Study on Dimensional Control of Poly(3-hexylthiophene) and Their Corresponding Applications

胡, 建臣

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氏名・(本籍・国籍)	フ　　ジェンチェン 胡　　建　臣（中　国）
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論文調査委員	（主　査）准教授　若　山　　裕 （副　査）教　授　安　達　千波矢　　教　授　石　原　達　己

論　文　内　容　の　要　旨

During the past decades, organic semiconductors were scrutinized to identify, understand and apply the useful optoelectronic properties inherent in the electron-rich π-systems. Polythiophene (PT), a conjugated polymers, proved sustained and robust subjects of study, because in PT molecules electrophilic reactions favor sites α to the sulfur atom, enchaining dramaticlly 2,5-couplings to form an extended π-system with quasi one-dimensional delocalization. Besides, the architecture of PTs is to the benefit of three-dimensional self-assembly, generating mesoscopic order and intrinsically large magnitude electronic properties, including efficient two-dimensional charge transport with high mobility, current density and ambipolar transport. Poly(3-hexylthiophene) (P3HT), as a typical representative of PTs, have been studied for fabricating transistors, solar cells and polymer Light Emitting Diodes (LEDs), etc.. Addtional ring substituents make its solubility improved, causing it's possible to carry out the preparation in a simple solution process. To develop the P3HT based organic devices, the carrier process of P3HT should be pay more attention to.

In this thesis, we focus on carrier process of P3HT and the attempt on fabrication of devices with different nanostructures which meet the demands of improvement of the carrier process.

In Chapter 1, a development of PTs and actuality of P3HT applications were described and some current issues of P3HT applications were figured out. Based on the listed current issues of P3HT applications, we proposed the corresponding programs to handle them.

In Chapter 2, we designed and fabricated sub one-dimensional (sub-1D) P3HT nanorod arrays (p-type) by using anodized aluminum oxide (AAO) template and tried to make the nanorods n-type fullerene(C_{60}) was deposited to fill in the interspace to form interdigitate junction (comb-shaped interface) which has a great advantage to enable effective charge generation, dissociation and transportation.

In Chapter 3, we reported on fabrication process of 2,3,5,6-tetrafluoro-7,7,8,8-tetracyanoquinodimethane (F4-TCNQ) highly doped P3HT one-dimensional (1D) nanowires by using AAO templates. We found that the conductivity of P3HT/F4-TCNQ nanowires prepared in porous alumina template was improved because of F4-TCNQ concentration increased by capillary force in nano-pores. F4-TCNQ was doped in different ratios, 0.1 wt%, 1 wt% and 10 wt%. Four-probe scanning tunneling microscope (STM), integrated with a scanning electron microscope (SEM), was employed to measure the resistivity in individual nanowires precisely. The four-probe STM technique has significant benefits in providing stable contacts, suppressing the contact resistance effect, accessing individual measurement nano-objects, and especially in allowing multiple measurements on the same nanowires. The resistivities of P3HT/F4-TCNQ nanowires were tuned in the range of 0.1-10 Ωcm by changing the F4-TCNQ concentration from 10 to 0.1 wt., which were 2-4 orders of magnitude smaller than those of the corresponding P3HT/F4-TNCQ thin film composites. In contrast, the resistivities of F4-TCNQ doped P3HT films were around $4\text{-}5 \times 10^3$ Ωcm, almost independent of the F4-TCNQ concentration.

In Chapter 4, developed a new technique to fabricate large scaled polymer solar cells (PSCs) in low cost. For this purpose, an indium tin oxide/ poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate)/poly(3-hexylthiophene-2,5-diyl):[6,6]-phenyl-C61-butyric acid methyl ester (ITO/PEDOT:PSS/P3HT:PCBM) multilayer structure (2D film) was fabricated in one step in solution process by utilizing solution interfacial tensions. Another strong point of this technique is that the film can be formed almost without any waste of materials. Besides, the active layer thickness can be controlled by the volume of P3HT:PCBM solution. Additionally, this technique is easily to be utilized on soft substrate like polyethylene terephthalate (PET).

In Chapter 5, a summary of Chapter 2, 3 and 4 was provided. Besides, perspective of P3HT applications were listed.

論 文 審 査 の 結 果 の 要 旨

本研究は有機半導体であるP3HTを様々な次元性を持ったナノ構造に加工する技術を確立し、飛躍的な導電性の向上や有機太陽電池への応用を実証している。ここでは高分子鎖の配向といった分子レベルでの構造制御から、櫛形ヘテロ界面のナノレベル構造制御、非平衡現象を利用した高密度ドーピング、高効率大面積バルクヘテロ薄膜作製法など基礎物性から応用展開まで系統的に取り組んでいる。これらの研究は基礎・応用両面において有機機能材料化学に寄与するところが大きく、価値ある業績と認める。