Fabrication of One-Dimensional Organic Nanomaterials and Their Optoelectronic Applications

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This paper reviews the recent research and development of one-dimensional (1D) organic nanomaterials synthesized from organic semiconductors or conducting polymers and their applications to optoelectronics. We introduce synthetic methodologies for the fabrication of 1D single-crystalline organic nanomaterials and 1D multi-component organic nanostructures, and discuss their optical and electrical properties. In addition, their versatile applications in optoelectronics are highlighted. The fabrication of highly crystalline organic nanomaterials combined with their integration into nanoelectronic devices is recognized as one of the most promising strategies to enhance charge transport properties and achieve device miniaturization. In the last part of this review, we discuss the challenges and the perspectives of organic nanomaterials for applications in the next generation soft electronics, in terms of fabrication, processing, device integration, and investigation on the fundamental mechanisms governing the charge transport behaviors of these advanced materials.

Keywords: Organic Nanomaterials, Organic Electronics, Organic Semiconductors, One-Dimensional Nanomaterials, Single Crystals, Organic Nanowires, Optoelectronics.

1. INTRODUCTION
Organic semiconducting materials have attracted great interest because of their various advantages such as large-scale synthesis, solution processability, and simple electronic tunability by molecular design. Furthermore, their compatibility with flexible and lightweight substrates enables their versatile implementation to the electronic circuits, energy and bio-related applications.¹⁻³ In particular, organic semiconductors based on π-conjugated molecules can be easily transformed into one-dimensional (1D) nanostructures that can potentially exhibit novel optical, electrical, and chemical properties.⁴⁻⁶ Examples of these assemblies include nanowires (NWs), nanorods, nanoribbons, nanobelts, whiskers, and nanotubes. In general, 1D organic nanostructures can be fabricated with various methods including solution-phase synthesis, vapor-phase synthesis, template-assisted synthesis, electrospinning, nanolithography, soft lithography, and so on. In particular, 1D nanostructures based on organic semiconductors have a large surface-to-volume ratio compared to bulk materials and possess a highly crystalline nature that is beneficial to efficient charge transport. To date, 1D organic nanomaterials based on organic semiconductors and conducting polymers have shown great potential in diverse applications covering organic light-emitting diodes,⁷ field-effect transistors,⁸⁻¹¹ phototransistors,¹²,¹³ photovoltaic.
cells, nanoscale lasers, memory devices, and sensors. To integrate 1D organic nanostructures into device applications, they need to be well-aligned and assembled in desired positions.

In addition to the synthesis of conventional single-component 1D nanostructures, the concepts of multi-component nanomaterials such as p–n junctions, core–shell NWs, composites and multi-shape nanostructures such as porous, branched structures, hollow structures have been recently suggested. Progress in nanoscale heterostructures is important for optoelectronic applications as well as for understanding their charge transport mechanisms as they typically exhibit different optoelectronic properties due to the effective functional interplay between two different materials and their unique structural effects.

This review will focus on the recent advances on the fabrication of 1D organic nanomaterials, the functionalization into multi-shape and multi-component nanostructures, and their optoelectronic applications. Self-assembly of small molecules or polymers can lead to the formation of organic 1D nanostructures in solution or in vapor phase. Template-assisted fabrication methods can also contribute

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to the growth of 1D nanostructures in both phases. In the following section, an electrospinning method with the applications of electrosprun fibers will be discussed. Furthermore, this review will address advantages of optoelectronic devices based on single-component or multi-component nanomaterials in comparison with bulk and thin-film based electronics. Finally, concluding remarks about 1D organic nanomaterials and their future perspectives in optoelectronic applications will be discussed.

2. FABRICATION OF 1D ORGANIC NANOMATERIALS

2.1. Self-Assembly in Solution Phase

Self-assembly of π-conjugated molecules into 1D nanostructures is generally dependent upon the balance of the π–π interaction of π-conjugated backbone and the hydrophobic interaction between alkyl side chains. Typically, 1D organic nanostructures can be successfully obtained via strong co-facial π–π stacking interactions, while reducing the unfavorable lateral growth driven by hydrophobic interactions between alkyl side chains.25 Strong intermolecular coupling between the π-stacked molecules provides an efficient pathway for charge transport along the long axis of NW. 1D organic nanostructures can be synthesized in various forms such as wires, rods, ribbons, tubes, and belts via solution-phase self-assembly.

Conjugated organic semiconducting materials with low solubility in organic solvent can self-assemble into 1D structures via recrystallization. A representative n-type organic semiconductor, N,N′-bis(2-phenylethyl)perylen-3,4,9,10-tetracarboxylic diimide (BPE-PTCDI) is sparingly soluble in a common organic solvent such as o-dichlorobenzene and benzonitrile at room temperature, whereas it becomes soluble upon heating with refluxing. BPE-PTCDI molecules tend to self-assemble into needle-like 1D structures via strong π–π interactions of π-conjugated backbones upon cooling the hot, homogeneous solution. Furthermore, the diameter and the length of BPE-PTCDI wires can be tuned across a wide range by controlling the cooling speed and the nucleation density tunable by the non-solvent (i.e., methyl alcohol) mixing ratio. The size of BPE-PTCDI wires can be easily tuned, ranging from several tens of nanometers to several micrometers in diameter and several tens of micrometers to several millimeters in length (Fig. 1(a)). The structural characterization with transmission electron microscopy (TEM) revealed that BPE-PTCDI molecules self-assemble into 1D nanostructures with the π-planar distance of 3.4 Å (Figs. 1(b) and (c)).11

The above-mentioned, non-solvent-mediated recrystallization method is also applicable to the preparation of highly conjugated π-channel organic semiconductors such as hexathiapentacene (HTP).26 Furthermore, 1-cyano-trans-1,2-bis-(3′, 5′-bis-trifluoromethyl-biphenyl)ethylene (CN-TFMBE) molecules can self-assemble in a mixture solvent of o-dichlorobenzene and methyl alcohol via recrystallization spin coating or drop-casting into highly fluorescent 1D nanomaterials, which is driven by the interactions between cyano-stilbene backbone and terminal –CF₃ units.26,27 The CN-TFMBE NWs were synthesized with diameters around 100 nm and showed enhanced fluorescence emission in the solid state, as shown in Figures 1(d) and (e). The strong intermolecular π–π stacking was induced by the planarization of the CN-TFMBE owing to the CN group and CF₃ end groups (Fig. 1(f)).

In the case of organic semiconducting molecules, which are highly soluble in organic solvents, 1D nanostructured self-assemblies can be prepared by transferring the molecules from a good solvent to a non-solvent, followed by rapid dispersion.29 This method refers to rapid solution dispersion method. The propoxylethylperylen-3,4,9,10-tetracarboxylic-3,4,9,10-diimide (propoxylethyl-PTCDI) molecules have sufficient solubility in hydrophobic solvents such as chloroform, due to the flexibility of propoxylethyl side chain. The concentrated solution can readily self-assemble to form nanobelt crystals upon injection of a small amount of solution into methanol.29 Similarly, injection of a small volume of concentrated 6,13-bis(triisopropylsilyl)ethynyl pentacene (TIPS-PEN)/toluene solution into acetoneitrile can lead to the formation of TIPS-PEN microribbon through the self-assembly on the basis of rapid solution dispersion method (Figs. 1(g)–(i)).9

When conjugated molecules have different solubility in two organic solvents, self-assembly can be induced at the interface between two different solvents through binary phase transfer. The solvent interface is typically formed by the difference in the polarity and the density of two solvents. PTCDI molecules with alkyl side chains (PTCDI-Cn) can be transformed into 1D nanostructures via the self-assembly at the methyl alcohol/chloroform interface.8 Moreover, solvent vapor diffusion can be used to form 1D organic single crystals. Lv et al. reported the organic single crystal of the tetrachlorinated diperylene bisimide (C₁₂₋₄Cl₂PBI) grown by solvent vapor diffusion method.30 The C₁₂₋₄Cl₂PBI solution dissolved in toluene was prepared in a small vial and exposed to methyl alcohol vapor. The nanoribbons with a diameter of around 2 μm were obtained after 3 days.

Moreover, Goto et al. reported a novel method to fabricate organic single-crystal arrays with a large-scale from solution-phase synthesis.31 They prepared aligned organic single-crystal arrays by controlling the supersaturation region of 3,9-bis(4-ethylphenyl)-peri-xanthenoxanthene (C₅₋₂Ph-PXX) solution. In the specific micropatterns, the supersaturation of organic solution could be changed locally, leading to the formation of single crystals (Fig. 2). This simple method allowed the formation of the aligned organic single crystal arrays. The careful control of the micropatterns may provide the formation of 1D single crystals.
A wide range of approaches have also been utilized to prepare 1D self-assembled crystalline polymer nanostructures. Poly-(3-alkylthiophene) (P3AT) NWs could be obtained by the thermally induced crystallization of P3AT in a poor solvent.32 The crystallization of π-conjugated polymers, which contain electron-rich backbones and aliphatic chains, is usually prohibited by the entangled chain conformations.5 Most of the related reports have focused on only the fabrication of nanostructured P3ATs because of their strong self-organizing characteristics.

Highly crystalline 1D polymer NWs with low solubility in organic solvent can be synthesized in a dilute solution (0.05–1%). By heating the mixture of highly diluted P3AT solution with poor solvent, followed by a cooling process, conjugated molecules can be stacked via π–π interaction between aromatic backbones without unfavorable stacking from aliphatic chains. P3AT NWs can be obtained in various conditions; controlling the length of alkyl chain, molecular weight, concentration, crystallization temperature, solvent, and regioregularity of polymer materials in an initial good solvent to a marginal or poor solvent.5

Kiriy et al. reported the formation of NWs by the aggregation of regioregular head-to-tail P3ATs by using a selective solvent, hexane, which is a good solvent for alkyl chains, but a poor solvent for polythiophene backbones.33 The addition of hexane into the chloroform solution of P3AT induced an ordered aggregation of the P3AT main-chain driven by a solvophobic interaction. Kim et al. reported a self-seeding method of poly(3-hexylthiophene) (P3HT) with a dilute solution in chloroform to fabricate 1D microwires (MWs).34 Slow cooling the solution from 40 °C to 10 °C over three days created crystalline seeds in solution to facilitate the growth of P3HT MWs in a solution phase. Figure 3(a) shows the polarized OM image and SEM images of 1D single-crystalline P3HT MWs. The width and length of the wires were approximately 1–3 μm and 30–500 μm, respectively. The molecular and crystallographic structures of P3HT chains in a single-crystal MW are depicted in Figure 3(b).

Aside from P3AT single crystalline NWs, several different semiconducting polymers can be utilized to fabricate 1D nanostructures in solution phase. Dong et al. fabricated highly crystalline NWs using poly...
Figure 2. (a) Schematic diagram of the solubility-supersolubility relationship. Points ABC represent three states of solution at the same temperature with different solution concentrations; point A is a stable state, where crystallization does not occur. As the concentration increases over solubility curve, crystallization occurs at points B and C. The spontaneous crystallization can occur at point C because nucleation occurs only in a supersaturated solution. (b) Schematic diagram of crystal growth in the nucleation control region. (c) Two regions of the micropattern; nucleation control region and growth control region (up) and the schematic growth model (bottom). (d) Polarized OM image of well-aligned 12 × 32 arrays of C_{2}Ph-PXX films. Reprinted with permission from [31], O. Goto, et al., Adv. Mater. 24, 1117 (2012). © 2012, Wiley-VCH.

(para-phenylene ethynylene) derivatives with thioacetate end groups (TA-PPE) via slow self-assembly from a dilute solution (0.05–1.0 mg ml$^{-1}$) under a volatile solvent pressure in a closed jar. Slow evaporation of the solvent could induce self-assembly of the polymer molecules with strong intermolecular interactions. Large-area and well-defined TA-PPE NWs were obtained through the self-assembly of TA-PPE in tetrahydrofuran (THF)

Figure 3. (a) Polarized OM image of 1D single-crystalline P3HT MWs and SEM images of single P3HT MW with a side-view image showing the rectangular cross-section with well-defined facets. (b) Molecular and crystallographic structures of P3HT chains with schematic representation of P3HT chains into single-crystalline MWs along the π–π stacking direction. (a) and (b) Reprinted with permission from [34], D. H. Kim, et al., Adv. Mater. 18, 719 (2006). © 2006, Wiley-VCH. (c) SEM images of the growth processes of the self-assembled TA-PPE NWs. (d) Schematic diagram of possible molecular packing in the 1D TA-PPE NW and the SAED pattern of an individual TA-PPE NW. (c) and (d) Reprinted with permission from [35], H. Dong, et al., J. Am. Chem. Soc. 131, 17315 (2009). © 2009, American Chemical Society.

2.2. Self-Assembly in Vapor Phase
Physical vapor transport (PVT) method has been frequently adopted to grow highly crystalline nanostructures
of organic semiconductors that are insoluble in common organic solvents. The main advantage of this method is that the direct growth on any substrates can be realized with less contamination. In a typical PVT process, quartz tubes are loaded in a furnace with a temperature gradient. Then, organic semiconductors are sublimated at the high-temperature region and the vaporized molecules are carried by the flow of inert gas to the lower temperature region. Then, the nucleation of the crystal growth takes place at the lower temperature region, eventually leading to the formation of highly purified crystals.

High-purity single crystals can be obtained from the PVT method without unintentional doping, which is generally caused by solvent molecules or other impurities. Moreover, this method can be employed to induce the direct growth of highly crystalline organic semiconductor nanomaterials onto a desired substrate. Similarly, the deposition of thin films or crystals by the condensation of vaporized target materials onto surfaces is defined as the physical vapor deposition (PVD) method. Because single crystals fabricated in a vapor phase are free of defect or impurity, this method provides great potential to fabricate nanostructures for investigation on their intrinsic electrical and optical properties of organic nanomaterials.

Recently, NWs or nanostructures based on small molecule organic semiconductors have been frequently fabricated in the vapor phase. Vertically aligned NWs were successfully fabricated in the vapor phase from 1,5-diaminoanthraquinone (DAAQ). The NWs could be grown on the sharp tip with high surface energy as shown in Figures 4(a)–(d).

Furthermore, fabrication of multi-component nanostructures is available via the PVT method. Yao and co-workers fabricated vertical arrays of $p$–$n$ junction NWs based on a $p$-channel semiconductor, copper phthalocyanine (CuPc), with an $n$-channel semiconductor, 5,10,15,20-tetra(4-pyridyl)-porphyrin ($\text{H}_2\text{TPyP}$), via the one-step PVT process (Fig. 4(e)). The $\text{H}_2\text{TPyP}$ nanostructures could provide nucleation sites for the growth of CuPc crystals before the formation of heterojunction structures. The strong $\pi$–$\pi$ stacking interactions between the two $\pi$-conjugated systems ensured good interfacial connection.

![Figure 4](image.png)

**Figure 4.** (a) Scheme of vapor phase self-assembly for synthesis of DAAQ nanowires (NWs), (b) SEM image of vertically aligned DAAQ NWs, (c) TEM image of a single NW with the selected area diffraction pattern (inset), and (d) DAAQ NWs grown on a tungsten tip. (a)–(d) Reprinted with permission from [39], Y. S. Zhao, et al., *ACS Nano* 4, 1630 (2010). © 2010, American Chemical Society. (e) Schematic of $p$–$n$ heterojunction crystal growth in the one-step PVT process based on CuPc-$\text{H}_2\text{TPyP}$. Reprinted with permission from [40], Q. H. Cui, et al., *Adv. Mater.* 24, 2332 (2012). © 2012, Wiley-VCH. (f) TEM image with the corresponding selected area diffraction (SAED) patterns, and optical images of single nanoribbon based on F$_6$CuPc-CuPc. Reprinted with permission from [41], Y. Zhang, et al., *J. Am. Chem. Soc.* 132, 11580 (2010). © 2010, American Chemical Society.
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at the junction part. Briseno and co-workers reported the fabrication of F$_16$CuPc-CuPc nanoribbons (Fig. 4(f)) and found that the selective growth behaviors of vaporized molecules into crystals are strongly dependent upon the similarity in molecular structures and lattice parameters at the interfacial basal planes.$^{41}$

Furthermore, dendritic organic heterojunctions with aluminum tris(8-hydroxyquinoline) (Alq$_3$) MW trunks and DAAQ NW branches were prepared with the two-step PVD growth process.$^{42}$ In this system, the Alq$_3$ MWs were used as seeds for the vertical growth of DAAQ NWs. Similarly, Yan et al. reported the site-selective growth behaviors of 4,4′-bis(phenylethynyl)anthracene (BPEA) NWs using the PVD method.$^{43}$ The condensation of vaporized BPEA molecules took place on the sharp tips of silver NWs.

Park et al. demonstrated a selective preparation method of pentacene crystals into 1D wires and two-dimensional (2D) disks via a vaporization-condensation-recrystallization (VCR) process, which can induce the growth of organic crystals in various geometries by controlling the precursor temperature (Fig. 5(a)).$^{44}$ In the VCR process, 1D wires were obtained at a higher temperature (Fig. 5(b)), while 2D disk crystals were obtained at a lower temperature (Fig. 5(c)). The crystal-plane-dependent photoluminescence (PL) properties of pentacene 1D wires and 2D disks revealed a correlation between the arrangement of pentacene molecules and optoelectronic performance (Figs. 5(f)–(n)). Pentacene 1D wires possessing (010) and (001) planes exhibited strong PL to the (010) plane when the light beam was normal to the (010) plane, while PL was not detected with regard to the (001) plane. Similarly, pentacene 2D disk crystals having only (001) crystal plane did not show the PL property.

2.3. Template-Assisted Synthesis

Template-assisted synthesis has been adopted to prepare 1D organic nanostructures composed of the oligomer/polymer materials with less conjugated molecules in the backbone. The two kinds of templates are broadly defined as hard templates and soft templates. Particle track-etched membranes such as polycarbonate membrane are a common hard template for the synthesis of 1D nanostructures. The diameters of nanostructures can be easily modified by controlling the pore size of the template adopted. Anodic aluminum oxide (AAO) membranes and mesoporous silica also exemplify hard templates.$^{45-47}$

A broad range of techniques have been attempted to fill the pores of templates. Among them, vapor deposition polymerization (VDP) based on hard templates is one of the simplest ways to fill the pores of AAO membrane with polymeric materials. The diameter and length can be easily tuned by varying the type of template. It is also possible to prepare NW arrays with tailored architecture via control of the pore density. Furthermore, the wall thickness of nanotube can be easily controlled in nanometer scale by injecting different amounts of monomers (Fig. 6). By using two different monomers, multi-layered nanotubes can also be fabricated. Capillary force between monomer and template enables to realize fabrication of nanotubes with desired thicknesses and 1D nanostructures.$^{49}$

Figure 5. (a) Schematic diagram of crystal growth of pentacene 1D wires and 2D disk crystals via the VCR process. (b) 1D pentacene wires fabricated at a relatively higher temperature region (T = 350 °C) region. The SAED patterns of (d) 1D wire and (e) 2D disk pentacene crystals. (f) TEM, (g) PL, and (h) SAED pattern images of 1D pentacene wire which possesses (010) plane parallel to substrate. (i) TEM, (j) PL, and (k) SAED pattern images of 1D pentacene wire which have (001) plane parallel to substrate. (l) TEM, (m) PL, and (n) SAED pattern images of 2D disk pentacene with (001) plane parallel to substrate. All of the insets are the corresponding TEM images. Reprinted with permission from [44]. J. E. Park, et al., Angew. Chem., Int. Ed. 51, 6383 (2012). © 2012, Wiley-VCH.
Figure 6. TEM image of polypyrrole (PPy) nanotubes via the VDP method. Thickness of the PPy nanotubes can be controlled by changing the injection amount of monomer as (a) 0.07 ml, (b) 0.14 ml, (c) 0.21 ml. (a)–(c) Reprinted with permission from [48], J. Jang and J. H. Oh, Chem. Commun. 882 (2004). © 2004, Royal Chemical Society. (d) Multi-cell nanotubes based on PAN/rhodamine B/PAN coaxial nanotubes structure by using the VDP method. (e) TEM images and (f) confocal microscope image of the fabricated PAN/rhodamine B/PAN nanotube. (d)–(f) Reprinted with permission from [49], K. J. Lee, et al., Chem. Mater. 18, 5002 (2006). © 2006, American Chemical Society.

Lee et al. fabricated organic NWs by stacking small molecules such as pyrene and 1,4-bis(2-(5-phenyloxazolyl)]benzene in the pores of the AAO membrane. They introduced organic molecules into the pores of the AAO membrane via sublimation. Strong π–π interaction between organic dyes makes it possible to maintain their 1D structure even after removing the AAO template.50

As soft templates, assemblies of block copolymers are widely used to synthesize various nanomaterials. The morphology of block copolymer assemblies can be modified by controlling the composition of polymers and their molecular weight.51 Moreover, surfactants,52 liquid crystals,53,54 and polymer nanofibers55 have been used as soft templates.

Moreover, Alam et al. demonstrated a novel and versatile method to fabricate NW arrays based on 5,6,11,12-tetraphenylnaphthacene (Rubrene), Alq3, and [6,6]-Phenyl C61 butyric acid methyl ester (PCBM).56 Figure 7(a) shows the illustration of the fixed-angle centrifugation-assisted growth method. The AAO template was placed in the bottom of organic solution and organic NWs could be obtained by the centrifugation of the solution together with AAO template, as shown in the SEM images in Figures 7(b)–(e).

2.4. Electrospinning

Electrospinning is a straightforward and cost-effective method to fabricate nanofibers from diverse polymer solutions or melts under high electric field. Although the first patent mentioning electrospinning had been reported as early as the 1900s,57–59 over the last two decades a broad range of research has been reported to obtain 1D nanomaterials such as nanofibers, nanotubes, and nanobelts using electrospinning due to the recent increasing attention on nanomaterials.

The basic instrumental setup for electrospinning is as follows:

(1) prepare a certain polymeric solution,
(2) deliver the polymeric solution with a desired feed rate through the needle,
(3) apply the high voltage onto the solution and
(4) collect the products in the form of nanofibers.

Numerous experimental parameters should be considered for precise control of nanofiber quality, such as solution parameters (including viscosity, surface tension, types of polymers, concentration, and conductivity) and external...
parameters (applied voltage, feed rate, type of collector, distance between needle to collector and environmental conditions).

When high voltages are applied onto the solution, the surface charge will be accumulated on the solution droplet. Because there is certain limitation on the maximum surface charge per surface area (such as Rayleigh limitation), the droplet explosion will occur in order to increase their surface area as increasing surface charges. When the polymers dissolved in the solution contain high enough chain entanglements, the droplet will be elongated rather than exploded to increase their surface area, resulting in fiber thinning. These fiber thinning is the main factor to obtain nanosized fibers using electrospinning. After jet-launching, the spun-fiber will be discharged during the flying onto the collector, implying relaxation of the elongation force of the thin fiber. If the jet fibers were not fully dried before losing the surface charges, surface tension of the solution will again become the main factor to decide the nanofiber quality, and bead-on-a-string morphology can be obtained. Therefore, one can say that electrospinning is the result of the competition between the elongation force and the surface tension of the polymeric solutions.60

As mentioned previously, enough entanglement of the polymeric chain is the decisive requirement for electrospinning. Therefore, there are intrinsic limitations to prepare 1D nanomaterials composed of low-molecular weight organo-electronic materials. Polymeric materials such as conducting polymers should be considered for 1D nanomaterials from electrospinning. However, conducting polymers have low solubility to get enough chain entanglements in general. In addition, a high degree of crystallization of conducting polymers or organo-electronic materials is required for better performance in electronic devices, but the rapid crystallization can result in needle blocking during the electrospinning. Therefore, in general, it is difficult to obtain 1D organic nanomaterials for electronic applications using electrospinning.

Several additional modifications on conventional electrospinning should be followed to obtain 1D nanomaterials for electronics and optoelectronics. Chang et al. reported the preparation of PVDF (a piezo-electric polymer) nanofibers using near-field electrospinning.61 In conventional electrospinning, the product is typically in the form of a nanofiber web because of whipping instability. In near-field electrospinning, the spun fiber is rapidly collected prior to the whipping instability, so that aligned nanofibers can be obtained. They said that the aligned spun-fiber contained a high degree of β-crystals which is important for piezo-properties because of the electrical stretching during the electrospinning, resulting in high performances in polymer-based nanogenerators.

Figure 8 illustrates typical examples of preparing semiconducting nanofibers using electrospinning. The fluorescence micrographs of emitting electrospun fibers are presented with the molecular structures for the semiconducting polymers: poly[(9,9-dioctylfluorenyl-2,7-diyld-alt-co-(9-hexyl-3,6-carbazole)] (Fig. 8(a)), poly[(9,9-dioctylfluorenylene-2,7-diyld-co-(1,4-diphenylenevinylene-2-methoxy-5-f2-ethylhexylxyg-benzene)] (Fig. 8(b)), poly[(9,9-dioctylfluorenyl-2,7-diyld-co-(1,4-benzof2,10,3g-thiadiazole)] (Fig. 8(c)), and poly[(2-methoxy-5-(2-ethylhexylxyg-1,4-(1-cyanovinylene phenylene))-co-f2,5-bis(N,N0-diphenylamino)-1,4-phenyleneg] (Fig. 8(d)), respectively.62

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Figure 8. Fluorescence microscopy images of semiconducting polymer nanofibers fabricated by electrospinning with molecular structures of the polymers. NWs based on (a) poly[(9,9-dioctylfluorenyl-2,7-diyld-alt-co-(9-hexyl-3,6-carbazole)], (b) poly[(9,9-dioctylfluorenylene-2,7-diyld-co-(1,4-diphenylenevinylene-2-methoxy-5-f2-ethylhexylxyg-benzene)] (c) poly[(9,9-dioctylfluorenyl-2,7-diyld-co-(1,4-benzof2,10,3g-thiadiazole)] (d) poly[(2-methoxy-5-(2-ethylhexylxyg-1,4-(1-cyanovinylene phenylene))-co-f2,5-bis(N,N0-diphenylamino)-1,4-phenyleneg]. Reprinted with permission from [62], F. Di Benedetto, et al., Nat. Nanotechnol. 3, 614 (2008). © 2008, Nature Publishing Group.
The diameter of the electrospun fiber can be tuned by controlling viscosity, surface tension, conductivity of the polymer solution, electric field, collection distance, and so on. By varying the composition of materials, multi-component or multi-shape nanofibers can be easily fabricated via electrospinning. Nanofibers of binary blends of two polymers can be produced by electrospinning with the mixed solution of desired materials. For fabricating multi-shape nanofibers such as porous nanofibers, both methods of inducing the phase separation phenomena or collecting electrospun nanofibers, followed by selective removal of one component, can be utilized. After electrospinning of the mixture of Poly-L-lactide (PLLA) and poly(vinyl pyrrolidone) (PVP) solution, either PLLA or PVP can be selectively removed, and then porous nanofibers can be obtained (Fig. 9(a)).63 PLLA in dichloromethane solution can be used to fabricate porous PLLA nanofibers,24–64 due to the use of a volatile solvent such as dichloromethane, as shown in Figure 9(b). Furthermore, porous fibers can be used as templates for the formation of various tubes or nanostructures by removing the fiber templates under annealing at elevated temperatures. Moreover, Figure 9(c) represents hollow nanofibers produced by co-spinning two immiscible liquids with a coaxial, two-capillary spinneret, followed by selective removal of the core components. Electro co-spinning of PVP in mineral oil with ethanol and titanium tetraisopropoxide liquids was prepared to fabricate hollow nanofibers from composite containing amorphous TiO₂ and PVP. Octane treatments on the electro co-spun fibers can induce extraction of oily cores and fabricate hollow nanofibers.65


Jeong et al. recently described an interesting method to prepare P3HT nanofibers using core–shell electrospinning.68 They introduced additional solvent into the shell to prevent needle blocking, locating P3HT/chloroform solution into the core. High-performance organic field-effect transistors (OFETs) were fabricated with polyelectrolyte gate dielectric and electrospun P3HT nanofibers by utilizing this approach.69 The operational voltage, field-effect mobility and on/off ratio of the prepared OFET were 2 V, ∼2 cm² V⁻¹ s⁻¹, and 10⁷, respectively.
Beyond core–shell electrospinning, a polymeric blend or composite of conducting polymer has been adopted to produce 1D conducting polymer nanomaterials using electrospinning. Jeong and co-workers have reported on the fabrication of nanofibers with poly(e-caprolactone) (PCL)/P3HT blend. After selective dissolution of the PCL contents on blended nanofibers, an aligned surface morphology of P3HT could be obtained, because of the shear stress of P3HT during electrospinning. They suggested that fine nanofibers composed of P3HT can be prepared by adequate control of the shear stress. Srivastava et al. has reported on the preparation of nanofibers of PVP and their composites with PPy. They employed a multi-spinnerette electrospinning device using microchannels cast in polydimethylsiloxane (PDMS) for scale-up. The pyrrole monomer and initiator for polymerization were introduced onto PVP solution. The insoluble PPy compartments were well-dispersed into a PVP solution, resulting in PPy/PVP composite nanofibers.

Yoon et al. has reported on the electrospinning of nanofibers with polymer blend containing diacetylene molecules. The diacetylene incorporated into polymer nanofibers could be polymerized by UV irradiation in the matrix. By utilizing the end functional groups of diacetylene, diverse types of poly(diacetylene) electrospun nanofibers could be prepared. Because these poly(diacetylene) showed different PL properties under certain environments, the nanofiber-web prepared could be used as colorimetric sensors for detecting harmful organic solvent vapors such as chloroform and toluene.

Aside from polymer materials, small molecules such as rubrene can also be used as the raw materials of electrospun fibers. Dhakal et al. fabricated organic rubrene nanofibers via electrospinning. The rubrene was mixed with a minimal amount of PEO to obtain the optimum viscosity required for electrospinning. The electrospun rubrene nanofibers showed uniform features with a diameter of around 600 nm as shown in Figures 10(a) and (b). After a 10-second collection, large-scale rubrene nanofibers were obtained and their confocal Raman spectra revealed that nanofibers have both amorphous and the crystal phase of the rubrene molecule within the electrospun nanofibers (Figs. 10(c) and (d)).

Besides the above-mentioned methods, nanolithography and soft lithography are also powerful techniques for the preparation of tailored nanostructures. In addition, the single particle nanofabrication technique (SPNT) method suggests a great platform to fabricate NW and nanostructures. Fullerene NWs and $p-n$ junction NWs based on polyfluorene (PFO)/PC$_{61}$BM heterojunction prepared by the SPNT method are illustrated in Figure 11.
3. OPTOELECTRONIC APPLICATIONS OF ORGANIC NANOMATERIALS

3.1. Optoelectronic Devices Based on Single-Component Organic Nanomaterials

Highly crystalline or single-crystalline organic semiconductor NWs typically show higher charge carrier mobility than that of thin film counterparts because of their high structural perfection. Their grain-boundary-free features are beneficial to efficient charge transport, leading to high-performance electronic devices.\(^{12}\)

BPE-PTCDI MWs prepared by non-solvent nucleation method have shown high charge carrier mobility that reached up to 1.4 cm\(^2\) V\(^{-1}\) s\(^{-1}\) with good air-stability.\(^{11}\) In order to realize NW/MW array with controllable alignment and density, the filtration-and-transfer (FAT) alignment method has been developed. The organic NWs or MWs can be aligned by fluid flow through a PDMS mask mold in a simple vacuum filtration setup. The filtered, aligned NW/MW patterns could be transferred onto the desired device substrate via hydrophobic interactions. OFETs based on BPE-PTCDI NW/MW arrays exhibited superior performance compared to thin film OFETs.

Organic phototransistors (OPTs) based on BPE-PTCDI NWs were prepared by Oh and co-workers\(^{12}\) to analyze their optoelectronic properties (Figs. 12(a)–(c)). OPTs based on a single BPE-PTCDI NW exhibited high photoresponsivity, on/off switching ratio, and substantially high external quantum efficiency (EQE) compared to thin-film-based BPE-PTCDI phototransistors. A significant mobility enhancement was observed when the incident optical power density increased and the wavelength of the light source matched the light absorption range of the photoactive material. The photoswitching ratio was strongly dependent upon the incident optical power density, whereas the photoresponsivity was more dependent on matching the light-source wavelength with the maximum absorption range of the photoactive material. BPE-PTCDI NW-OPTs exhibited much higher external quantum efficiency (EQE) values (\(\approx 7900\) times larger) than thin-film OPTs. This is attributed to the intrinsically defect-free single-crystalline nature of the BPE-PTCDI NWs. In addition, compared to the thin-film OPTs, the single-crystalline BPE-PTCDI NW-OPTs exhibited two orders of magnitude higher charge accumulation/release rates from deep traps under on/off switching of external light sources.

Gemayel et al. prepared OPTs based on single or multi perylenebis(dicarboximide) (PDIs) NWs.\(^{31}\) They also observed enhanced phototransistor performance compared to thin-film OPTs (Figs. 12(d) and (e)). The photoresponse in OPTs could be tuned by controlling the device geometry and the morphology of the active semiconducting layer from thin films to NW structures.

Chou et al. utilized self-assembled BPE-PTCDI NWs for nonvolatile transistor memory devices (Figs. 12(f) and (g)).\(^{20}\) The NWs with small diameters resulted in a large memory window. Their stable current response to the write-read-erase-read (WRER) cycles revealed high possibility of organic n-type semiconducting NWs for applications in nonvolatile OFET memory devices. In addition, self-assembled NWs based on alkyl-chain-substituted PTCDI derivatives were used as n-channel semiconductors in OFETs with an electron mobility of 0.01 cm\(^2\) V\(^{-1}\) s\(^{-1}\).
and utilized as high-performance complementary inverters combined with p-channel HTP NWs.

The charge transport characteristics of polymer NWs have been intensively investigated. Self-assembled polymer NWs based on P3ATs have attracted great interest due to their high charge carrier mobilities and self-organizing property. Single component system or blended system of semiconducting/insulating polymers has been actively studied. Lu et al. fabricated poly(3-butylthiophene) (P3BT)/polystyrene (PS) composite films and found that the increased mobility of free charge carriers, in particular hole mobility, contributes to the enhanced electrical conductivity of this semiconductor/insulator composite. Qiu et al. also reported polymer NW/insulator composite OFETs based on P3HT and PS. By using marginal solvent such as CH$_2$Cl$_2$, a network of highly crystalline P3HT nanofibers embedded in an insulating PS matrix was prepared. The P3HT/PS NW OFETs showing reasonably high charge carrier mobilities are promising upon considering the low-cost, environmentally stable, and flexible device characteristics (Figs. 13(a)–(e)). By optimizing the composite fabrication conditions, the devices based on P3HT/PS blend films achieved mobility up to 0.01 cm$^2$ V$^{-1}$ s$^{-1}$, which is comparable to the pristine P3HT film.

Similarly, Kim et al. prepared self-assembled crystalline P3BT NWs and their nanocomposites with insulating PS, and utilized them for high-performance OFETs. The intra-NW carrier mobility reached up to 0.2 cm$^2$ V$^{-1}$ s$^{-1}$ (Figs. 13(d) and (e)). The self-organized P3BT NWs provided efficient charge transport pathways with enhanced performance compared to the pure P3BT NW films.
In addition, Mitsui et al. prepared solution-processed single-crystal FETs using novel p-type semiconductors, naphtho[2,1-b:6,5-b']difuran derivatives (C₈-DPNDF),⁹¹ which exhibited a hole mobility of up to 3.6 cm² V⁻¹ s⁻¹ when using a solution sustaining piece to control the crystal growth (Fig. 14). They found that these features originate from the dense crystal packing and the resulting large intermolecular π-orbital overlap as well as from the small reorganization energy, all of which originate from the small radius of an oxygen atom.

For the large scale and facile synthesis of highly crystalline small molecule nanostructures, dip-coating with a proper controlling of solvent drying speed can be adopted.⁹²-⁹⁴ Jang et al. reported a facile one-step growth of self-aligned acene crystal arrays.⁹⁵ In the dip-coating process, solvents with a low boiling point, such as dichloromethane or chloroform, can be used to produce continuous and uniform crystals over substrates. Figure 15(a) shows a schematic illustration for crystal growth of TIPS-PEN or fluorinated 5,11-bis(triethylsilyl)ethynyl anthradithiophene (FTES-ADT) by dipping with a fast evaporation process. The uniformly grown TIPS-PEN crystals were utilized to fabricate FETs, achieving hole mobility up to 1.5 cm² V⁻¹ s⁻¹ (Figs. 15(b)–(f)).

In order to fabricate dense arrays of single crystals over large-area substrates with controlled crystal alignment, Bao and co-workers developed a droplet-pinned crystallization (DPC) method.¹⁰ The schematic illustration for growth process of C₆₀ crystals is depicted in Figure 16(a). The DPC method induced the fabrication of aligned C₆₀ single crystal arrays along an additional piece.
of silicon wafer and is scalable to a 100-mm wafer substrate, with around 50% of surface coverage with aligned crystals. The needle-like C_{60} single crystal FETs revealed the maximum electron mobility of up to 11 cm² V⁻¹ s⁻¹, while the ribbon-like C_{60} single crystal FETs showed an electron mobility of \( \sim 3 \) cm² V⁻¹ s⁻¹ (Figs. 16(b)–(e)).

3.2. Energy Harvesting Devices Based on Single-Component Organic Nanomaterials

Organic semiconductor nanomaterials have attracted great interest for their applications to organic photovoltaic cells. The active layer in organic solar cells is typically based on bulk heterojunction (BHJ) system in which the photoactive layer consists of a bicontinuous blend of an electron donor (p-type) and an electron acceptor (n-type). Since the exciton diffusion length is approximately 5–10 nm in most organic semiconductors, the nanoscale control of the photoactive layer is critical for achieving high power conversion efficiency (PCE). In order to tune the surface morphology in nanoscale, thermal annealing,96–98 and additive processing99–103 have been extensively employed. As another promising strategy, implementation of NWs in the active layer as either the donor or acceptor component has been explored.14, 16, 17, 104–106 Since diameters of NWs and dimensions can be well matched to the exciton diffusion length, charge separation can be ensured before recombination occurs in NW solar cells. Furthermore, their high carrier mobilities, high absorption coefficients,107 and longer exciton diffusion lengths make them a competitive alternative to the BHJ layer.8

In the case of the NW solar cells based on small-molecule organic semiconductors, C_{60} NWs,79 and N,N′-dioctyl-3,4,9,10-perylenedicarboximide (PTCDI-C_{8}) NWs108,109 have been utilized as nanostructured electron donor (n-type) materials. On the other hand, research on polymer NW solar cells have mostly focused on the p-type NWs, such as P3ATs and their block copolymers, while a small number of n-type polymer NW solar cells were reported; for instance, n-type poly (benzobisimidazobenzophenanthroline) (BBL) nanoribbons were reported by Briseno et al.82

While most of the NW solar cells have been fabricated with either donor or acceptor NWs, BHJ solar cells incorporating both donor and acceptor NWs have also been demonstrated recently. Jenekhe and co-workers110 fabricated a series of oligothiophene-functionalized naphthalene diimides (NDI-nTH and NDI-nT) NWs and utilized as electron acceptor layers in all NW BHJ organic solar cells, NDI-3TH NWs/poly(3-hexylthiophene) NWs. The NW-based solar cells exhibited PCE of 1.15%. The device structure was composed of ITO/PEDOT:PSS (40 nm)/P3HT (15 nm)/active layer (\( \sim 80 \) nm)/1,3,5-Tris(1-phenyl)-1H-benzimidazol-2-yl]benzene (TPBI) (3 nm)/Al (80 nm), where the P3HT and TPBI thin films acted as the
electron-blocking and hole-blocking layers, respectively, and the active layer consisted of P3HT NWs:NDI NWs (1:1 wt/wt) blend.

In particular, periodically and vertically aligned BHJ solar cells are considered to be ideal the architecture for achieving high efficiency. In this device architecture, photogenerated excitons can be easily separated, and the charge carriers have their efficient pathways to each corresponding electrode. The efficient charge transport via vertically aligned NW system has been demonstrated. Figure 17(a) shows organic solar cells based on vertically aligned [6,6]-phenyl-C_{61}-butyric styryl dendronester (PCBSD) nanorods. Fabrication of nanorods was realized via the AAO templating method in combination with the cross-linkable property of PCBSD materials. Li and co-workers \textsuperscript{15} prepared organic/organic ordered nanostructured heterojunction solar cells with a P3HT:Indene-C_{60} bisadduct (ICBA) BHJ upper layer that showed enhanced photocurrent with high PCE of 7.3%.

He et al. fabricated interpenetrating nanostructured polymeric heterojunctions by double nanoimprinting method, and utilized them in the photoactive layer for organic solar cells (Fig. 17(b)). \textsuperscript{111} Poly[(9,9-dioctylfluorene)-2,7-diyl-alt-[4,7-bis(3-hexylthien-5-yl)-2,1,3-benzothiadiazole]-2',2''-diyl] (F8TBT)/P3HT films were nanoimprinted, resulting in ordered nanostructured array with about pitch size of 20 to 50 nm. The PCE of nanostructured solar cells reached 1.9%, which is 50% higher than that of the blend devices.

3.3. Optoelectronics Based on Multi-Component Nanomaterials

Aside from the application of single-component organic nanomaterials, development of multi-component nanomaterials and their application in optoelectronics have been extensively studied. Multi-component organic nanomaterials are very promising candidates as building blocks for high-performance optoelectronic components on a scale ranging from micro- to nano-scale.\textsuperscript{112-115} Although several inorganic NW heterojunctions have been successfully obtained by crystal growth, multi-component organic nanomaterials such as p-n junctions, core–shell NWs, and organic–inorganic hybrid nanostructures have been far
less developed and incorporated into nanoscale photonic devices. Assembling organic NWs into complex heterostructures is critical for realizing multi-functional devices and understanding charge transport and energy transfer phenomena.

Briseno and co-workers fabricated organic single-crystalline $F_{16}$CuPc-CuPc $p-n$ junction nanoribbons by PVD method (Fig. 18). The selective crystallization of $n$-type $F_{16}$CuPc on $p$-type CuPc single-crystalline nanoribbons realized ambipolar charge transporting properties in OFETs with balanced mobilities of 0.05 and 0.07 cm$^2$ V$^{-1}$ s$^{-1}$ for $F_{16}$CuPc and CuPc, respectively. The single $F_{16}$CuPc-CuPc nanoribbon solar cells exhibited PCE of 0.007%.

Multi-component NW building blocks can be prepared from the controlled growth of two different materials. Figure 19 shows Alq$_3$-DAAQ wire-on-wire nanostructures and their optical waveguiding properties. The preformed Alq$_3$ MWs, made by self-assembly in the liquid phase, were used as nucleation sites for the site-specific vapor growth of DAAQ branches to give dendritic heterostructures. When the trunk was excited with a focused laser beam, emitted light of various colors was simultaneously channeled from the branched nanowires via both waveguiding and energy transfer. The intensity of the out-coupled emissions was modulated effectively by changing the polarization of the incident light.
Lee and co-workers recently reported on the high-speed, large-area printing of highly aligned individual NWs. High-speed electrohydrodynamic printing of organic NW was utilized to fabricate organic semiconducting NW arrays on specific substrates. The prepared P3HT:PEO blend NW FETs exhibited field-effect mobility of up to 9.7 cm\(^2\) V\(^{-1}\) s\(^{-1}\). Furthermore, complementary inverter circuit arrays based on the aligned NWs were also fabricated (Fig. 20).

### 4. CONCLUDING REMARKS

Organic nanomaterials are promising building blocks for soft and flexible electronics. 1D nanomaterials and 1D multi-component nanostructures based on small molecules and polymers can be widely employed for use in optoelectronic devices such as OFETs, OPTs, organic memories, organic solar cells, and optical waveguides. The structural perfection of single-crystalline organic 1D NWs that are self-assembled along the \(\pi-\pi\) stacking direction enables efficient charge transport; therefore, one can expect enhanced optoelectronic properties compared to thin film counterparts. Furthermore, the in-depth study on the fundamental charge transport mechanism of organic electronic materials can be effectively performed using 1D tailored organic nanomaterials. Moreover, a multi-component system composed of more than two different materials provides unique and multi-functional properties emerged from the combination of each compound. Inter-molecular interactions enable efficient energy transfer or tunable optoelectronic behaviors. The synthetic approaches of organic nanomaterials and the fabrication techniques of nanoscale optoelectronics need to be further advanced to realize fully integrated soft, stretchable, cost-effective, and energy-efficient nanoelectronics.
ABBREVIATIONS

1-D One-dimensional
2-D Two-dimensional
AAO Anodic aluminum oxide
AFM Atomic force microscopy
Alq3 Aluminum tris(8-hydroxyquinoline)
BBL Poly(7-benzimidazobenzophenanthroline)
BJH Bulk heterojunction
BPE-PTCDI N,N′-bis(2-phenethyl)-perylene-3,4,9,10-tetracarboxylic diimide
BPEA 4,4′-bis(phenylethynyl)anthracene
C12-4ClDiPBI Tetrachlorinated diperylene bisimide
C8-DPNDF Naphtho[2,1-b:6,5-b]xanthenoxanthene
CN-TFMBE 1-cyano-trans-1,2-bis-(3,4:9,10-diimide)-2-fluorinated 5,11-bis(triethylsilylethynyl)anthracene
C8-DPB Fluorinated 5,11-bis(triethylsilylethynyl)anthracene
CuPc Copper phthalocyanine
DAAQ 1,5-diaminoanthraquinone
dBCP Benzophenone 2-carboxyanhydride
DPC Droplet-pinned crystallization
EQE External quantum efficiency
FTES-ADT Fluorinated 5,11-bis(triethylsilylethynyl)anthradithiophene
H,TPyP 5,10,15,20-tetra(4-pyridyl)porphyrin
ICBA Indene-C60 bisadduct
LMW Microwire
MW Nanowire
OFET Organic field-effect transistor
OM Optical microscope
OPT Organic phototransistor
P3AT Poly-(3-alkylthiophene)
P3BT Poly(3-butyliithiophene)
P3HT Poly(3-hexylthiophene)
PCBM [6,6]-phenyl C60 butyric acid methyl ester
PCBSD [6,6]-phenyl-C60-butyric styril dendron-ester
PCE Power conversion efficiency
PCL Poly-(ε-caprolactone)
PDH Poly(diethylaminothiophene)
PDI Perylenebis(dicarboximide)
PDMS Poly(dimethylsiloxane)
PDT Poly(ethylene oxide)
PEO Poly(dodecylthiophene)
PFO Polyfluorene
PL Polarized light microscopy
PLLA Poly-L-lactide
POM Polarized optical microscopy
Ppy Poly(pyrrole)
PS Polystyrene
PTCDI Perylene-3,4,9,10-tetracarboxylic-3,4,9,10-diimide
PVD Physical vapor deposition
PVP Poly(vinyl pyrrolidone)
PVT Physical vapor transport
Rubrene 5,6,11,12-tetraphenylnapthacene
SAED Selected area electron diffraction
SEM Scanning electron microscopy
SPNT Single particle nanofabrication technique
TA-PPE Thiouacetate poly(para-phenylene ethynylene)
TEA Transmission electron microscopy
THF Tetrahydrofuran
TIPS-PEN Triisopropylsilylethynyl pentacene
TPBI 1,3,5-Tris(1-phenyl-1H-benzimidazol-2-yl)benzene
UV Ultraviolet
VCR Vaporization-condensation-recrystallization
VDP Vapor deposition polymerization
WRER Write-read-erase-read.

References and Notes

Received: 10 August 2013. Accepted: 26 August 2013.