Aligning One-Dimensional Nanomaterials by Solution Processes

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ABSTRACT: One-dimensional nanomaterials, including both nanowires (NWs) and nanotubes (NTs), have been extensively investigated in the decades because of their unique physicochemical properties. Particularly, aligning NWs/NTs into a network or complex micropatterns has been a key issue for its unique integrated functionalities, which enjoy benefits in versatile applications. So far, solution processes remain the most effective strategy to align NWs/NTs, which also bear advantages of mild operation condition and large-scale production. In this perspective, particular attention is drawn to the currently widely used solution coating approaches for aligning NWs/NTs, including the Langmuir–Blodgett film technique, solution shearing approaches, and methods of tri-phase contact line manipulation. We also proposed several perspectives in this field.

1. INTRODUCTION

One-dimensional (1D) nanomaterials including nanowires (NWs) and nanotubes (NTs) bear unique electronic, optical, and magnetic properties for its high aspect ratio and mechanical flexibility, which have been exploited in various applications of optoelectronics,4−9 biological sciences,10−13 energy harvesting,14−17 and chemical sensing,18−22 and thus serve as an attractive research topic in the decades. Particularly, aligned NWs/NTs have shown various integrated properties in both mechanical and electrical regard,23,24 which has been currently widely used in developing flexible electronic devices, such as flexible electrodes,25−28 stretchable electronics,29−31 and e-skin.32,33 In the fabrication of the electronic devices, the precise location and orientation of NWs/NTs are essential and crucial to achieve their advantages of integrated physicochemical properties for the adequate device performance. Therefore, trying to realize highly aligned NWs/NTs in various micropatterns remains a main challenge in building blocks for high-performance micro/nanodevices.

In nature, highly aligned 1D materials have been observed in a centuries-old technique for transporting timber by the river.34 Inspired by this, diverse solution processes such as drop casting,35,36 spray coating,37−40 Langmuir–Blodgett (LB) film,41−44 solution shearing,45−48 external electric/magnetic field,49,50 three-phase contact (TCL) line manipulating,51−54 etc. have been developed to align NWs/NTs into functional patterns. So far, solution processes remain the most effective strategy for aligning NWs/NTs, which also bear advantages of mild operation condition and large-scale production. In this perspective, here, we reviewed solution processes aiming to align NWs/NTs that were widely used in recent years, which were summarized into three categories based on different driving forces (Figure 1): (i) LB technique, where NWs/NTs are aligned as a consequence of anisotropic contraction of the solution film under the external forces from barriers; (ii) solution shearing methods, where directional shearing forces of solution on other medium were used, including the blown bubble approach, blade coating method, spray coating route, and fluid flow-directed technique; (iii) approaches by manipulating the TCL. Each approach has advantages and limitations. Here, both the fundamental principles and the applications were discussed in detail for each category. Finally, future perspectives of aligning NWs/NTs are proposed, which may shed light on the controllable alignment of 1D nanomaterials using solution processes.

2. LB TECHNIQUE

As a general method to align 1D nanomaterials under compression by external force, the LB technique originated from an ancient lumbering operation in North America in the 19th century, in which aligned timbers covered the river surface, forming a spectacular sight (Figure 2a).34 Then, a similar “logs-on-a-river” approach on a nanoscopic level was reported to direct NW assembling, which was called the LB compression technique.44,55 As shown in Figure 2b, in the LB trough, a dispersion of NWs in an organic solvent spreads on the water surface, generating a randomly distributed and loosely packed NW monolayer at the air–water interface. NWs are then compressed slowly by computer-controlled barriers with the surface pressure monitored. The surface pressure is a cognitive factor that affects the assembly process and the final morphology of the NW film in the LB technique, which can be adjusted by mechanically moving one or two barriers on the liquid surface.56

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To better understand the alignment process, thermodynamic analysis and large-scale molecular dynamics theoretical evaluation were taken to reveal the NW assembly mechanism (Figure 2b).47 By comparing the theoretical prediction and the experimental measurement, several stages of the alignment process can be identified. During the initial stage, a low surface pressure was shown, where NWs are packed loosely in the liquid surface with a far distance between each other. As the two barriers get closer, randomly dispersed NWs tend to bundle and align with each other, and meanwhile, the free energy becomes lower because the free space for NWs becomes limited gradually. The surface pressure at this stage is dominated by the conformation entropy due to the still far distance for the short-range interactions such as van der Waals forces until the formation of a well-defined freestanding NW monolayer structure, where the attractive potential from the increased weak molecular interactions dominates. Moreover, large-scale molecular dynamics theoretical evaluation was also introduced to numerically simulate the alignment process. The compression generates densely aligned NWs that are parallel to the trough barriers. The resulting NW film can be transferred from the air–liquid interface onto desired solid substrates by either horizontal or vertical lift-off. By transferring multiple times, an NW network with controllable layers and mesh size can be obtained.27

This versatile and general method has attracted explosive attention worldwide for the major advantage of the ability to align various NWs over large areas with ultrahigh packing density (Figure 2d–g),42,58−60 which can be utilized to fabricate integrated functional micro/nanodevices.61–65 For example, Ag and W18O49 NWs have been manipulated to fabricate an NW network for the fabrication of transparent field-effect transistors (Figure 2h).42 Flexible and transparent electrodes were large-scale-fabricated by co-assembling Ag NWs with Te NWs using the LB technique and then etching away Te NWs, leaving Ag NWs with controllable pitch (Figure 2i).57 Moreover, aligned Ag NW monolayers have been used as excellent substrates for surface-enhanced Raman spectroscopy with large electromagnetic field enhancement factors (Figure 2j).44 Although the technique has been proven to be versatile for aligning various NWs/NTs, there are still some limitations inherent in this assembly method. For example, (i) the reorganization of NWs/NTs during transfer from the air−liquid interface onto desired solid substrates leads to overlapping features and gaps within the dense arrays.64,65 (ii) The aligning process is usually conducted at the air−water interface, which may be inappropriate for some materials and devices that are easily damaged by water.66 (iii) For hydrophilic nanosized building blocks, the surface of the nanomaterials must be functionalized with hydrophobic ligands for the LB experiment, which greatly restricts their future application.63,67,68

3. SOLUTION SHEARING-INDUCED ALIGNMENT

Except for the LB technique, solution shearing is widely used to align NWs/NTs with the parallel direction to that of the shearing force. The shearing force is an unaligned force, which tends to push two parts of the body into opposite directions. The shearing force is often generated by the motion of a fluid or solid against another one. Different types of motion means have been developed, such as the spreading/dewetting of a solution on the surfaces, the mechanical movement of two plates/rolls in opposite directions, and the relative motion of a solution and the surfaces. Under solution shearing force from these motions, NWs/NTs have been aligned for functional patterns and high-performance micro/nanodevices.55 The main viable methods that are currently used in recent years have been reported as the blown bubble, blade coating, spray coating route, and fluid flow-directed techniques.

Blown bubble technique is a simple and efficient alignment method, which consists of (i) preparation of a homogeneous and stable NW/NT suspension, (ii) expansion of the suspension to form a bubble with controlled pressure and expansion rate, (iii) transfer of the bubble film to desired substrates (Figure 3a,b).69 Using this technique, a uniformly aligned NW/NT film on rigid or flexible curved surfaces can be obtained with controllable density and large-area scales (Figure 3c), which can be further used for a field-effect transistor (FET) (Figure 3d). This method offers significant advantages compared to other methods. However, it is limited by both the lack of tight control over the distribution of NWs/NTs and the possible contamination from the polymer coating, which impedes this method for mass production applications.70

Blade coating is a powerful and widely used method for NW/NT alignment, in which the NW/NT solution is sandwiched between a shear blade and a substrate. As the shear blade moves, the meniscus of the solution separates and dries, depositing aligned NWs/NTs on the substrate with the direction of NWs/NTs parallel to that of movement. For better alignment of NWs/NTs, special treatment was performed for the substrates, for instance, alternating patterns of solvent wetting and dewetting regions (Figure 3e)72 or optimizing the substrate temperature (Figure 3g).71 Thus, densely aligned SWCNTs are deposited on the patterned substrate and further used for FET (Figure 3f). Orthogonal aligned Ag NW networks by two-step blade coating are applied...
in constructing quantum dot light-emitting diodes (QLEDs) (Figure 3i). The limitation of this method lies in the pretreatment of the substrates complicating the fabrication process.

Another attractive approach for large-scale and highly aligned NWs/NTs was the spray coating route (Figure 3j). In this alignment process, the NW/NT suspension was pressurized to spray through a nozzle onto a temperature-controlled substrate, including silicon, glass, and flexible substrates. The fast flow of the NW/NT suspension causes solution shearing, resulting in the alignment of NWs/NTs. The alignment with controlled orientation and density can be obtained over a large scale under controlled conditions of the nozzle flow rate, droplet size of the sprayed NW suspension, spray angle, and the temperature of the receiver substrate. This route is promising for mass production, but the aggregation of the nanomaterials in the alignment process is non-negligible.

Microfluidic flow is usually used to align NWs/NTs by combining surface patterning techniques. Micrometer-sized channels are used to confine the fluid flow. When homogeneous suspensions pass through the microfluidic channels, NWs/NTs are deposited on the substrates and aligned along the flow direction driven by solution shearing. The alignment degree of NWs/NTs can be controlled by the flow rate, which is because higher flow rates produce larger solution shearing, resulting in better alignment.
Moreover, the distribution density of NWs/NTs can be regulated by the flow time and the concentration of NWs/NTs in the solution. Through multiple repeating of the fluid-directed alignment process, complex geometry patterns such as layer-by-layer crossed NW/NT network have been obtained. The challenge for the alignment obtained by this microfluidic flow technique is to precisely control the position of the deposited NWs/NTs.

4. ALIGNMENT BY MANIPULATING TCL

Furthermore, the manipulation of solid–liquid–gas TCL of NW/NT solution on the substrates has been developed as another efficient method to align various NWs/NTs, which benefits from surface tension and capillary flow. In an evaporation-induced alignment process, NWs/NTs are aligned around the TCL due to solvent evaporation. The simplest demonstration of the alignment assisted by TCL shift is through the use of an evaporating liquid droplet, where NWs/NTs deposited onto a substrate reorient perpendicular to the direction of the TCL to minimize fluid drag. During evaporation, the liquid moves to the edge of the droplet to compensate the solvent loss caused by evaporation, which is called Marangoni flow. Thus, a liquid flow toward the solvent–substrate–air TCL is generated, which transports NWs/NTs to the evaporation front and aligns them along the direction of the flow, that is, perpendicular to the direction of the TCL (Figure 4a). When the solvent completely evaporates, NWs/NTs are ideally expected to align along the radial direction of a circle (Figure 4b). In a word, solid–liquid–air TCL is crucial for the assembly because pinning of the contact

Figure 3. Alignment of NWs/NTs under solution shearing. (a–d) Blown bubble approach (Copyright 2007 Nature Publishing Group). (a) Schematic depiction of blown bubble process to fabricate an aligned NW/NT film. (b) Photographs of directed bubble expansion process. (c) Images of the aligned NWs by this method, which was transferred to a large-scale Si wafer (150 mm). (d) Application of Si NW alignment induced by blown bubble approach for the FET device. (e, f) Solution shearing technique (Copyright 2015 Wiley-VCH Verlag GmbH & Co. KGaA). (e) Schematic illustration of SWCNT aligned by solution shearing. (f) AFM images of the aligned SWCNTs on hydrophilic–hydrophobic patterned substrates. (g–i) Blade coating approach (Copyright 2016 American Chemical Society). (g) Schematic of a viscous force-assisted blade coating method to assemble the aligned Ag NW networks. (h) SEM image of the aligned Ag NW networks with their electrothermal properties in the inset. (i) Schematic structure and photograph of fabricated QLEDs based on the obtained Ag NW networks. (j) Schematic of the spray coating process and the resulting Si NW alignment (Copyright 2012 American Chemical Society).
line is the prerequisite to induce the outward capillary flow. Various approaches have been developed to achieve TCL manipulation for NW/NT alignment.

Through the observation of contact line deposition and alignment of NWs in an evaporating droplet (Figure 4d,e), Yang et al. programmed the stick–slip motion of the solvent contact line during dip coating to align NWs over large areas (Figure 4c). By this programmed dip coating, selective positioning of NW arrays with controllable density and spacing can be achieved (Figure 4f,g). Recently, Ko et al. fabricated...
highly conductive and transparent electrodes using a capillary printing technique (Figure 4k), in which highly aligned Ag NW arrays were produced by the anisotropic dragging of Ag NW solutions via a pretreated PDMS stamp (Figure 4j,h). The key technologies of this strategy are the physical confinement to prealign Ag NWs in the PDMS nanochannels and the subsequent alignment of NW by controlling the TCL, which exerts capillary forces on the meniscus-trapped Ag NWs due to solvent evaporation (Figure 4i). In addition, very recently, our group has achieved aligning Ag NWs by facile bioinspired directional liquid transfer to control the TCL (Figure 4l,o). The conical fiber array (CFA) was used to guide Ag NW solution directional transfer under cooperative effect of the Laplace pressure difference, asymmetrical retention force, and gravity (Figure 4m). The receding TCL of the Ag NW solution can be finely controlled by CFA-guided directional liquid transfer during the dewetting process (Figure 4n), which enables Ag NW alignment at the edge of TCL.

5. CONCLUSIONS AND OUTLOOK

In conclusion, we reviewed the recent progresses on aligning NWs/NTs by solution processes, including the LB film technique, solution shearing approaches, and methods of TCL manipulation. These strategies enjoy advantages of mild experimental conditions and large-scale production. Aligned NWs/NTs, which are normally prepared by solution processes, have shown versatile perspective applications in many fields: (1) Aligned NWs/NTs are helpful in making a high-performance FET, which is the building block and active element of integrated circuits for signal amplification and readout. For example, it has been reported that the aligned CNTs may exhibit higher mobility and on-current density due to its lower tube-to-tube junction resistance compared with that of random-network CNTs. Moreover, aligned metallic NWs/NTs can be considered as the gate, source, and drain electrodes for the flexible FET. (2) Aligned NWs/NTs serve as important building blocks in making flexible electronic devices when conductive NWs/NTs were used, such as artificial skins, wearable devices, and stretchable displays. Compared to conventional rigid wafer-based electronic conductors, the conductive NW/NT film can remain continuous and maintain high conductivity under iterative deformation by bending, stretching, and stressing. Particularly, the aligned NWs/NTs show anisotropic conductivity, which endows the directional recognition of flexible electronic devices. Under iterative forces from two directions, spaces between the aligned NWs/NTs are periodically changed in head-to-head or side-by-side direction, resulting in the continuous and relative changes of the resistance of the aligned NW/NT film. This may cast a new light on the directional force sensing of the artificial skins and wearable devices. (3) Aligned NWs/NTs can be used in making electrochromic devices. For example, aligned WO$_3$ NWs can be used in making smart windows by a reversible electrochemical process. Compared with disordered NW/NT networks, aligning NWs/NTs can achieve precise deposition of NWs/NTs. Meanwhile, by adjusting the density of NWs/NTs, the balance of the optical transmittance and the conductivity of the aligned NW/NT film can be simply implemented. Therefore, in the preparation process, the uniformity in large area and the suitable density of NWs/NTs are always desirable. In addition, aligned NWs/NTs can serve as conductive micropatterns by which localized electrodeposition can be realized. Despite various advantages of solution processes in aligning NWs/NTs, solution processes still suffer from limitations, including the reorganization of NWs/NTs during transfer, nonuniformity over a large area, and difficulty in micropatterns without templates. Here, we proposed several perspectives in aligning NWs/NTs: (1) trying to accurately control the alignment and orientation of NWs/NTs at macroscale; (2) trying to realize the large-area uniformity for the NW/NT alignment; (3) trying to achieve the NW/NT alignment with various densities; (4) aligning NWs/NTs without an external field, special operation skills, and complex deposition parameters. Taken together, the solution process techniques remain the most efficient approaches to align NWs/NTs for the future applications in diverse electronic devices.

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Notes

The authors declare no competing financial interest.

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