

博士論文の内容の要旨
Abstract of Doctoral Dissertation

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論文題目 Dissertation Title	Study on high-performance conductive fibers towards multifunctional e-textile: fabrication and applications (多機能電子テキスタイルを目指した高機能導電繊維の創製に関する研究)

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Fiber is a symbol of human civilization, being ubiquitous yet obscure in society over most of history. In the last two decades, it has enjoyed a Renaissance with the advent of one-dimensional electronic devices made of fibers and/or owning a fiber shape. We would like to call them fiber-based electronic devices that mainly include 1D fiber-shaped and 2D fabric-shaped electronic devices. Thereafter, fiber-based multifunctional electronic devices have mushroomed and consequently expected to be integrated into a smart system like the human body as an ultimate goal, in which functions of energy generation, energy storage, context sensing, making response, communication, and data processing are combined and coordinated. However, until now it is at the very early stage. In the work, a series of high-performance conductive fibers are prepared via wet-spinning and electro-spinning techniques. Furthermore, as-fabricated conductive fibers are used as functional units to assemble or integrate fabric electrodes. To confirm their applicability, finally we demonstrate the multimode and multifunctional applications on e-textiles. Detailed research contents are as follows:

(1) An ion-induced self-assembly is used to approach continuous and large-scale fabrication of poly (3, 4-ethylenedioxythiophene): poly (styrene sulfonate) (PEDOT: PSS) fibers with an array microstructure, mimicking the spider fluff. The formation of copper complex with fluff-like shape occurs spontaneously on the surface of PEDOT fibers without any additional post-treatment or demanding condition, which is difficult to achieve by other methods. And these biomimetic PEDOT: PSS-Cu²⁺ fibers possess exceptionally large contact area as well as good sensitivity, which endowing it with ultralow detection limit and fast response time. Benefiting from enhanced performance, we envision that braided fabric decorated with such biomimetic fibers demonstrates potential applications for wearable electronic textiles and human-machine interface.

(2) Sodium alginate (SA) as a modifier was added to the wet-spinning solution to prepare PEDOT: PSS/SA composite fibers with high processability applicable to various wearable fields. Interestingly, continuous core-shell PEDOT: PSS/SA composite fibers were fabricated via a single-nozzle technique with regular needles instead of coaxial needles. Apparent shell wrapped on the surface of the fibers with core-shell

structure was proved to be SA gel. Additionally, the mechanical properties of composite fibers were opposite in dry/wet state. The former was strong-yet-brittle, the latter was weak-yet-ductile (typical properties of SA hydrogel). The presence of SA shell improved the stability and durability of electroconductive composite fibers. Combined with several simple proof-of-concept experiments, PEDOT: PSS/SA composite fibers are demonstrated to be a promising candidate for wearable sensors.

(3) Core-shell hydrogel fiber with adjustable three-dimensional (3D) helical structure was fabricated, in which $\text{Ti}_3\text{C}_2\text{T}_x$ (MXene)/PEDOT were modified onto the surface of helical sodium alginate/polyacrylic acid (SA/PAA) hydrogel fibers *via* the combination of in situ polymerization and electrostatic assembly. The hybrid fiber possess a insensitive conductance (<5% resistance change) under various deformation, including stretching (0-800%), bending (0-180°), pressuring and twisting, attributed to both the double tortuous conductive network and pre-strain release from helical structure. Moreover, conductance-stable helical fibers could be appropriately assembled into coaxial energy fibers and integrated into fabric, both acting as strain-insensitive energy storage device and self-powered wearable sensor.

(4) A spatial multi-level nanofibrous membrane with grid-like microstructure of uniform distribution was fabricated, in which carboxylated carbon nanotubes (CCNTs)/ PEDOT was modified onto the surface of grid-like polyurethane (PU) nanofiber *via* the combination of metal mesh template, in situ polymerization and ultrasonic treatment. Nanofibrous membrane enables a pressure sensor with high sensitivities (5.13 kPa^{-1}), fast response/recovery time (80 ms and 120 ms) and ultralow detection limit of 1 Pa. In addition, as a scalable and integrable platform, we also demonstrate its multifunctional applications for electro-thermal conversion and energy harvesting.

(5) Couple techniques of template-based electrospinning and coating were used to prepare a novel pressure sensing platform that features the $\text{Ag}/\text{Ti}_3\text{C}_2\text{T}_x$ (MXene)-modified grid-like polyurethane (PU) nanofibers sandwiched between the encapsulation layer and interdigital electrodes. Resulting nanofibrous sensor possesses high pressure sensitivity (15.3 kPa^{-1}), low limit (1 Pa), broad detection range (0-40 kPa) and excellent stability over 1000 loading/unloading index cycles. Meanwhile, owing to the ultrathin, ultralight, ultraflexible peculiarities, it can robustly conform to non-irregular surfaces and maintain healthy interfacing. In several conceptual displays, we demonstrate it outstanding response properties of human motion in a full range. Assembled tactile array can be further used to detect the pressure stimulation of objects in spatial area by force mapping.

In summary, this work develops a series of 1D/2D functional fiber-based components, respectively, as well as shows how these components can be integrated or assembled into wearable e-textiles and further demonstrate multifunctional applications in sensing, energy harvesting and storage. We believe this advanced toolbox can provide some useful references for the development of e-textiles.