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Electrospinning based all-nano composite materials: Recent achievements and perspectives

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ABSTRACT

Nanoscale composite materials have received more and more attention in recent years. All-nano composite material produced by electrospinning is a rising field, which is in the basic research and primary development stage. Due to the multifunctional properties of these composite materials, they are expected to be applied in many fields. This review focuses on recent progress in reported electrospinning based all-nano composite materials for various applications, such as membrane filtration, adsorption of harmful substances, photo/chemical/electro catalysis, biological medicine, electronic nanodevices, energy storage and conversion, and electromagnetic interference shielding and stealth materials. Finally, we propose our personal insights into future research opportunities and challenges in the hope of stimulating more researchers to engage in this novel field of electrospinning based all-nano composite materials for innovative applications.

1. Brief introduction

In 2000, for the first time, Nobel Prize winner Mr. MacDiarmid brought electrospinning technology to China. Together with Prof. Wanjin Zhang, they proposed the concept of functional organic nanoscale material [1]. Soon afterwards, the new concept of all-nano composite material was put forward by Prof. Ce Wang, in which organic/organic, inorganic/organic, organic/inorganic components were combined diversely via the electrospinning technology [2–7]. Through electrospinning, we could obtain one-dimensional materials with different microstructures, such as nanowires [8], nanotubes [9], nanobelts [10], nanofibers [11], etc. The one-dimensional all-nano composite materials could be assembled into two-dimensional all-nano composite materials [12,13], and further into three-dimensional all-nano composite materials [14,15] (Fig. 1). It is well known that one-dimensional all-nano composite fibers via electrospinning could be deposited onto the collector to obtain two-dimensional fibrous membranes or mats directly [6,7]. In addition, three-dimensional all-nano composite material from electrospinning is a novel research hotspot, which have been studied by many teams, such as Ding's group, Jiang's group, Fong's group, etc. The three-dimensional all-nano composite materials are usually prepared based on freeze-drying a dispersion of short cross-linked or thermally induced self-agglomerated one-dimensional all-nano composite fibers [16–19]. The above is the concept introduction for electrospinning based all-nano composite materials. Because of their multi-components and different morphology, all-nano composite materials prepared through electrospinning have been extensively studied, that tries to find out the synergistic effect, interface effect and even abnormal phenomenon among the components or structures.

2. Applications of electrospinning based all-nano composite materials

The electrospinning based all-nano composite materials possess lightweight, super-soft and ultrathin properties; moreover, through the combination and regulation of organic or inorganic nanocomponents, electrospinning based all-nano composite materials could show amazing functionalization, intelligentialize and the features of superfine fibers, which give them many excellent performances and make them suitable in many fields [20–24]. The application fields of electrospinning based all-nano composite materials mainly focus on membrane filtration, adsorption of harmful substances, photo/chemical/electro catalysis, biological medicine, electronic nanodevices, energy storage and conversion, and electromagnetic interference shielding and stealth materials (Fig. 1). The details are presented below.

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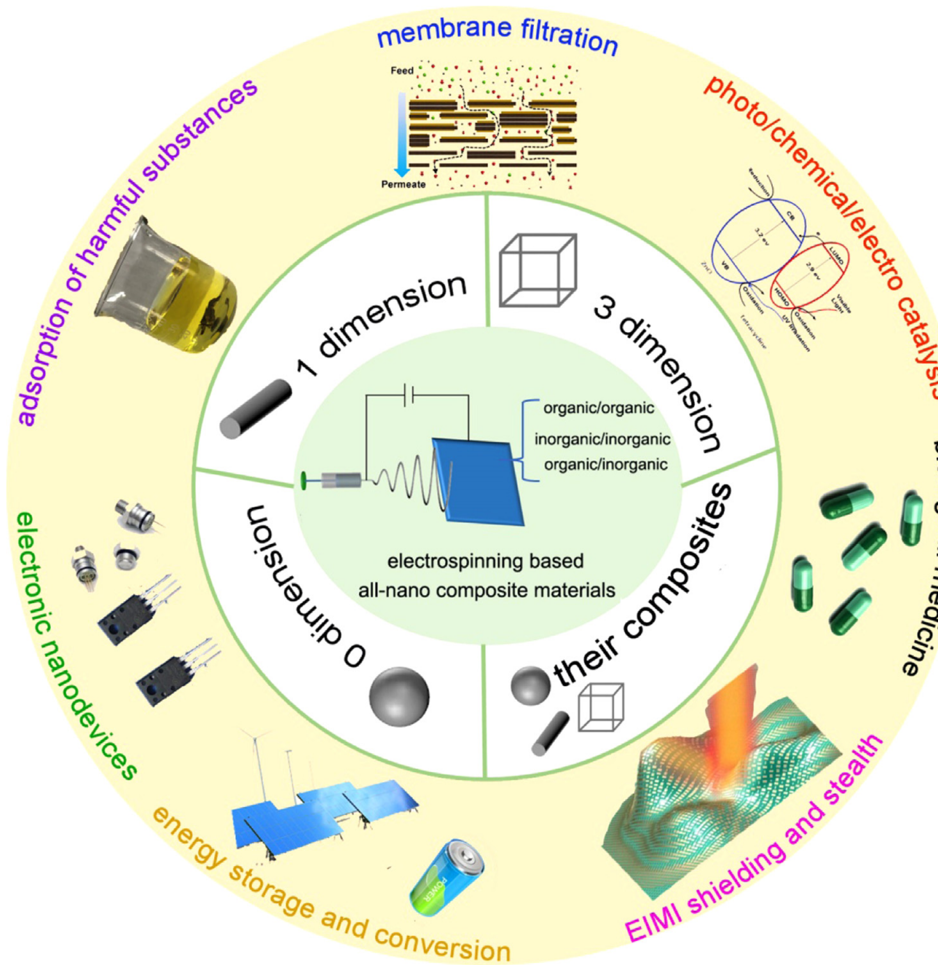

Fig. 1. Schematic illustration of the compositions and applications of electrospinning based all-nano composite materials.

2.1. Membrane filtration

As the fiber preparation technology, electrospinning has one advantage for the fabrication of fibrous membranes. Thus, the good film-forming properties of electrospinning is employed in some applications, such as membrane filtration [25–28]. The flowing mediums are usually water and air. Many strategies are studied to improve the filtration performance of electrospun fiber membranes, including *in-situ* blended electrospinning [29], fiber surface functionalization [30] and membrane surface coating [31].

The water filtration involves in wastewater purification, desalination and oil–water separation. Cai and co-workers prepared polyhydroxybutyrate–calcium alginate/carboxyl multi-walled carbon nanotubes composite nanofibrous filtration membranes (PHB–CaAlg/CMWCNT) via electrospinning technique combined with redissolving the top hydrogel nanofibrous layer to form dense thin film as barrier layer on PHB nanofibrous substrate through suitable water mist wetting process and Ca^{2+} crosslinking (Fig. 2**a**) [32]. The membrane could reject 98.2% Brilliant blue, with a flux of 32.95 $L\,m^{-2}\,h^{-1}$ at 0.1 MPa (Fig. 2**b**). Our group has demonstrated the fabrication of Cu nanocluster immobilized electrospun nanofibrous mats. After a surface modification process by n-hexadecyl mercaptan immersion, the composite showed the super-wetting property with a high WCA of 154.6° and a sliding angle lower than 8.0°, which exhibited high oil-water separation efficiency higher than 99.40% for heavy oil and 98.50% for light oil. Wang et al. used electrospinning technique to fabricate polytetrafuoroethylene nanoparticles embedded polysulfone nanofiber membrane [33]. The nanofiber membrane was tested in desalination by direct contact membrane distillation, showing good distillation performance: a permeate flux greater than 32.7 $kg\,m^{-2}\,h^{-1}$ (80 °C feed temperature, 20 °C permeate temperature, 30 $g\,L^{-1}$ NaCl feed concentration) and a rejection factor higher than or equal to 99.99% with a permeate electrical conductivity lower than 11.9 $\mu S\,cm^{-1}$. In addition to traditional contact membrane distillation, direct solar desalination is an emerging and promising technology. For example, Zhu's group demonstrated a flexible Janus membrane which were decoupled into different layers, with an upper hydrophobic carbon black nanoparticles (CB) coating polymethylmethacrylate fiber layer for light absorption, and a lower hydrophilic polyacrylonitrile fiber layer for pumping water [34]. The Janus membrane showed high efficiency (72%) and stable water output (1.3 $kg\,m^{-2}\,h^{-1}$, over 16 days) under 1-sun, suitable for direct solar desalination. In another study, Jin and co-workers prepared nylon-6/carbon black composite fiber cloth for efficient and durable solar steam evaporation [35]. The fiber cloth could absorb 94% of the solar spectrum and give rise to a state-of-the-art solar energy utilization efficiency of 83% during the pure water evaporation process. It could also be hand-washed more than 100 times without degrading its performance and thus offered a potential mechanism for foulant cleaning during practical solar steam generation and distillation processes.

Air filtration is also an important application for electrospun fiber membranes, due to their interconnected nanoscale pore structures, highly specific surface areas, fine diameters, and porous structure as well as their ability to incorporate active chemistry on a nanoscale surface [36]. Many polymer or mixture have been prepared into fiber membranes to treat growing air pollution problem. Ding' group prepared silicon nitride/polyvinyl butyral nanofibrous membranes

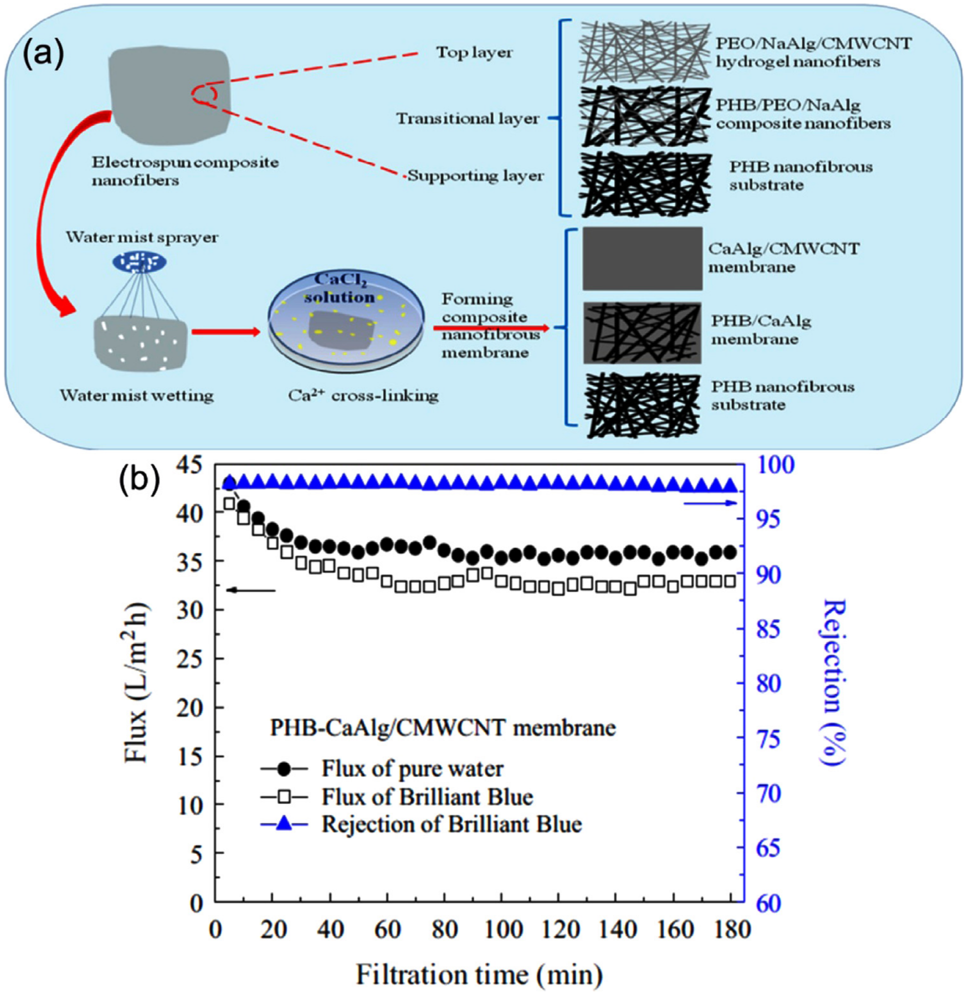

Fig. 2. (a) The schematic diagram for the preparation of PHB–CaAlg/CMWCNT composite nanofibrous filtration membranes. (b) The flux and rejection rate of PHB–CaAlg/CMWCNT composite nanofibrous membrane for Brilliant blue. Reprinted with permission from Ref. [32], Copyright 2016, Elsevier.

followed by introducing hydro-oleophobic fluorinated polyurethane. The resultant membranes achieved a high filtration efficiency of 99.950%, low air resistance of 55.0 Pa, and robust durability, providing a guidance for the design of electret air filtration materials used in haze environment [28]. Wang and co-workers incorporated the nanoparticles of zeolitic imidazolate framework-8 (ZIF-8) into poly (lactic acid) (PLA) to prepare porous electrospun membranes [37]. The PM capture measurement demonstrated that the PLA/ZIF-8 porous membranes had significantly enhanced $PM_{2.5}$ removal efficiency compared to that of the pure PLA membrane. Qian and co-workers used electrospun polyimide nanofibers as building blocks to construct a three-dimensional hierarchically porous architecture via freeze-drying as thermal insulators and particulate air filters [38]. The 3-D structural filter showed ultralow density, high-temperature stability, low thermal conductivity and excellent performance in $PM_{2.5}$ filtration. Remarkably, electrospun fiber based anti-smog masks have been sold by many companies in China, which inspire researchers to realize the industrialization of electrospinning.

2.2. Adsorption of harmful substances

To removal pollutants from wastewater, adsorption is also a preferred method and is employed by many researchers due to its advantages of the easy operation, low cost, and high efficiency [39,40]. As the key in the adsorption method, the performance of adsorbent plays a decisive role. The all-nano composite materials prepared by electrospinning technique show high porosity, easy preparation, easy modification, and recoverability, which makes them as suitable adsorbents in wastewater treatment [41–43]. According to the composition, the electrospinning based adsorbents can be divided into inorganic all-nano adsorbents, organic all-nano adsorbents, inorganic/organic composite all-nano adsorbents. The adsorbed pollutants are also diverse classes, such as heavy metals, organic dyes, antibiotics, endocrine disrupting compounds, pesticides, oil contamination, microorganisms, radioactive metal ions, inorganic phosphorus and so on.

Chen et al. fabricated flexible silica/mesoporous alumina core-shell fibers through a facile and applicable one-step coaxial electrospinning technique [44]. The core-shell fiber adsorbents exhibited good adsorption performance toward Congo red with an adsorption of 115 mg g^{-1} within 48 h. Besides, the form of the silica/mesoporous alumina fibers was kept well throughout the reuse process, and the adsorption capacity decreased only slightly after several cyclic experiments. Recently, our group reported the hierarchical aminated polyacrylonitrile (PAN)/γ-AlOOH electrospun composite nanofibers by a combination of electrospinning process, chemical modification and hydrothermal reaction [45]. The adsorption behaviors toward Pb^{2+}, Cu^{2+} and Cd^{2+} ions in aqueous solution were investigated. The maximum monolayer adsorption capacities for Pb^{2+}, Cu^{2+} and Cd^{2+} were 180.83 mg g^{-1}, 48.68 mg g^{-1} and 114.94 mg g^{-1}, respectively. Adsorption mechanism analysis proved that the amine groups and γ-AlOOH crystals synergistically contributed to the high adsorption activity of the hierarchical aminated PAN/γ-AlOOH composite nanofibers. The obtained results suggested their potential application in heavy metal ions removal from wastewater effluents. Abbasizadeh and co-workers synthesized a novel polyvinyl alcohol (PVA)/titanium oxide (TiO_2) nanofiber adsorbent modified with mercapto groups by electrospinning [46]. The results showed that the adsorption capacity of radioactive U (VI) for the modified PVA/TiO_2 nanofibers was remarkably greater than those of the unmodified nanofibers. The maximum adsorption capacity of U(VI) was estimated to be 196.1 mg g^{-1} at 45 °C with pH of 4.5. Thermodynamic parameters showed that the nature of U(VI) adsorption onto the nanofiber was endothermic and spontaneous, and was favored at higher temperature.

2.3. Photo/chemical/electro catalysis

Electrospinning provides an effective way to fabricate inorganic ceramic nanofibers, which are suitably used as catalysts (photo/chemical/electro) [47]. On the other hand, the electrospun mats from stable polymer or ceramic fibers are also good catalytic supports due to their large surface area and a high porosity [5,48,49]. Generally, electrospinning based all-nano catalysts could be synthesized from blending electrospinning or post-loading functional nanoparticles.

In the report of Gao's group, a novel hierarchical MnOx/TiO_2 composite nanofiber was fabricated by combining the electrospinning technique and hydrothermal growth method [50]. The synthesized nanomaterial, which comprised primary TiO_2 nanofibers and secondary MnOx nanoneedles, presented high catalytic oxidation performance on low-concentration acetone. The synergistic effect of the specific hierarchical nanofibrous morphology, the abundant surface-adsorbed oxygen, the superior redox property, and the sufficient specific surface contributed to the high catalytic activity. Wu and co-workers fabricated MWCNTs/Ag_3PO_4/polyacrylonitrile (PAN) ternary composite fiber membranes (TCFMs) with good photocatalytic performance by electrospinning technique combined with in situ Ag_3PO_4 forming reaction [51]. The as-prepared MWCNTs/Ag_3PO_4/PAN TCFMs showed enhanced photocatalytic activity and stability for degrading rhodamine B (RhB) in batch processing systems, which mainly ascribed to fast electron transfer from Ag_3PO_4 to MWCNTs and the resulting high electron–hole (e^-–h^+) separation efficiency (Fig. 3). Radical trapping experiments revealed that holes (h^+) and superoxide radicals ($\cdot O^{2-}$) played primary roles in RhB degradation. In another study of Chen' work, they search for non-noble metal catalysts with high electrocatalytic activity for the hydrogen evolution reaction (HER) [52]. They reported a novel WO_{3-x} catalyst on carbon nanofiber mats (CFMs) through electrospinning and carbonization process. The as-prepared catalysts exhibited substantially enhanced activity for the HER, demonstrating a small overpotential, a high exchange current density, and a large cathodic current density. The remarkable electrocatalytic performances resulted from the poor crystallinity of WO_{3-x}, the high electrical conductivity of WO_{3-x}, and the use of electrospun CFMs.

2.4. Biological medicine

Electrospinning is a simple, cost-effective, and reproducible process to prepare both biocompatible synthetic and natural polymers fibers high-surface area, micro-porosity, and the ability to load drugs or other biomolecules into the fibers; moreover these electrospun porous nanofibers can mimic the micromorphology of the native extracellular matrix (ECM) and enhance the cell migration and proliferation, the electrospun nanofibers have a wide range of applications in biomedical fields such as tissue engineering scaffolds, wound dressing materials and carriers for drug delivery [53–55].

Due to the preparation feature from polymer solution, electrospun fibrous structures have been recognized as drug-delivery system candidates. So, the most direct drug-release system could be obtained from the blending electrospinning of drug/polymer mixed solution. Indeed, controlling the release of drugs is essential for sustained delivery application. Various possibilities to control the release of hydrophilic or hydrophobic drug from electrospun fibers and different electrospinning methods such as emulsion electrospinning and coaxial electrospinning for drug loading are explored by many researchers. Prabhakaran and co-workers produced either metformin hydrochloride (MH) or metoprolol tartrate (MPT) loaded poly(3-caprolactone) (PCL) or poly(3-

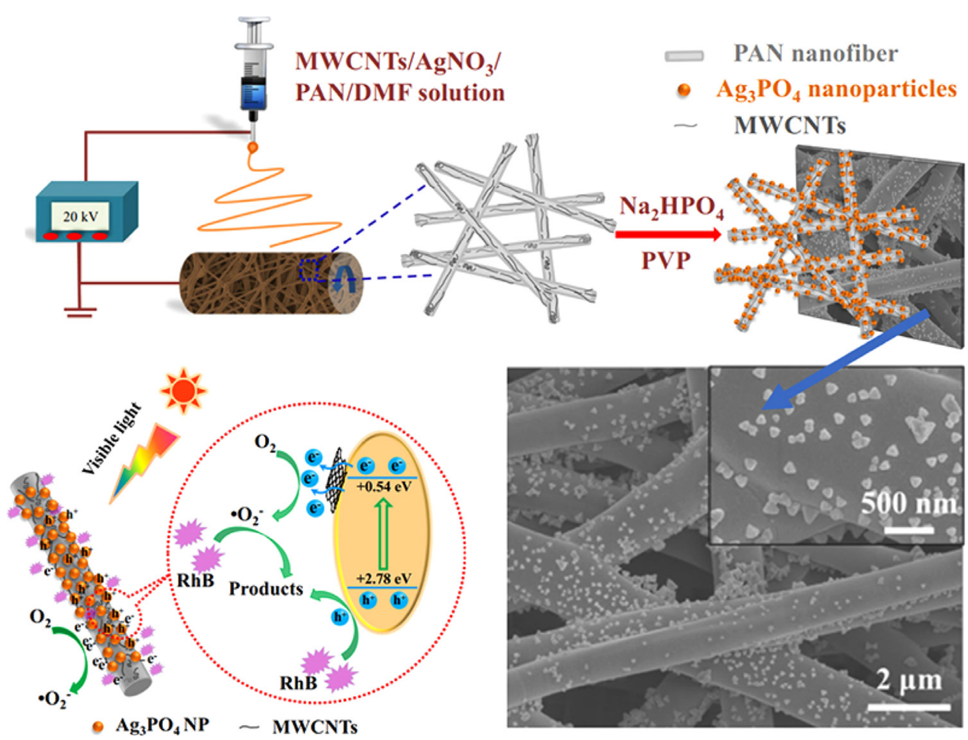

Fig. 3. Schematic diagram of the preparation of MWCNTs/Ag_3PO_4/PAN TCFMs and their proposed reaction mechanism for photocatalytic degradation of RhB. Reprinted with permission from Ref. [51], Copyright 2018, Springer.

hydroxybutyric acid-co-3-hydroxyvaleric acid) (PHBV) nanofibrous scaffolds by emulsion electrospinning [56]. *In vitro* release studies indicated that the emulsion electrospun nanofibers significantly alleviated the burst release and produced a sustained release of drugs compared to the blended electrospun nanofibers. Between the two polymers studied, PCL demonstrated a better drug delivery carrier compared to PHBV, and MPT incorporated nanofibers showed less burst release than the others.

For tissue engineering scaffolds, several tissue substitutes (such as neural tissue, bone tissue, cartilage tissue, vascular tissue, ocular tissue, skeletal muscle tissue, etc.) have been fabricated by electrospinning. The compositions and structures of electrospun fibers are regulated to meet the needs of different tissues [19,57,58]. Ramakrishna and co-workers demonstrated the possibilities of fabricating aligned poly(L-lactic acid) (PLLA) fibrous scaffolds by electrospinning technique [59]. It was obvious that the fiber diameter could easily be tailored by manipulating the processing parameters. The *in-vitro* cell experiment results showed that the direction of neural stem cell elongation and its neurite outgrowth was parallel to the direction of PLLA fibers for aligned scaffolds. No significant changes were observed on the cell orientation with respect to the fiber diameters. the experimental results suggested the aligned nanofibrous PLLA scaffold could be used as a potential cell carrier in neural tissue engineering. Recently, Mo's group reported a novel tri-layer tubular graft consisted of Poly(L-lactide-co-caprolactone)/collagen (PLCL/COL) fibers and Poly(lactide-co-glycolide)/silk fibroin (PLGA/SF) yarns via a three-step electrospinning method [60]. The tri-layer vascular graft consisted of PLCL/COL aligned fibers in inner layer, PLGA/SF yarns in middle layer, and PLCL/COL random fibers in outer layer (Fig. 4**a**). The human umbilical vein endothelial cells (HUVECs) and smooth muscle cells (SMCs) proliferated well on the materials. Fluorescence staining images demonstrated that the axially aligned PLCL/COL fibers prearranged endothelium morphology in lumen and the circumferential oriented PLGA/SF yarns regulated SMCs organization along the single yarns. The outside PLCL/COL random fibers performed as the fixed layer to hold the entire tubular structure. The in vivo results showed that the tri-layer vascular graft supported cell infiltration, scaffold biodegradation and abundant collagen production after subcutaneous implantation for 10 weeks (Figs. 4**b** and 4**c**), revealing the optimal biocompatibility and tissue regenerative capability of the tri-layer graft. Moreover, Fong and co-workers reported the 3D electrospun nanofibrous polycaprolactone (PCL) scaffolds through a novel technique, named thermally induced nanofiber self-agglomeration [58]. After the functionalization with hydroxyapatite and bone morphogenic protein 2, the composite scaffolds showed synergistically enhanced osteogenic differentiation capabilities of C2C12 cells. Importantly, *in vivo* studies indicated that this synergism was able to generate significantly increased new bone in an ectopic mouse model.

Electrospinning based fibers' high specific surface area, porous structure and high porosity can help water and oxygen penetrate, meanwhile provide hemostatic and absorbability properties, making them very promising as wound dressing materials. At the early stage, research focus on the electrospinning of a great many biodegradable and biocompatible synthetic and natural polymers, such as PCL, PVA, PLA, PU, PEO, PVP, polysaccharides, gelatin, and collagen. In the past ten years, to satisfy the demand for novel wound dressings, some functional substances, such as antibiotics, enzyme and growth factors, etc., are added into the electrospun fibers to improve the wound healing performance. Li et al. prepared ciprofloxacin (CIF) loaded poly(N-isopropylacrylamide) (PNIPAAm)/poly(L-lactic acid-co-ε-caprolactone) (PLCL) electrospun fibers [61]. L929 fibroblasts could proliferate on the fibers, indicating their biocompatibility. The CIF-loaded fibers were found to have potent antibacterial activity against *E. coli* and *S. aureus. In vivo* tests on rats indicated that CIF-loaded PNIPAAm/PLCL fibers have enhanced healing performance compared to CIF-loaded PLCL fibers or a commercial gauze. Cui' group reported the basic fibroblast growth factor (bFGF) grafted electrospun poly(lactide-co-glycolide) (PLGA) fibers through polydopamine (PDA) assistance, which maintained original fiber properties and growth factor bioactivity [62]. *In vitro* experiments showed that the bFGF-grafted PLGA electrospun fibrous scaffolds had highly enhanced adhesion, viability, and proliferation of human dermal fibroblasts. *In vivo* results showed that such scaffolds shortened wound healing time, accelerated epithelialization and promoted skin remodeling.

2.5. Electronic nanodevices

In the past few years, much attention has been paid to the electronic properties of metallic and semiconducting nanoscale materials for their potential applications in fabricating nanoscale electronic devices. In Section 2.3, we have demonstrated that electrospinning could provide an effective way to fabricate inorganic nanofibers. Due to their long and continuous morphology which could promote electron transfer and migration, all-nano composite materials synthesized by electrospinning have been studied as a model of electronic devices. Our group reported the fabrication of Au-doped polyacrylonitrile (PAN)–polyaniline (PANi) core–shell nanofibers via electrospinning and subsequent gas-phase polymerization [63]. The as-prepared Au-doped PAN-PANi core–shell nanofibers provided a very high field-effect mobility of up to 11.6 cm^2 V^{-1} s^{-1}, without crystallizing the molecular structures of polymers. This high mobility could be due to the nanofiber structure, which promoted charge transfer and reduced the grain-boundary effect, and the doping of Au nanoparticles, which served as "conducting bridges" between the PANi semiconducting domains. This approach was also suitable for other conducting polymers. Thus, this work may eventually lead to a new and simple method for fabricating high performance polymer field effect transistors.

Gas sensor devices represent an ideal application to take advantage of electrospinning based all-nano composite materials. The constituents for gas sensor devices are semiconducting materials, including SnO_2, ZnO, TiO_2, In_2O_3 and so on. Loading with functional nanoparticles or constructing hierarchical structures are often investigated to improve sensing property. Kim's group reported the nanocrystalline fiber mats of unloaded and Pd-loaded SnO_2 synthesized by electrospinning followed by hot-pressing and calcination at 450 or 600 °C [64]. The crystallization and microstructure evolution were strongly dependent on the amount of Pd loading, as well as on the calcination temperature. The Pd-loaded SnO_2 fiber mats demonstrated significantly enhanced sensitivity to H_2 compared to their unloaded counterparts, while the sensitivity to NO_2 was reversed with the unloaded SnO_2 sensors demonstrating higher sensitivity than their Pd-loaded counterparts. These observations could be attributed to the catalytic effect of the PdO nanocrystallites in promoting the oxidation of H_2 to H_2O and enhancing electron depletion at the surface of the SnO_2 crystallites. Zhang and co-workers fabricated a novel hierarchical heterostructure of α-Fe_2O_3 nanorods/TiO_2 nanofibers with branch-like nanostructures using a simple two-step process called the electrospinning technique and hydrothermal process [65]. A high density of α-Fe_2O_3 nanorods (about 200 nm in diameter) was uniformly deposited on a TiO_2 nanofibers backbone. Different nanostructured α-Fe_2O_3/TiO_2 heterostructures were also investigated by controlling the volume ratio of the reactants. The α-Fe_2O_3/TiO_2 heterostructures with a proper mixture ratio of the reactants sensor exhibited obviously enhanced sensing characteristics, including higher sensing response, lower operating temperature, faster response speed, and better selectivity in comparison with other ones. Moreover, the α-Fe_2O_3/TiO_2 heterostructures sensor also exhibited excellent sensing performances compared with α-Fe_2O_3 nanorods and TiO_2 nanofibers sensors.

2.6. Energy storage and conversion

As the performance of energy devices depends significantly upon

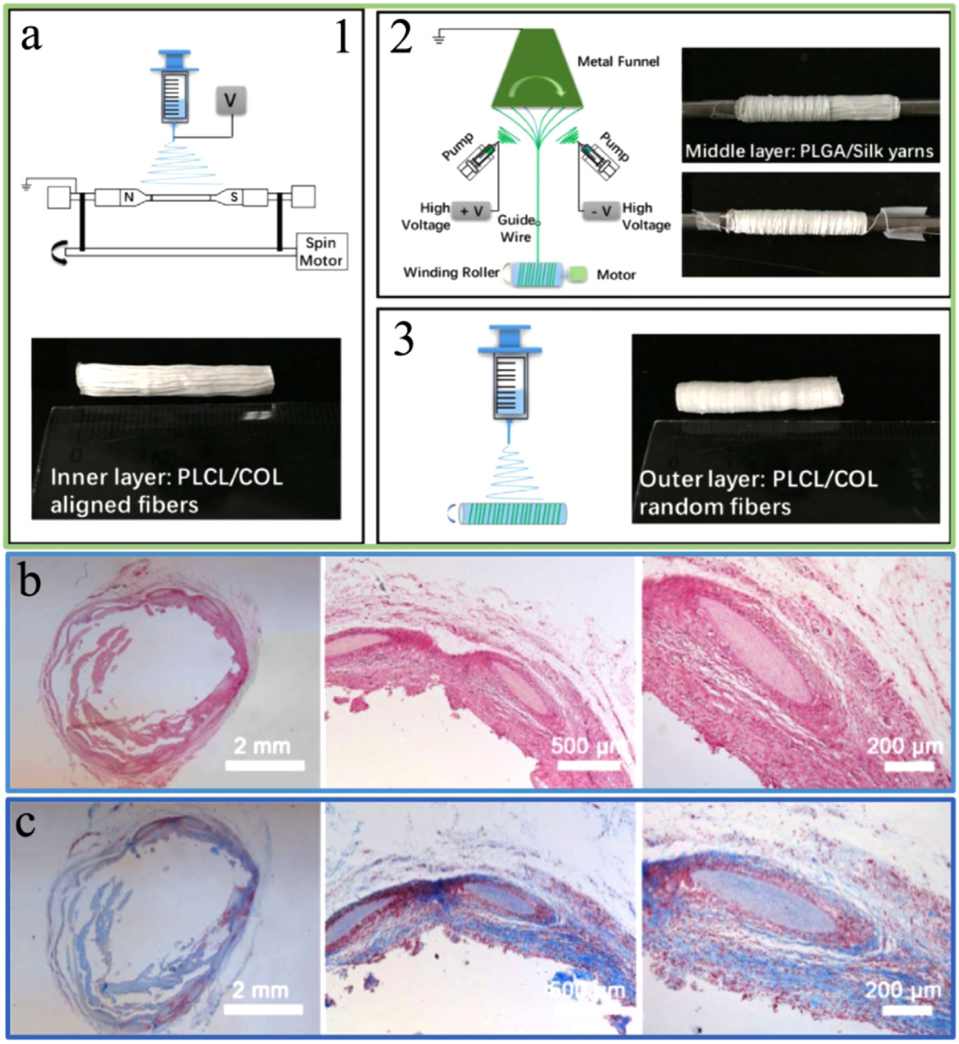

Fig. 4. (a) The schematic diagram showing the fabrication of tri-layer tubular graft. H & E staining images (b) and Masson's trichrome staining images (c) of the transplanted grafts after subcutaneous embedding in mice for 10 weeks. Reprinted with permission from Ref. [60], Copyright 2017, Elsevier.

the functional materials, the dimensionality, directionality and compositional flexibility of electrospinning based all-nano composite materials are increasingly being investigated for the targeted development of electrode and electrolyte materials, where the specific properties associated with nanoscale features such as high surface area and aspect ratios, low density and high pore volume allow performance improvements in energy conversion and storage devices, ranging from dye sensitised solar cells, fuel cells, lithium ion batteries, supercapacitors and piezoelectric nanogenerators.

A typical dye sensitised solar cell (DSSC) is composed of a photoanode, a counter electrode, and an electrolyte containing a redox couple, such as iodide/triiodide (I^-/I_3^-), that is interspersed between the electrodes to establish an electrical connection (Fig. 5**c**). Due to the structure and composition of electrospinning based all-nano composite materials, they are often investigated as counter electrode or photoanode. Qiu and co-workers prepared Co_9S_8 nanoparticle/carbon nanofiber (CNT) composite materials (Figs. 5**a** and 5**b**) via an electrospinning followed by calcination and hydrothermal reaction as an efficient and low-cost Pt-free counter electrode (CE) for DSSCs [66]. The DSSC assembled with the prepared Co_9S_8/CNFs composite as a CE exhibited excellent electrocatalytic activity and a power conversion efficiency (PCE) of 8.37% (Fig. 5**d**), which was comparable to that of the DSSC with conventional thermally deposited Pt as the CE (8.50%). Therefore, the Co_9S_8/CNFs composite can be used as an efficient and low-cost promising alternative CE in DSSCs to replace traditional Pt electrode. For photoanode, electrospun TiO_2 materials with different morphology, such as nanofiber, nanowire, nanorods, microspheres, hollow-nanotube and nanotube have been studied [67].

Fuel cells are energy devices that can convert chemical energy into electrical energy through a redox reaction between the anode and the cathode in the presence of catalysts. Because of their high efficiency without the emission of pollutants, they have received considerable attention as an alternative energy source. The potential of electrospinning based all-nano composite materials for fuel cells are focused on anodes, cathodes, or proton conductive membranes. Alvi et al. prepared an effective and low cost Pd-Ce bimetallic decorated carbon nanofibers by electrospinning as anode for the high-performance methanol fuel cells [68]. Pullithadathil' group reported the preparation of Pt grafted N-doped mesoporous carbon nanofibers (Pt/MPCNFs) as a cathode catalyst using electrospinning for high-performance PEM fuel cells [69]. Performance analysis in a single PEM fuel cell showed twice the power density (428 mW cm^{-2}) with Pt/MPCNFs compared to commercial electrocatalyst membranes due to the effective enhancement in the triple phase boundaries. Due to its good membrane properties, electrospun fiber membranes were also employed as proton conductive membrane in fuel cells. Kawakami's group fabricated the composite membranes composed of phytic acid-doped polybenzimidazole electrospun nanofibers and Nafion matrix [70]. The composite membrane showed higher proton conductivity than the recast-Nafion membrane without nanofibers.

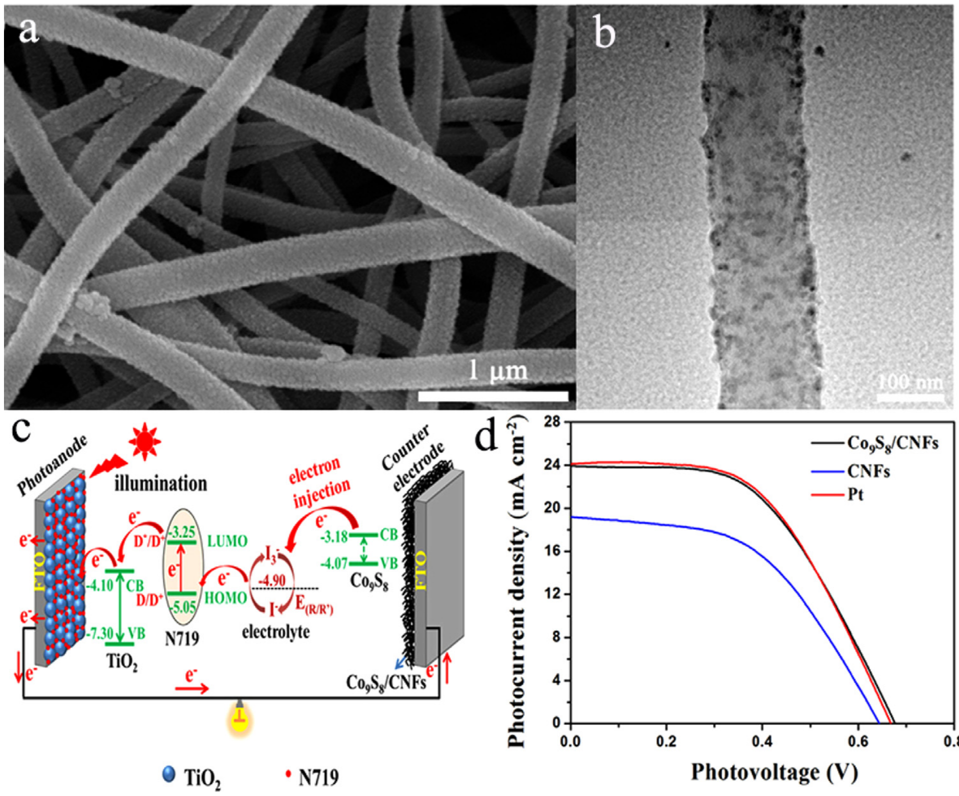

Fig. 5. SEM (a) and TEM (b) images of Co_9S_8/CNFs composite. (c) Schematic diagram of the work principle of DSSC. (d) Photovoltaic performance of DSSCs fabricated using Pt, CNFs and Co_9S_8/CNFs as CEs. Reprinted with permission from Ref. [66], Copyright 2018, Elsevier.

Among various secondary batteries, lithium ion batteries (LIBs) have attractive advantages, such as high energy density, low maintenance, and relatively low self-discharge [71]. A LIB consists of an anode and a cathode separated by an electrolyte containing dissociated lithium salts, which enables the transfer of lithium ions between the two electrodes. Electrospinning based all-nano composite materials are adopted to design component materials (anodes, cathodes, and separators) for LIBs to improve the efficiency and the durability. Mu and co-workers prepared flexible TiO_2/SiO_2/C films as self-supporting electrodes for lithium-ion batteries (LIBs) by using an electrospinning approach [72]. The TiO_2/SiO_2/C film anode exhibited excellent high-rate capability with a capacity of 115.5 mAh g^{-1} at 8 A g^{-1} (9.8 C rate) and good storage performance. It also showed high long-term cycling stability of 700 cycles at 200 mA g^{-1} with a capacity of 380.1 mAh g^{-1} and a high capacity retention of 88.3%. In the study of Duan' group, Nb-Doped $LiNi_{0.4}Co_{0.2}Mn_{0.4}O_2$ (Nb-NCM) nanobelts have been successfully fabricated through a facile electrospinning method followed by the calcination [73]. When used as a cathode for lithium-ion batteries, the as-electrospun Nb-NCM nanobelts exhibited superior electrochemical performances, which could be attributed to their unique features, such as their 1D nanostructure, well-crystallized nature and Nb-doping, which could improve the ionic conductivity of Nb-NCM nanobelts and stabilize the electrode/electrolyte interface. Ding et al. developed electrospun porous polyurethane@graphene oxide (PU@GO) membranes as ideal separator candidates for high-performance LIBs [74]. Electrolyte-uptake capacity and ionic conductivity of the flexible PU@GO separators were superior to those of commercial Celgard separators at 733% and 3.73 mS cm^{-1}, respectively, resulting in better battery performances in terms of impedance, specific discharge capacity and cycle life.

With large surface area, high porosity, low density, good directionality, and tunable composition, electrospinning based all-nano composite materials are regarded as ideal candidates for electrodes in supercapacitors (SCs). Electrospinning based all-nano composite materials including 1D carbons, metal oxides, metal sulfides, metal nitrides, conducting polymers and composite nanomaterials with various types of architectures as electrodes for SCs have been studied. Tian et al. prepared different nanostructures of Ni–Mn oxide (Ni–Mn–O) on flexible electrospun carbon nanofibers (ECNFs), which were further used as binder-free electrodes for supercapacitors [75]. Among three typical nano-structures, Ni–Mn–O nanosheets on ECNFs showed superior electrochemical properties with a good specific capacitance (368.6 F g^{-1}) and excellent rate properties (88.6% retention at 20 times initial current density) in a three-electrode system. Furthermore, an aqueous symmetry cell and a solid-state symmetry supercapacitor are assembled using two pieces of flexible ECNFs@Ni–Mn–O nanosheets. The maximum energy density and power density based on the solid-state supercapacitor were up to 20.4 W h kg^{-1} and 5.2 kW kg^{-1}, respectively. Shao and co-workers fabricated the freestanding electrospun carbon nanofibers/carbon nanotubes/polyaniline (CNFs/CNTs/PANI) ternary composites [76]. As expected, the ternarycomposites as electrodes presented high specific capacitance (i.e., 315 F g^{-1} at 1 A g^{-1}) and dramatic rate capability (i.e., 235 F g^{-1} at 32 A g^{-1}) in three-electrode configuration. Moreover, the as-fabricated flexible solid-state supercapacitor based on the ternary composites also achieved desired electrochemical properties with high capacitance, high-rate capability, high energy/power density (i.e., 5.1 Wh kg^{-1} at 10.1 kW kg^{-1}), and remarkable cycling stability (i.e., 92% capacitance retention after 10,000 cycles at 2 A g^{-1}).

Dielectric materials with high dielectric permittivity, good mechanical properties, and excellent thermal stability are highly desired n the embedded capacitors and energy-storage devices. Electrospinning based all-nano composite materials with ceramic fillers and conductive fillers are good candidates as dielectric materials [77,78]. Hou and co-workers fabricated polyimide/carbon nanotubes (PI/MWCNTs) nanocomposites by electrospinning and hot-pressing [79]. The PI/MWCNTs nanocomposites with 20 vol% concentration of MWCNTs had the highest dielectric permittivity of 217 (at 1 kHz), which was 57.5 times higher than the pure PI. Compared to the maximum energy storage density of pure PI (0.404 J cm^{-3}), the best maximum energy storage density of 1.957 J cm^{-3} was achieved by incorporating 12 vol% of MWCNTs into PI. Furthermore, they prepared polyimide/$BaTiO_3$/MWCNTs three-phase nanocomposites [80]. The investigation shows that the dielectric permittivity of the as-fabricated three-phase nanocomposites with 40 vol% BTPs and 10 vol% MWCNTs loads is as high as1061.98 at 100 Hz with a lower dielectric loss of 0.23, and the corresponding energy storage density is 4.773 J cm^{-3}, 11 times that of the pure PI.

In addition to photoelectric conversion, thermoelectric conversion and piezoelectric conversion attract extensive interest by researchers in recent years. Compared with thermoelectric conversion, piezoelectric conversion properties are more suitable for electrospinning based all-nano composite materials. Most research focus on poly(vinylidene fluoride) (PVDF) based electrospun nanofibers. Yu et al. proposed a novel approach to prepare electrospun polyvinylidene fluoride (PVDF) nanofibers with high piezoelectric performance [81]. PVDF nanofibers were doped with inorganic salts without the use of any postpolarization treatment. The optimized piezovoltage of PVDF nanofibers doped with $FeCl_3 \cdot 6H_2O$ was 700% higher than that of a device with undoped nanofibers. A large area, flexible piezoelectric material that consisted of aligned arrays of electrospun fibers of the polymer poly(vinylidene-fluoride-co-tri-fluoroethylene) were reported by Persano and co-workers (Fig. 6**a**–6**c**) [82]. The resulting material offered exceptional piezoelectric characteristics, to enable ultra-high sensitivity for measuring pressure, even at exceptionally small values (0.1 Pa) (Fig. 6**d**–6**f**). The collective results suggested utility in a variety of sensor- and energy harvesting components, with lightweight construction, attractive mechanical properties and potential for implementation over large areas at low cost, with application opportunities in human motion monitoring and robotics.

2.7. Electromagnetic interference shielding and stealth

In recent years, the electromagnetic waves have become a serious problem because of the proliferation of electronic devices which justifies an active quest for novel and effective electromagnetic interference (EMI)-shielding materials or microwave absorbing materials in a wide variety of applications. The shielding or absorbing performance toward electromagnetic waves mainly depends on electrical and magnetic properties of the materials. Due to their low density and tunable composition, electrospinning based all-nano composite materials are explored for preparing electromagnetic interference (EMI)-shielding and microwave absorbing materials. Li et al. fabricated the self-healing superhydrophobic polyvinylidene fluoride/Fe_3O_4@polypyrrole (F-PVDF/Fe_3O_4@PPy_x) fibers with core–sheath structure by electrospinning of a PVDF/Fe_3O_4 mixture and in situ chemical oxidative polymerization of pyrrole, followed by chemical vapor deposition with fluoroalkyl silane [83]. The F-PVDF/Fe_3O_4@$PPy_{0.075}$ fiber film produced a superhydrophobic surface with self-healing behavior. Moreover, the maximum reflection loss (R_L) of the F-PVDF/Fe_3O_4@$PPy_{0.075}$

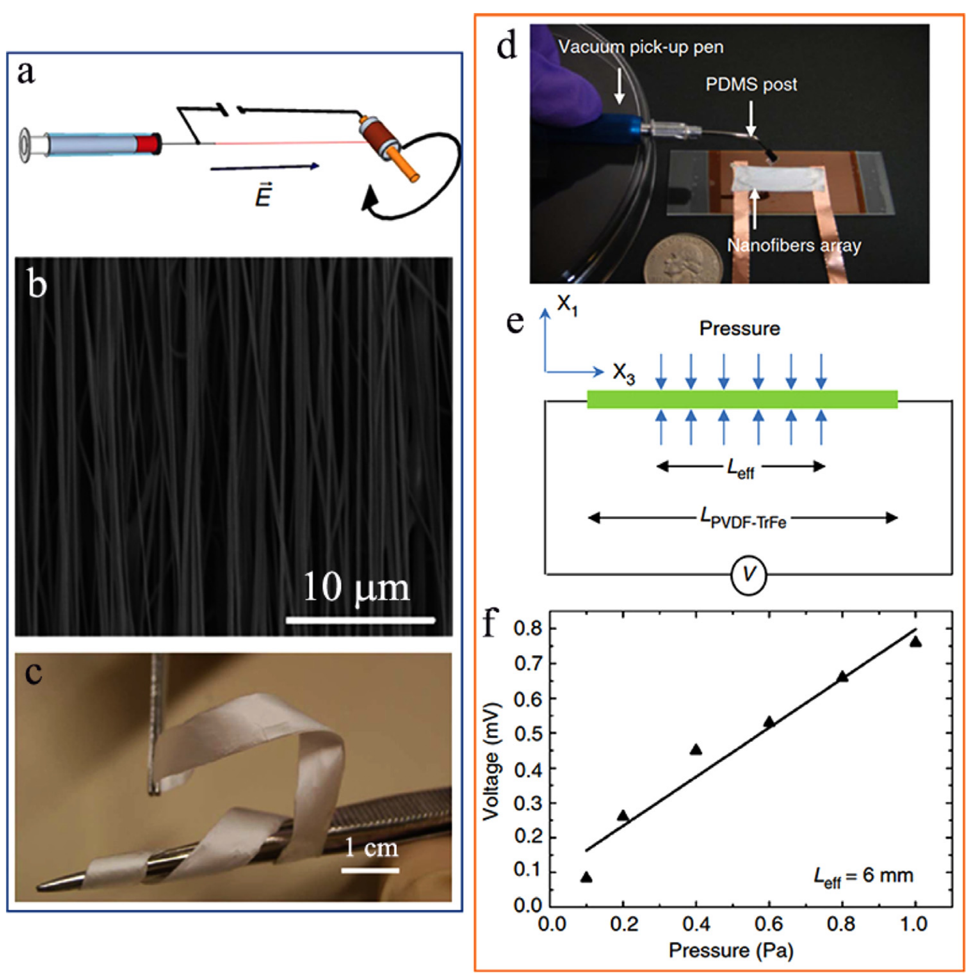

Fig. 6. (a) Schematic illustration of the experimental setup for electrospinning highly aligned arrays. (b) SEM micrograph of fiber arrays. (c) Photograph of a free-standing film of highly aligned piezoelectric fibers. Experimental and theoretical studies of responses of pressure sensors. (d) Photograph of the manipulator used to apply pressures. (e) Schematic illustration of an analytical model for the response of arrays. (f) Experimental (symbols) pressure response curve in the low-pressure regime (0.1–1 Pa). Reprinted with permission from Ref. [82], Copyright 2013, Springer Nature.

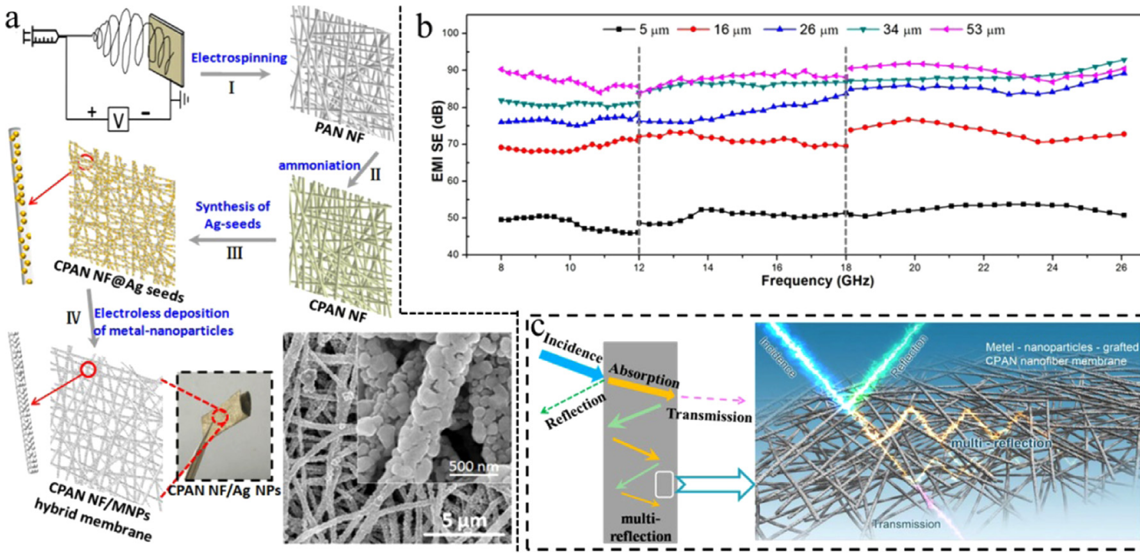

Fig. 7. (a) Schematic representation of the procedure for the fabrication of Ag nanoparticles decorated electrospun polymer nanofiber membrane. (b) EMI-shielding effectiveness of the lightweight hybrid membrane with different thicknesses. (c) Schematic description of the EMI shielding mechanism.

fiber film reached −21.5 dB at 16.8 GHz and the R_L below −10 dB was in the frequency range of 10.6–16.5 GHz with a thickness of 2.5 mm. The microwave absorption performance was attributed to the synergetic effect between dielectric loss and magnetic loss originating from PPy, PVDF and Fe_3O_4. Low-density, flexible and high-performance electromagnetic interference (EMI) shielding materials are urgently required to treat increasingly serious radiation pollution. Hsiao et al. developed lightweight and flexible reduced graphene oxide (GO)/water-borne polyurethane (WPU) composites through layer-by-layer assembly of two oppositely charged suspensions of GO onto electrospun WPU fibers, followed by the reduction by hydroiodic acid [84]. The composites exhibited substantially enhanced electrical conductivity (approximately 16.8 S m^{-1}) and showed a high EMI-shielding effectiveness (approximately 34 dB) over the frequency range from 8.2 to 12.4 GHz. A recent study in our group, we prepared a highly flexible porous electrospun polymer nanofibers (NFs) decorated with Ag nanoparticles (NPs) membrane via electrospinning and electroless plating processes (Fig. 7**a**). A superior EMI-shielding effectiveness of ≈90 dB was achieved for the lightweight hybrid membrane (53 μm) in X/Ku/K-band (Fig. 7**b** and 7**c**), which is superior to pure metal and most of the synthesized EMI-shielding materials. The excellent EMI-shielding efficiency was attributed to the high conductivity of Ag NPs and favorable porous structure in the hybrid NF membrane. In addition, the resultant hybrid membrane showed a reasonable mechanical strength and excellent flexibility.

For microwave absorbing materials with a low reflection coefficient for the electromagnetic illumination in microwave frequency, their further application is the stealth technology, which is indispensable in the modern military and detection industries. The development of a cost-effective microwave absorbents in wide bandwidth on the basis of electrospun all-nano composite materials is still a very challenging task. Besides, infrared stealth is also a major focus with increasing demands for advanced detection and stealth technology. Zhang et al. prepared freestanding membranes composed of $SiO_2@Bi_2O_3$ hierarchical core/shell fibers by a combination of two fabrication methods: electrospinning and hydrothermal reaction [85]. The infrared emission rates of the resultant $SiO_2@Bi_2O_3$ composite membranes were evaluated in comparison with pure SiO_2 fibers in 2–22 μm wavebands. It was theorized that the coating of Bi_2O_3 nanoparticles contributed to the decrease of infrared emissivity, and the infrared emission properties of $SiO_2@Bi_2O_3$ composite fibers were related to the α-Bi_2O_3 phase. The results favourably indicated prospects of $SiO_2@Bi_2O_3$ composite fibrous membranes for applications in infrared stealth camouflage.

3. Conclusion and perspective

Electrospinning affords us a remarkably easy and versatile technique for the formation of superfine fibers with multi-component and multi-structural nano composite materials with many desirable properties. Electrospinning based all-nano composite material is a rising field, which is in the basic research and primary development stage. Although researchers have developed a lot of all-nano composite materials, in addition to the gas filtration products (such as anti-$PM_{2.5}$ masks and automotive filters), other products haven't been accepted by investors and manufacturers. Medical electrospun superfine fibers are good materials (e.g. dressing, patch, etc.); however, the waiting time for medical approval and money are daunting. Polyimide nanofiber Li-ion battery separator has a good market prospect, so it is necessary to expand production and increase investment. Thus, how to do and what can be done with the superfine fiber composite materials is still a question to ponder.

A new round of scientific and technological revolution and industrial change have come to us at a rapid pace. At present, various new materials, new technologies and new industries emerge in endlessly. Following the development of the times, the industrialization of electrospinning based all-nano composite materials has begun to emerge. Based on the above statements, there are still some problems existing in the industrialization of electrospinning based all-nano composite materials. The main bottlenecks that limit the further industrialization are large scale preparation technology, materials' mechanical strength and suitable applications. Among them, applications seem to be more important. According to the current situation, there is no doubt that we should still follow the research direction of modification, substitution and intelligent manufacturing to expand the applications of electrospinning based all-nano composite materials in the next five years. Modification is to improve the intrinsic properties of the traditional materials through using superfine fibers or their composite materials as modifying materials (such as air filtration products). Substitution is taking advantage of special characteristics of electrospinning based all-nano composite materials, such as ultralight, supersoft, large specific surface area, high porosity, synergistic effect and the effect of the interfaces, to develop new nano-scale composite materials that are not yet available in the world, discover new principles and properties to replace some traditional products. For example, electrospinning based all-nano composite materials with low-density could be applied as ultra-lightweight products to replace some aerospace and aerospace material. Losing weight of one gram would bring out great economic and practical significance toward national defense. At last, intelligent manufacturing is research and development of intelligent perception, intelligent control and other key technologies, making electrospinning based all-nano composite materials being applied in the novel fields of autonomous health monitoring, marine engineering and intelligent ground equipment. We trust that with unremitting efforts, electrospinning based all-nano composite material products will come into our lives in the near future.

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