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Thermally Switchable One- and Two-Dimensional Arrays of Single-Walled Carbon Nanotubes in a Polymeric System

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Abstract: Fabrication of highly ordered arrays of single-walled carbon nanotubes (SWNTs) has been of great interest for a wide range of potential applications. Here, we report thermally switchable one- and two-dimensional arrays of individually isolated SWNTs formed by cooperative self-assembly of functionalized SWNTs and a block copolymer/water system. Small-angle X-ray scattering measurements reveal that when the block copolymer/water system is in an isotropic phase, two-dimensional hexagonal arrays of SWNTs are formed by depletion attraction, and when the block copolymer/water system is in a lamellar phase, one-dimensional lattices of SWNTs intercalated in the polar regions of the polymeric lamellar structure are formed by entropically driven segregation and two-dimensional depletion attraction. These two SWNT arrays are thermally interchangeable, following the temperature-dependent phase behavior of the block copolymer/water system.

Introduction

Single-walled carbon nanotubes (SWNTs) have remarkable electrical, thermal, and mechanical properties[1,2] which provide a broad range of potential applications, such as nanoscale electronic devices,[3,4] sensors,[5−7] energy storage materials,[8,9] and reinforcement for materials.[10,11] Self-assembly or guided assembly of SWNTs into highly ordered superstructures with well-defined morphology, density, and direction has been of great interest as a route to collectively enhance their physical properties and is the key to the realization of various potential applications of SWNTs.[12] Recently, many efforts have been made toward this purpose using methods such as solvent evaporation,[13−15] electric field,[16] and flow field.[17] The assembled SWNT superstructures achieved so far, however, have been limited to simple ones such as preferentially oriented SWNTs on substrates or nematic SWNTs in bulk; ordered SWNT superstructures with more complex geometries such as one- or multi-dimensional arrays have not been achieved yet.

Amphiphilic molecules, such as block copolymers, exhibit rich phase behavior and have been used extensively as excellent templates for highly ordered nanostructure materials with various architectures.[18−21] Therefore, utilization of the rich phase behavior of block copolymers may provide a general and inexpensive way for fabricating a large variety of self-assembled and highly ordered arrays of SWNTs without going through complicated preparative procedures. The temperature-dependent phase behavior of block copolymers may also provide new

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possibilities of making thermally interchangeable, ordered SWNT arrays, which can lead to the development of thermosensitive hybrid materials. Furthermore, understanding the self-assembly behavior of SWNTs, which are the most representative one-dimensional nanoparticles, in polymeric systems may provide key information for the phase behavior of one-dimensional nanoparticles interacting with surrounding materials, which has not been fully exploited yet. Therefore, this approach should be applicable to designing new self-assembled superstructures of other one-dimensional nanoparticles, such as semiconducting, metallic, or magnetic nanorods which are currently of great interest for various potential applications.

Here, we report thermally reversible, highly ordered one-dimensional lattices and two-dimensional hexagonal arrays of individually isolated SWNTs formed by cooperative self-assembly of functionalized SWNTs and pluronic block copolymers in water. To the best of our knowledge, this is the first demonstration of highly ordered SWNT superstructures made using the self-assembling phase behavior of polymeric systems.

Results and Discussion

The individually isolated and non-covalently functionalized SWNTs (*p*-SWNTs)[22] were fabricated by (1) dispersing SWNTs in water using cationic surfactant, cetyltrimethylammonium 4-vinylbenzoate (CTVB), which has polymerizable counterions, and (2) permanently fixing the surfactant monolayer on the SWNTs by *in situ* free radical polymerization of the counterions, followed by ultracentrifugation and freeze-drying. Small-angle neutron scattering measurements of *p*-SWNTs in D_2O indicate that isolated SWNTs of average diameter, 1.0 nm, are cylindrically encapsulated by polymerized surfactant monolayers whose thickness is about 2.0 nm, resulting in 5.0 nm for the diameter of *p*-SWNTs. Atomic force microscopy measurements indicate that the length of the *p*-SWNTs is ca. 500 nm. A schematic of a *p*-SWNT is shown in Figure 1a. The *p*-SWNTs fabricated in this way are very stable and easily redispersible in water[22] or alcohols[23] by only a few minutes of mild vortex mixing, even after harsh processing such as freeze-drying. The excellent stability and processability of *p*-SWNTs provide a unique opportunity to explore the rich phase behavior of block copolymers as templates to fabricate various self-organized and highly ordered SWNT superstructures in polymeric systems. For efficient homogeneous mixing and systematic incorporation or guided self-assembly of *p*-SWNTs, a polymeric system that exhibits both an isotropic phase and a highly ordered phase depending on temperature is desired. Therefore, the pluronic block copolymer PE6200 ($PEO_{10.5}-PPO_{30}-PEO_{10.5}$) was selected as a model system, which shows an isotropic phase at room temperature and a lamellar phase near $T = 50$ °C at 50 wt % in water.[24]

Mixtures of *p*-SWNT/PE6200/water (10/50/50 by weight) were prepared by mixing *p*-SWNT/water with PE6200 at room temperature. The UV−vis−NIR spectrum of *p*-SWNTs mixed with the PE6200/water system is nearly identical to that of *p*-SWNTs dispersed in water, showing sharp van Hove transition peaks, which indicates that most of the SWNTs in the PE6200/water system exist in an individually isolated form, as they do in water (Figure 1b).[25] The radial breathing mode of the Raman spectrum of the *p*-SWNTs mixed with the PE6200/water system shows a 3.7 cm^{-1} shift to higher frequency compared to that of the bundled SWNTs in water, which further confirms the existence of the debundled and individually isolated SWNTs in the PE6200/water system (Figure 1c).[26]

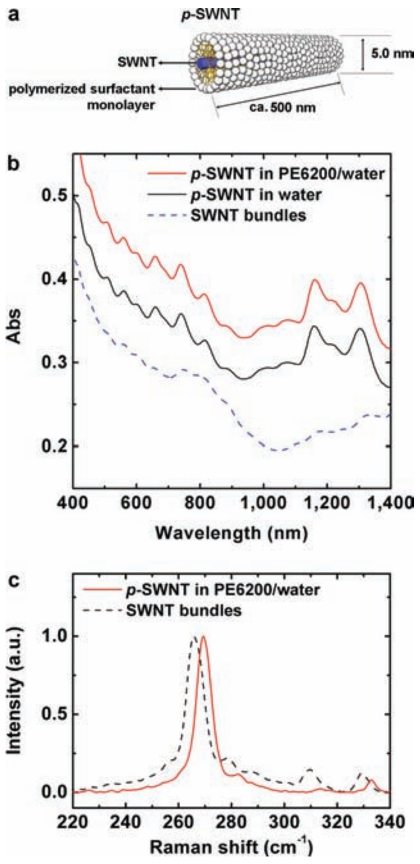

Figure 1. (a) Schematic of isolated *p*-SWNT structure. The SWNT (1 nm diameter) is surrounded by a monolayer of polymerized surfactant (CTVB). (b) UV−vis−NIR spectra of *p*-SWNTs in PE6200/water system (red solid line), *p*-SWNT in water (black solid line), and SWNT bundles in water (blue dashed line). Lines are vertically shifted for visual clarity. (c) Raman shift from *p*-SWNTs in PE6200/water system (red) and bundled SWNTs in water (black). Intensities are normalized to 1 for the peak near 270 cm^{-1} for visual clarity.

Small-angle X-ray scattering (SAXS) measurements of PE6200/water (50/50 by weight) with and without *p*-SWNTs were recorded as the temperature was increased from 20 to 60 °C. The SAXS intensities of the PE6200/water system without *p*-SWNTs show that the PE6200/water exists as an isotropic micellar phase at 20 °C, as indicated by an intermicellar correlation peak, and makes a transition into a lamellar phase at ca. 45 °C, as indicated by the 1:2 ratio of peak positions (Figure 2a).[24] The interlayer spacing of the lamellar structure at 50 °C is 9.5 nm. The relatively weak intensity of the first-order Bragg peak compared to the second-order Bragg peak indicates that there is a subdomain structure in this lamellar

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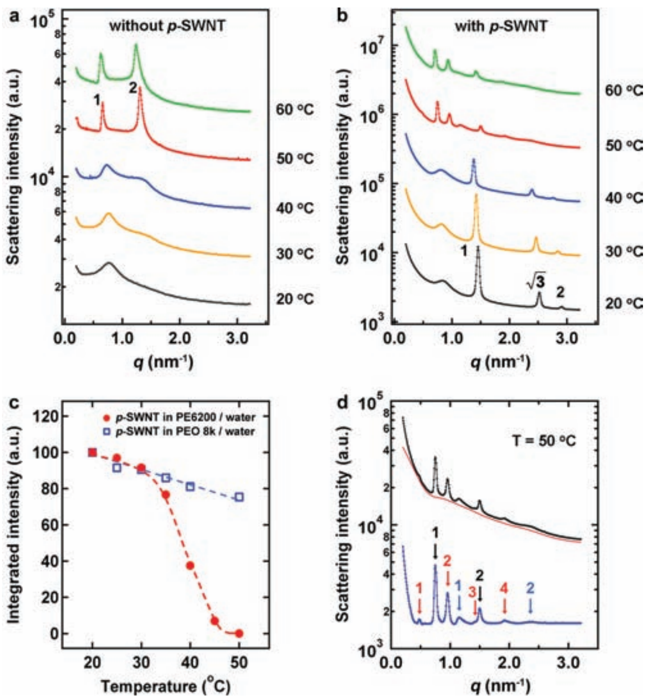

Figure 2. SAXS intensities of (a) PE6200/water (50/50 by weight) and (b) *p*-SWNT/PE6200/water (10/50/50 by weight) at various temperatures (during heating). Scattering intensities are shifted vertically for visual clarity. (c) Integrated intensities of the first-order hexagonal peaks of the *p*-SWNT/PE6200/water system (red solid circles) and the *p*-SWNT/PEO/water system (5/40/60 by weight) (blue open squares). The integrated intensities are normalized to 100 at 20 °C. The dashed lines are guides for the eye. (d) From the scattering intensity at 50 °C (black line), smooth background (red line) has been subtracted to help identify scattering peaks (blue line). The numbers indicate the positional ratio of the scattering peaks belonging to three different groups, indicated by red, blue, and black.

system.[27] Form factor analysis shows that a 2.3 nm water-rich gap exists in the middle of the polar domain of the lamellar structure (with a polar domain thickness of 6.5 nm and an apolar domain thickness of 3.0 nm), which explains the ratio of the Bragg peak intensities (see Supporting Information, Figure S1).

The scattering patterns of the PE6200/water with *p*-SWNTs (Figure 2b) are dramatically different from those of the PE6200/water without *p*-SWNTs. At temperatures between 20 and 40 °C, the PE6200/water with *p*-SWNTs shows very sharp scattering peaks, indicating a highly ordered phase, while the PE6200/water without *p*-SWNTs shows a broad peak, indicating an isotropic micellar phase. At temperatures above 50 °C, where the PE6200/water by itself is in a lamellar phase, a new set of scattering peaks is observed in addition to the lamellar peaks, indicating a new structural order induced by *p*-SWNTs.

The three scattering peaks observed at temperatures between 20 and 40 °C clearly satisfy a $1:\sqrt{3}:2$ ratio of peak positions, indicating a hexagonally ordered structure (see Supporting Information, Table S1). The lattice parameter *a,* the center-to-center distance between nearest-neighboring cylinders of the two-dimensional hexagonal structure at 20 °C, is 5.0 nm, which is consistent with the diameter of *p*-SWNTs (5.0 nm). Since PE6200 by itself does not form any ordered structure at this temperature, the observed scattering pattern clearly indicates that the highly ordered hexagonal arrays are formed by *p*-SWNTs. The formation of the hexagonal arrays of *p*-SWNTs in PE6200/water is understood in terms of a depletion attraction between *p*-SWNTs. When colloidal particles are mixed with homopolymers, a depletion attraction between colloidal particles develops, inducing a cluster of particles.[28] When rod-like particles, such as DNAs[29] or microtubules,[30] are mixed with PEO homopolymers in water, they form hexagonally packed arrays due to the depletion attraction. In fact, SAXS intensities of *p*-SWNTs mixed with PEO homopolymers in water showed hexagonal scattering peaks, indicating hexagonally packed arrays that form due to depletion attraction (see Supporting Information, Figure S2). Although the critical micellization temperature of 50 wt % PE6200 in water is around 20 °C,[24] a large fraction of PE6200 exists in unimeric form near the critical micellization temperature.[31] Therefore, the depletion attraction force, which drives *p*-SWNTs to form hexagonal arrays at 20 °C, is induced by the PE6200 unimers, which have a radius of gyration of 1.3 nm[32] (Figure 3a). Since the diameter of a PE6200 micelle is expected to be ca. 10 nm,[33] which is larger than that of a *p*-SWNT, it is unlikely that the depletion attraction which induces the hexagonal arrays is from PE6200 micelles. This is the first demonstration of highly ordered hexagonal arrays of individually isolated SWNTs in a polymeric system formed via depletion-force-driven self-assembly.

As the temperature increases, the intensity of the hexagonal peaks decreases gradually until ca. 30 °C and then rapidly to zero near 45 °C. This is in stark contrast to the intensity behavior of *p*-SWNTs mixed with PEO homopolymer/water (Figure 2c). While the gradual decrease of intensity for the *p*-SWNT/PEO/water system can be attributed to the reduced depletion force or osmotic pressure with temperature,[34] the rapid decrease of intensity to zero in the *p*-SWNT/PE6200/water system can be understood in terms of the interplay between the depletion force and the hydrophobic interaction of PE6200. As the temperature increases above the critical micellization temperature, the volume fraction of unimers which are responsible for the depletion force decreases, while the volume fraction of micelles increases.[31] In addition, since the increased hydrophobicity of PPO blocks with temperature causes PE6200 to form a lamellar structure, the depletion attraction due to the PE6200 unimers is dramatically reduced, resulting in rapid disintegration of the hexagonal arrays of *p*-SWNTs.

The background-subtracted scattering intensity of *p*-SWNT/PE6200/water at 50 °C is shown in Figure 2d. The whole set of scattering peaks cannot be indexed according to a single conventional morphology, such as lamellae or hexagonally packed cylinders. As indicated in Figure 2d, however, the scattering peaks can be put into three groups (red, blue, and black), all of which have well-defined peak position ratios of 1:2:3:4 or 1:2, indicating three different lamellar-type structures with three different periodicities, 13.1 (red), 5.4 (blue), and 8.4 nm (black) (see Supporting Information, Table S1). Since the surface of *p*-SWNTs is hydrophilic, the *p*-SWNTs are expected to be inserted in the polar region (PEO + water) of the lamellar structure of PE6200/water, increasing the lamellar spacing

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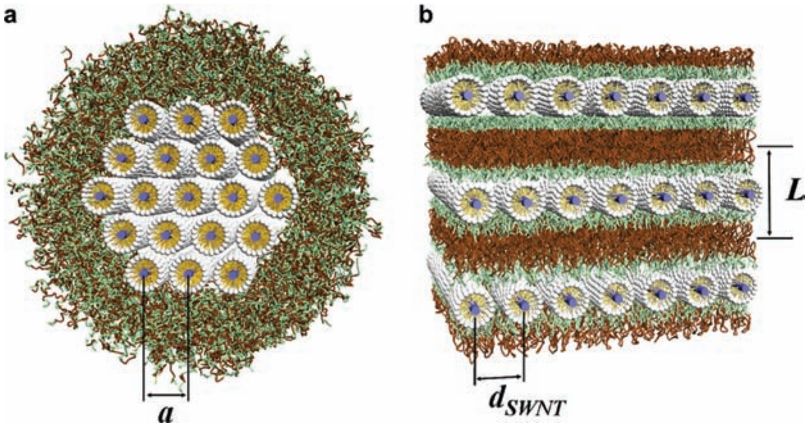

Figure 3. (a) Hexagonal array of *p*-SWNTs formed by depletion attraction in PE6200/water at low temperature. The center-to-center distance between neighboring SWNTs is given by the lattice constant *a* ($a = 5.0$ nm at 20 °C). (b) One-dimensional lattices of *p*-SWNTs intercalated in the polar region of polymeric lamellar structure (PE6200/water). The repeat distance of the lamellar structure is *L*, and the center-to-center distance between neighboring SWNTs in the one-dimensional lattice is d_{SWNT} ($L = 13.1$ nm and $d_{\mathrm{SWNT}} = 5.4$ nm at 50 °C).

accordingly. In fact, the periodic distance (13.1 nm) given by the red peaks is 3.6 nm larger than the lamellar spacing (9.5 nm) of PE6200/water without *p*-SWNTs at 50 °C. If we consider that the PE6200/water without *p*-SWNTs contains a 2.3 nm water-rich gap in the polar region, a gap of 5.9 nm, which provides a sufficient space for just one monolayer of *p*-SWNTs (diameter = 5.0 nm), should exist in the middle of the polar region of the lamellar structure, as indicated by the red peaks. Therefore, the increased periodic spacing of the red peaks strongly suggests that monolayers of *p*-SWNTs are intercalated in the polar regions of PE6200/water lamellar structure. The periodic distance (5.4 nm) given by the blue peaks is close to the diameter of a *p*-SWNT. This indicates that the intercalated *p*-SWNT monolayers are one-dimensional lattices of *p*-SWNTs with spacing comparable to the diameter of a *p*-SWNT. A form factor analysis (see Supporting Information, Figure S3), which successfully reproduced the amplitude ratios of the measured Bragg peaks (red peaks in Figure 2d), confirms the one-dimensional lattices of *p*-SWNTs intercalated in the polar regions of polymeric lamellar structures (Figure 3b). To the best of our knowledge, this type of ordered self-assembly of SWNTs in polymeric systems has not been previously reported. The black peaks, which give a periodic distance comparable to the lamellar spacing of the PE6200/water (50/50) system at 50 °C, are attributed to the lamellar structure of the PE6200/water without *p*-SWNTs coexisting with the lamellar structure intercalated with *p*-SWNT monolayers.[35]

The formation of one-dimensional lattices of *p*-SWNTs intercalated in the middle of polar regions of PE6200/water lamellar structure is understood in terms of entropically driven particle segregation and two-dimensional depletion attraction[36] induced by the two-dimensional nature of the lamellar structure. Experimental[37] and theoretical[38] studies have shown that, when large spherical particles are mixed with block copolymer systems of a lamellar structure, the particles are expected to be concentrated at the center of the domain to minimize the loss of conformational entropy of the polymer chains. Similarly, the concentration of *p*-SWNTs at the center of the polar domain can be understood by the entropically driven phenomenon. The water-rich gap in the middle of the polar region may further enhance this effect. Since *p*-SWNTs are confined in a two-dimensional water-rich space enclosed by two PEO + water layers and PEO chains may protrude into the water-rich space, two-dimensional depletion attraction between *p*-SWNTs can be induced by excluding PEO chains from the space between neighboring *p*-SWNTs. The two-dimensional depletion force makes *p*-SWNTs, residing as a monolayer in the middle of the polar domains, form a one-dimensional lattice with a repeat distance similar to the diameter of a *p*-SWNT.

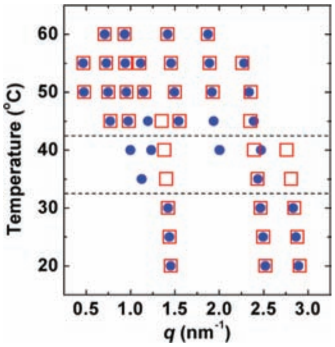

Figure 4. Scattering peak positions of the *p*-SWNT/PE6200/water (10/50/50 by weight) system during heating (red open squares) and cooling (blue solid circles).

The scattering patterns of the *p*-SWNT/PE6200/water system were measured during cooling from 60 to 20 °C and compared with those measured during heating (see Supporting Information, Figure S4). The peak positions of the scattering patterns observed during the heating and cooling cycles almost exactly overlap, except for some hysteresis near the structural transition region indicated by two dashed lines (Figure 4). This clearly

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(35) Since the lamellar structure intercalated with *p*-SWNT monolayers consumes additional water to hydrate *p*-SWNTs and to fill additional gap space induced by the intercalation of *p*-SWNTs, the volume fraction of water in the PE6200/water region without *p*-SWNTs should be lower than that in the PE6200/water (50/50) system, which explains the decrease of the lamellar spacing (from 9.5 to 8.4 nm).

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indicates that the self-assembled ordered arrays of *p*-SWNTs are thermally reversible equilibrium structures.

Conclusion

Thermally switchable and highly ordered one- and two-dimensional arrays of individually isolated SWNTs were fabricated by utilizing the self-assembling phase behavior of PE6200/water. When PE6200/water is in an isotropic phase, two-dimensional hexagonal arrays of *p*-SWNTs are formed due to depletion attraction. When PE6200/water is in a lamellar phase, one-dimensional lattices of *p*-SWNTs intercalated in the polar region of the polymeric lamellar structure are formed due to entropically driven segregation and two-dimensional depletion attraction. These two SWNT arrays are equilibrium structures and thus can be switched from one to the other by changing the temperature. This demonstrates that the phase behavior of polymeric systems provides a new route to fabricate thermally reversible and highly ordered arrays of individually isolated SWNTs. Considering the wide varieties of polymeric phase behavior available, this new approach may open up new opportunities to fabricate highly ordered SWNT superstructures with different architectures, which will lead to the realization of various potential applications of SWNTs. Furthermore, the temperature-dependent self-assembly behavior of SWNTs in polymeric systems may play an important role in developing new, thermosensitive hybrid materials. This approach should also be applicable for other one-dimensional nanoparticles, such as semiconducting, metallic, or magnetic nanorods, which are currently of great interest.

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Supporting Information Available: Description of materials and methods; form factor analysis of measured SAXS data; summary of peak positions and their relative ratios; summary of SAXS intensities of *p*-SWNT/PE6200/water during heating and cooling. This material is available free of charge via the Internet at http://pubs.acs.org.

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