CONCLUSIONS

To summarize, we presented mechanically and electrically robust 2-3 dimensionally organized and aligned SWCNT networks-polymer hybrid structures, which were never been demonstrated before. Our fabrication method can be scaled up to a large scale and compatible with the current micro-fabrication technology. These multi-dimensional micro-scale SWCNT networks-polymer hybrid structures have immediate and immense implications for the development micro-scale multifunctional flexible systems such as sensors, actuators, interconnects, transparent flexible electrodes, advanced micro-fluidic devices, and smart membrane systems.

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