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TOPICAL REVIEW

Recent progress on the poly(arylene ether)s-based electrospun nanofibers for high-performance applications

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Abstract
Poly(arylene ether)s (PAEs) engineering plastics are a type of high-performance material which are excellent in thermal resistance, mechanical properties, and have low dielectric constant and anti-corrosion. Over recent decades, PAEs further combined with electrospinning technology to fabricate as large surface-to-volume ratio and porosity membrane materials for high-performance applications. In this review, progresses of PAEs-based electrospun nanofibers and fiber reinforced composites including separate membranes, proton exchange membranes, oil-water separation filters, bio-scaffolds and humidity sensors, etc. are presented together with their corresponding high-performance applications in the fields of battery, wastewater treatment, bioengineering and sensor. Finally, current challenges and future development directions of PAEs-based electrospun nanofibers are discussed.

1. Introduction
Poly(arylene ether)s (PAEs) engineering plastics are widely employed in aerospace industry, medical equipment, automobile production, electronic manufacturing and other high-tech fields [1–4]. PAEs were employed as high-performance polymers for their excellent thermal resistance, good mechanical properties, easy processability, low dielectric constants, good corrosion resistance and oxidative stability [1, 5]. Such properties are derived from chemical architectures characterized by the presence of alternating rigid aromatic units and flexible oxygen ether bonds in PAEs’ backbone [4, 6]. The synthesis of the first high molecular weight PAE by R. N. Johnson in 1967. After that, in addition to the basic core structure, some extra polar bonds have been introduced to enhance inherent superior performances [7]. Representative functional groups, as shown in figure 1(A), include ketone (−(C=O)−), sulfone (−(O=S=O)−) and nitrile (−(C≡N), and are respectively named as poly(arylene ether ketone) (PEK), poly(arylene ether sulfone) (PES) and poly(arylene ether nitrile) (PEN) [6].

Nowadays, electrospinning technology has gained great attention due to advantages in nanofiber fabrication, such as easy operation, cost-effectiveness, easy incorporation of functional components in one-dimensional nanofiber composites, and enhancement of some properties driven from the material stretching induced by electrical force [8, 9]. This is an ancient technology dating back to the 18th century, but it was not widely applied in the field of fiber material preparation until the middle of the 20th century due to Sir Geoffrey Ingram Taylor et al.’s research on spinning principle and process [10]. The materials used to prepare nanofibers by electrospinning technology are mainly polymers. Such as cellulose acetate [11, 12], polylactic acid [13], polyvinylidene fluoride [14] and polyvinyl alcohol [15], etc for various unparalleled advanced applications.
including high flux oil/water separation, regenerative tissue engineering scaffolds, efficient batteries separator and etc [8, 16]. A typical electrospinning system consists of three elements: an injection syringe equipped with a metallic needle, a high voltage electric field system and a metallic collector as illustrated in figure 1(B) [10]. The highly enough viscous polymer solution or polymer melt is pushed out of the syringe and stretched as it forms a Taylor cone upon the opposite charge between syringe needle and collector. At the same time, the solvent of the polymeric jet is evaporated before reaching the collector, leading to the formation of individual fibers with a typical diameter lower than 100 nm [17]. The obtained nanofibers usually have ultrahigh surface-to-volume ratio and porosity, satisfactory flexibility and controlled topography [10, 17].

Inspired by the aforementioned features, a combination of electrospinning technology and poly(arylene ether)-based engineering plastics has been developed over the past decades. These investigations opened the way to high-performance PAEs with applications in various fields, such as proton exchange membranes for fuel cell, super-hydrophobic/hydrophilic membranes for oil-water separation, sensitive devices for humidity detection and electrospun scaffolds for bio-tissue engineering [18–20]. In these applications, PAEs-based electrospun nanofibers are mainly employed as tough matrices or further functionalized by other active materials. Benefiting from the excellent intrinsic high-performance, PAEs-based electrospun nanofibers are out-performing other materials with their remarkable thermal stability, corrosion resistance and mechanical properties [21, 22].

Consequently, considering above advantages, this review presents the last developments of PAEs-based electrospun nanofibers with a particular focus on novel fabrication approaches, associated beneficial material properties, and the most recent high-performance applications.

2. Fabrication approaches for PAEs-based electrospun nanofibers

2.1. PAEs-based single component electrospun nanofibers

PEK, PES and PEN are the types of PAEs usually employed for the fabrication of electrospun nanofibers. In the common method, PAEs are dissolved in polar organic solvents. N,N-dimethylformamide (DMF) with a high boiling point (153 °C) is the most common choice of solvent [23, 24]. Besides, some medium polar organic solvents with low high boiling point such as tetrahydrofuran (THF) and dichloromethane (CH2Cl2), are often used as alternatives or one part of solvent mixtures [25]. Thus, relatively long subsequent heat treatment (typically 24 h at 80 °C) is an indispensable step to remove excess solvent. However, in order to obtain much higher thermal resistance and mechanical properties, PAEs usually polymerized with large molecular weight. Or introducing abundant conjugated segments in the backbone or side chain to generate kinds of semi-crystalline polymers. Meanwhile, the above efforts are accompanied by a limitation in solubility and ultimately a reduction in the processability of PAEs in electrospun nanofiber fabrication [26, 27]. Thus, improving the electronic spinnability and seeking approached to guarantee the inherited high-performance of PAEs are always the critical aspect of the development of PAE-based electrospun nanofibers. Various advanced strategies, including hydrophilization pre-treatment, molecular modification and compound blending are developed depending on PAEs type.

2.1.1. Poly(arylene ether ketone) (PEK)

PEK is one of the earliest and most successful commercial PAEs, with poly(arylene ether-ether-ketones) Victrex PEEK™ (P1 in figure 2) as a particular example. P1 has a deposition temperature (T5%) of 520 °C and long-term service at high temperature (240 °C), as well as satisfying tensile strength at 97 MPa [28]. At the beginning of
their development, traditional PEKs were semi-crystalline and insoluble excepted in concentrated sulfuric acid \[29\]. Therefore, the excellent stability of PEK due to its semi-crystallinity was a double-edged sword that hindered its wide application. In order to improve the processability in electrospinning technology, the synthesis of amorphous PEKs has been extensively investigated.

One of the common ways to synthesize PEK with electrospinnability is to introduce the segment with a large spatial position to forbid regular molecular arrangement in the condensed phase. Qipeng Guo et al used phenolphthalein bisphenol monomer to synthesize a novel PEK (P2 in figure 2)\[30, 31\]. Phenolphthalein (PP) monomer holds a relatively large spatial position due to non-coplanar and twisted structure. By copolymerization with bihalogen benzophenone monomer via nucleophilic substitution polycondensation, PP can lead to amorphous polymers. Hui Zhang et al investigated that P2 can be facilely fabricated as nanofibers from DMF solution via electrospinning technology \[32\]. The obtained membranes showed fiber diameter range from 300 to 600 nm and high tensile strength located at 0.35–0.39 N·m²·g⁻¹. Hereafter, researches on the construction of amorphous PEK by the introduction of distorted non-coplanar segments never stopped. Another typical monomer is 4-((4-hydroxyphenyl)-2,3-phthalazin-1-one (DHPZ) evaluated by Xigao Jian et al

Figure 2. Chemical structures of various PEKs have satisfying electrospinnability.
The obtained new class of PEKs have outstanding thermal resistance and good solubility in polar organic solvents, becoming a suitable high-performance polymer for nanofibers made by electrospinning processing [34]. In addition, fluorene is a chemical group commonly used to improve solubility. Hai Li et al used 4,4’-(9H-fluorene-9,9-diy) diphen containing PEKs (P4 in figure 2) to prepare nanofibers with diameters ranging from hundreds of nanometers to several micrometers [35]. The obtained PEKs fibers did not exhibit beads or agglomerates thanks to the excellent solubility of polymers.

The other effective way to improve polymer solubility is to introduce polar groups such as carboxyl (−COOH), amino (−NH₂), sulfonic acid (−SO₃H) and 3-trifluoromethyl (−CF₃) groups in the polymer chain in order to enhance interaction with the solvent. Cui et al. taken −CF₃ as a hydrophobic group to synthesize a novel PEK (P4 in figure 2) and further fabricated a composite membrane via facile one-step electrospinning method [36]. The as-prepared membrane had a potential application in water/oil separation due to the superhydrophobicity. On the contrary, utilization of −COOH, −NH₂ and −SO₃H are aimed to bring hydrophilicity and chemical reactivity, which can be further reinforced by a porous architecture allowed by electrospinning technology. P5 (see in figure 2) is a carboxylic-functionalized hydrophilic PEK with good solubility in organic polar solvents [37, 38]. The functional bisphenol monomer, phenolphthalein, can be facilely obtained by ring-opening reaction of phenolphthalein. Mengzhu Liu et al demonstrated that P5 owns high mechanical strength, strong water absorption with a maximum of 55 g of water/g and 72% of absorbed water remained 210 min after exposure. These results reveal the great potential of P5 as strong water-absorbent material [37]. Sulfonated PEKs have attracted attention regarding their potential application in energy storage devices such as fuel cell membranes due to their excellent proton transport capacity [39, 40]. The corresponding typical structure is shown as P6 in figure 2. −SO₃H groups can be introduced by post-sulfonation of polymer in concentrated H₂SO₄ or by polymerization of sulfonated monomers [41]. Recently, lithiated sulfonyl PEKs gained attention because the ionomers act as an efficient electrode binder which enhances the ionic conductivity and lithium diffusion upon high rate cycling processes [42, 43]. Nan Lu et al employed the lithiated sulfonyl copolymerized PEK (P7 in figure 2) as a nanofiber matrix with initial Tₛₙₙ over 300 °C and high mechanical stability, and further improved voltage stability maintain from 0 to 2.0 V [39]. Furthermore, another ether modified PEK was proposed by Zhen Li et al [44]. This PEK was first chloromethylated to carry out P8 (see in figure 2) with good solubility and then prepared as a nonwoven membrane. However, the obtained membrane was weak in strength property and unable to maintain its skeleton when used as a LIB separator. Hence, prior to application, the chloromethyl groups on the P8 membrane were converted to hydroxymethyl groups and etherified. Finally, the former polymer has been further crosslinked to form P9 (see in figure 2), which endowed the fiber membrane with insolubility and reinforced mechanical strength and stability.

2.1.2. Poly(arylene ether sulfone) (PES)

PES contains sulfonyl groups in the backbone is a type of amorphous thermoplastic high-performance engineering polymers [45, 46]. The typical structure of PES was polymerized by bisphenol A and dichlorophenyl sulfone monomers (see P10 in figure 3) in 1965 [47]. Engineering applied P10 usually has a Tₛ over 195 °C and is fatigue-resistant at 150 °C. The most successful commercial product is Victrex PESTM (P11 in figure 3) developed in 1972 [45]. P11 not only possesses high thermal resistance with Tₛ of 200 °C-220 °C, satisfying mechanical properties upon temperature ranging from −100 °C to 200 °C, but also has unique superiority in hydrolysis resistance, creep resistance, biocompatibility and transparency [2]. Moreover, PES is suitable for preparing fiber materials by electrospinning technology due to the processability improved by the sulfone group and is widely used in biomedical and water purification fields [48, 49]. The commercial P10 electrospun fiber is known as a suitable carrier with excellent thermal resistance, good mechanical properties, high permeability, biocompatibility, as well as corrosion resistance [50]. Schaefer et al incorporated P10 and β-Cyclodextrin (CD) to prepare a micropollutant removal nanofiber composite for water purification [51]. The uniform and straight nanofiber composite show diameters of nanofibers at 518 ± 135 nm, the contact angle of membranes at 134.2° with good sorption reversibility due to the easy processability, hydrophobicity and durability of PES. To improve the weak interaction of PES with other low energy materials, the introduction of functional groups (e.g. −COOH, −SO₃H, −OH) is a convenient way [49, 52, 53]. Gang Zhang et al functionalized PES with active hydroxy units, which were post hydroxylated from methoxy groups (P12 in figure 3) [54]. The existing −OH formed hydrogen bonds between polymer chains, and endowed P12 with a high Tₛ (238.8 °C). Then, they prepared the novel PES as electrospinning nonwoven nanofiber membranes that can be further chemically modified by octadecltrichlorosilane (OTS). OTS was then covalently grafted on the functional P12 fibrous membrane, and thus endows the composite membranes with superhydrophobicity and acts as a durable separation material. On the other side, Kyunghwan Yoon et al functionalized PES via mixed solvent and oxidation process [55]. They evidenced that fabrication of nanofiber by dissolving PES in mixed solvents (N-methylpyrrolidone (NMP) and DMF) can enhance the mechanical properties by inter-fiber junction points. This was caused by fusion of solvent inducted fusion via adding NMP solvent with a high boiling point. At the same time, they also provide another
strategy to realize the significant enhancement of hydrophilicity of PES fiber membranes required by water filtration applications. The water contact angles of functionalized membrane decreased from 127° to 28° through a short oxidation treatment using ammonium persulfate.

On the other hand, because of a wide range of applications and outstanding performance in harsh conditions, PES has been taken as reference materials to develop new electrospinning approaches. In addition, PES has been investigated by a novel bubble electrospinning technology, which is a type of needleless technology [56]. Bubbles of PES were produced from a tube connected to a controllable syringe pump. After that, the high voltage will give a force to draw multiple jets by overcoming the surface tensions of PES bubbles. Different from common electrospinning technology, bubble electrospinning technology will not form Taylor cone, but tow multiple jets onto the collector at the same time. Moreover, the obtained fibers are all porous nanofibers by contrast with the smooth nanofibers prepared by common electrospinning technology. On the other hand, the advanced needleless electrospinning method can greatly improve the efficiency, but it is rarely used in PAE-based electrospun fibers preparation. Because PAEs are usually hydrophobic, it is easy to block pinholes due to precipitation upon the moisture in the air during electrospinning. Needleless electrospinning technology may be the most promising strategy to solve this problem which will greatly improve the membranes production efficiency, and promote the industrial development of PAE-based electrospun nano fibers in the future [57].

Guoqing Chang et al proposed an interesting electrospinning method [58], by equipping a motor on the spinneret, the syringe can rotate at a high speed (16 000 rad min⁻¹) while spraying the polymer solution. Thus, the self-binding yarns of PES were collected and possessed 50.1% higher elongation and higher tensile strength (30.8 MPa) than that of PES nonwoven membranes fabricated by traditional electrospinning approaches.

Poly(phthalazinone ether sulfone ketone) (PPESK) is a novel copolymer that combines the advantages of phthalazine, ketone and sulfone groups. The corresponding chemical structure is shown as P13 (see in figure 3) [59]. PPESK polymers have ultrahigh T_g, ranging from 263 °C to 305 °C, good mechanical properties, chemical resistance, solubility and processability. These advantages made them widely studied, especially for ultrafiltration, proton exchange membrane and separation membranes [59, 60]. To improve interfacial compatibility, Shikai Zhang et al provide a new preparation method to improve interfacial compatibility of...
proton exchange membranes (PEM) made by sulfonated poly (phthalazinone ether sulfone ketone (SPPESK, \textbf{P14 in figure 3}) \cite{61}). PPESK was used to form SPPESK by dissolution in the sulfonating reagent. Then, SPPESK was primarily prepared as a three-dimensional fiber membrane. Next, the dry electrospun membrane was immersed in SPPESK/glycol/H$_2$O solution to fill the inter-fiber voids and then, the solvent was evaporated to obtain a dry membrane. The following investigation brought evidence that the membrane welded with fibers can be employed as a dense and defect-free PEM with much higher interfacial compatibility, relatively better water swelling resistance and resulted in higher mechanical stability and slightly higher thermal resistance than the one without inter-fiber voids filling. Xue Gao \textit{et al} proposed another novel strategy to prepare SPPESK as PEM with higher tensile strength and cell power density \cite{19}. The principle was the formation of through-plane nano-scale proton conductive channels by means of aligned electrospun nanofibers.

2.1.3. Poly(arylene ether nitrile) (PEN)

PEN, apart from PEK and PES, hangs the functional cyano group (–CN) on the side chain \cite{1}. –CN is usually employed as a functional group to enhance heat resistance without weakening the processability of the polymer \cite{62}. The first commercial PEN was PEN-ID300$^{\text{TM}}$. It is obtained by polymerization of resorcinol and 2,6-difluoromethylbenzonitrile monomers via nucleophilic substitution (P15 in figure 4). PEN-ID300$^{\text{TM}}$ has the equal comprehensive high-performance as Victrex PEEK$^{\text{TM}}$, even better thermal performance with $T_g$ at 148$^\circ$C, and a relatively high flexural strength and tensile strength of 194 MPa and 132 MPa, respectively \cite{1}. These advantages should be attributed to the strong polar –CN group, which mostly improves the interactions between molecular chains by dipole-dipole interaction that results in the promotion of thermal and mechanical properties \cite{63}. However, the most inchoate PENs were suffering from crystallization during synthesis and were difficult to obtain as stable polymer products with high enough molecule weight. Consequently, their development has been initially restricted. In the last three decades, PEN gradually received noteworthy investigations as synthesis techniques improved \cite{62, 64}. Relatively to other PAEs, PEN electrospun nanofibers are rarely studied.

In order to reduce crystallinity and therefore to improve the processability of PEN by molecular design, some noncoplanar and twisted structures, such as PP, BPA, PLL and hydroquinone sulfonic acid sodium salt (SHQ), were incorporated in the backbone. Fanbin Meng \textit{et al} used a PEN copolymer (P16 in figure 4) derived from BPA and resorcinol monomers as an electrospun fiber matrix \cite{65, 66}. Iron phthalocyanine (FePc) was mixed with P16 in DMF and then, materials with various size distributions and morphologies were fabricated by modulating the P16 concentration, electric voltage, and distance between needle and collector. The as-prepared PEN/FePc nanocomposites showed beads-in-string structures, which were further transformed into thorns-like fibers through thermal post-treatment. During the process, the PEN with high-thermal resistance offered stable stems, while the self-assembly of phthalocyanines induced the growth of thorns. Due to the 3D feature, PEN/FePc has a great potential application in fiber-reinforced polymer composites. –COOH and –SO$_3$H are the
most functional groups for endowing polymer with hydrophilicity and reactivity. Penglun Zheng et al synthesized a novel fluorescent PEN copolymerized from hydrophilic 4, 4-bis(4-hydroxyphenyl)valeric acid (DPA) segments and hydrophobic biphenol segments (P17 in figure 4) [67, 68]. The prepared nanoscale porous electrospun fibers showed blue fluorescent property, high specific surface area and were grafted with reactive – COOH. Moreover, the –COOH can be further treated with NaOH to form –COONa, which offered nanofibers with good adsorption capacity of metallic cations. These features allow applications in optical sensing and water purification. Yingqing Zhan et al taken sultoned PEN as the matrix to design efficient hexavalent chromium removal materials (P18 in figure 4) [69]. The –SO₃H groups were reacted with polypropylene by in situ polymerization to form core/shell nanofibrous membranes with properties that enhance the reusability and handling property in wastewater treatment. Inspired by these advantages, they developed a variety of functional electrospun fiber materials based on easy-hanging PEN matrices with appropriated molecular structures (such as hydrophobic P19 in figure 4) for applications in harsh environments [70, 71].

2.2. PAEs-based blends and coaxial electrospun nanofibers

In order to overcome the weakness of single component electrospun fiber and expand more functions, blending of PAEs was carried out. Blending, which means combining multiple components, is an efficient and facile way to obtain multi-functional or unique 3D-structured electrospinning fibers. There are two common strategies employed for blending. One strategy is to spin pre-mixed composite solutions by conventional electrospinning technology, the other one is to spin separated components by coaxial electrospinning technology. The first method requires good miscibility between PAEs and components to form a uniform nanofiber matrix without agglomeration like beads. To ensure this, using all PAEs-based polymers is the best choice. For example, PES and SPEEK-Na had been blended and spun as fibers with uniform morphology by Qiong Du et al [72]. In this blended system, each PAE-based polymer performed its own functions. PES performs as skeletal polymer and provides excellent mechanical strength and thermal resistance to ensure the membrane with durability, while SPEEK-Na acts as branches with hydrophilic groups to provide oil/water separation ability. Due to the good compatibility of two blended polymers, a PAE-based 3D membrane with good dimensional stability and durability was fabricated for oil/water mixtures/ emulsions separation. The second choice is to blend polymers having similar solubility to PAEs. Weiwei Cui et al proposed a composite membrane constructed by PES and poly(vinylidene fluoride) (PVDF) [73]. Prior to electrospinning, PES and PVDF were dissolved in a mixture of THF and DMF to form a homogeneous solution. Morphology of obtained fibers was smooth and beadless that evidenced the good miscibility of two blended polymers. The presence of the PES decreased PVDF crystallinity and improved thermal-dimensional stability. These features resulted in the promotion of stable cycle performance, recovery rate and capacity retention when the PES/PVDF composite membrane was applied as a separator in lithium-ion batteries. Besides, although PAEs are almost exclusively dissolved in organic solvents, there also exhibits opportunities for blending with water-soluble polymers. Kun Jia et al put forward a novel electrospinning strategy [74]. They synthesized an amphiphilic aromatic multi-block copolymer (P20 in figure 4) having –SO₃K and –COOK groups. The synthesized amphiphilic PEN was employed as a polymer ligand for lanthanide ions (Tb³⁺ and Eu³⁺). When the ions coordinate with amphiphilic PEN, the molecular chain form microparticles by self-assembly and further sensitize the intrinsic fluorescent emission of both Tb³⁺ and Eu³⁺. Then, by dispersing the microparticles in water-soluble polyvinyl alcohol (PVA), luminescent nanofibers were prepared by electrospinning approach. The PEN-based ion-microparticles seamlessly emerged in the PVA nanofiber and were stretched as spindle-like beads. This proposed an approach for blending PAEs-based materials with water-soluble polymers to form novel functional electrospun fibers.

In the second place, coaxial electrospinning is a newborn technology that originated from conventional electrospinning technology at the beginning of the 20th century [75]. It’s a physical strategy for combining two components, including one spinnable material jetted from the outer chamber outlet with another spinnable/ non-spinnable material jetted from the inner chamber outlet to form core-shell fibers [76]. Taking advantage of this, spinnable PAEs have been used to construct nanofibers with functional polymers, liquid crystals, nanoparticles, etc. and developed advanced applications [77, 78]. One of the benefits is that PAEs-based nanofibers prepared from coaxial electrospinning can encapsulate the core component steadily. Yao Wu et al reported a blend of nanofibers mixing graphene oxide (Ssi-GO) in SPEEK nanofibers via coaxial electrospinning (see in figure 5) [77]. Compared to the Ssi-GO stretched and floated in the monaxial nanofiber membrane, the coaxial spinning Ssi-GO core was encapsulated in the SPEEK shell. The fragging force also elongated the Ssi-GO with the SPEEK fiber axial direction and resulted in a unique cambiform-like morphology. Thus, the Ssi-GO exhibited wrinkling voids, which promoted water uptake and restricted membrane swelling. Of course, the important aspects of the coaxial electrospinning method provide a way to merge multiple functions by designing various core-shell structures. Qianqian Yuan et al used the same SPEEK with different degrees of sulfonation
(DS) to act as core and shell components [79]. The modulation of mechanical strength and proton conductivity by different DS has been studied in detail. The results revealed that low DS core combined with high DS shell possessed superior performances. Specifically, the SPEEK core mainly controls the mechanical properties while the shell mainly controls the proton conduction. Besides, coaxial electrospinning technology is also a popular strategy to fabricate hollow ultrafine fibers. Based on its high mechanical strength, PES becomes a good choice to construct hollow structures. To find a facile way to prepare multifunctional ultrafine fibers, Zhimei Wei et al attempted a multilayer architectural strategy [80]. By employing dilute hydrophilic polymer solution as the core component and concentrated PES as the tough shell, two layers of hollow ultrafine fibers can be fabricated by coaxial electrospinning and phase separation in one step.

3. High-performance applications

As a marriage between the highly porous electrospun nanofibers and the excellent overall properties of engineering plastics, PAEs-based electrospun nanofiber membranes integrate the merits of both materials. At present, many potential frontier applications of PAEs-based electrospun nanofibers include separator membranes of energy storage batteries, proton exchange membranes, oil/water separation membranes, biological tissue materials and humidity sensors have been widely investigated.

3.1. High efficiency and dimension stable membranes for battery

3.1.1. Lithium-ion batteries separator

The separator is one of the critical components in lithium-ion batteries (LIBs) which physically separates the electrodes to prevent short circuits of the LIBs, at the same time, allowing the transports of Li\(^+\) between electrodes [81]. The chemical and physical properties of the separator significantly affect the storage of liquid electrolytes. An efficient LIBs system requires a separator possessing appropriated mechanical properties, chemical resistance, thermal resistance, porosity and wettability to ensure LIBs with high operational efficiency and safety. At present, most commercial separators for LIBs are polyolefin porous membranes based on polypropylene (PP) and polyethylene (PE), which are widely used due to their suitable mechanical properties, excellent chemical resistance, and appropriate thickness. Nevertheless, such separators do not have sufficient thermal stability, wettability and porosity. These separators’ weaknesses are limiting actual LIBs’ capacity, service life and safety [44].

Combining the thermal resistance of high-performance engineering plastics with the high porosity of electrospun materials, PAEs-based polymers are potential alternatives for LIBs polymer separators. Wenzheng Gong et al employed PPESK to fabricate electrospun nanofiber membranes with high strength by hot-pressing two pieces of membranes into one [82]. The obtained PPESK electrospun nanofiber membranes possessed satisfying tensile strength of 22.8 MPa and thermal stability of 200 °C. Thus, the PPESK separators will not melt or shrink at high working temperatures. Moreover, PPESK electrospun membranes have high porosity of 70%, electrolyte uptake of 525% and interfacial resistance at 268 \(\Omega\) which results in excellent ionic conductivity of 1.39 mS cm\(^{-1}\). Furthermore, they used coaxial electrospinning technology to fabricate PPESK fibrous membrane composited with PVDF to improve performance [83, 84]. The PPESK/PVDF has a thermal shutdown temperature of 170 °C, better electrolyte uptake and ionic conductivity than that of commercial PP/PE/PP separators by virtue of the comprehensive properties of polymer composite. Indeed, PPESK/PVDF solved the problem of narrow interval temperature between shutdown and failure point of commercial PP/PE/PP tri-layer
composite separators. The PP layer will melt when the internal temperature of the LIBs is over 130 °C, then, block the micropores and circuit the battery. PPESK/PVDF expanded the interval temperature by 40 °C which improved the safety issue of LIBs.

PAEs-based polymer composites with inorganic nanoparticles will lead to a separator membrane which not only has high thermal stability but also has enhanced affinity with liquid electrolytes. Hai Li et al used crystalline Al₂O₃ to modify PEK electrospun nanofiber membranes [35]. In comparison to the commercial polyolefin separators, PEK-Al₂O₃ almost showed no shrinkage at a high temperature (150 °C for 1 h) as shown in figure 6(A). The blending separator also exhibited high degradation temperatures over 500 °C, superior electrolyte wettability, as well as high electrolyte uptake of 561%. And the most important is the high discharge capacity of 158.2 mAh g⁻¹ during 100 cycles (see in figure 6(B)). Meanwhile, Hai Li et al also developed a series of fluorinated PAE-based separators, fluorinated poly(ether ether ketone) (FPEEK) [85] and fluorinated poly(aryl ether ketone) (FPPEK) [86] to improve safety and electrochemical properties. The fluorinated PAEs showed good compatibility with liquid electrolytes. Thus, both fluorinated PAE-based separators possessed higher electrolyte uptake than commercial PP separators. They showed better electrochemical properties with superior electrolyte uptake at 559% and 514%, respectively. Notably, they inherited the excellent thermal resistance and mechanical strength of PAEs. Especially, fluorinate FPPEK exhibited outstanding anti-shrinkage properties for 1 h at 150 °C without any disintegration before 500 °C. While FPEEK showed a strong tensile strength with a value of 27.7 MPa. Besides, the ionic conductivity of both separators reached 3.12 mS cm⁻¹ and 3.15 mS cm⁻¹, respectively. It has been demonstrated that fluorinated PAEs-based separators are promising high-performance substitutes allowing the production of safer and more durable LIBs.

3.1.2. Proton exchange membranes
Proton exchange membranes fuel cells (PEMFCS) have gained great interest due to their advantages of compatibility, reliability and high energy efficiency [87]. One of the most important parts of PEMFCs is proton exchange membranes (PEMs) which act as the key role in transport proton. At present, the commonest commercial PEMs are based on perfluoro sulfonic acid including Nafion™ and Aquion™. However, they suffer several disadvantages including high cost, excessive methanol permeability, and low conductivity due to obvious water loss at high internal temperatures [41]. To break through the mentioned limits, in recent years, sulfonated PAE-based polymers have taken place as a promising class of alternative PEMs materials due to low cost, lower methanol permeability, satisfying mechanical strength and excellent thermal stability at high temperature [88]. For this reason, various modified sulfonated PAE-based PEMs have been developed for PEMFCs applications.

Forming organic-inorganic composite membranes is also an effective method to improve mechanical-thermal stability while reducing the dimensional swelling of PEMs. Chanmin Lee et al prepared a SiO₂/SPEEK composite electrospun membrane and used it for a supporting skeleton in the Nafion matrix to form a dense film [89]. The micrograph results showed that impregnated SiO₂/SPEEK adhered well to Nafion (see in figures 7(A)–(C)). The commercial Nafion PEM is unstable and collapses to lose the function of proton conductive over 80 °C. By contrast and due to the superior mechanical and thermal resistances inherited from SPEEK, the obtained composite membrane performed better than Nafion PEM at temperatures as high as 120 °C and 40% relative humidity (RH). The maximum current density of composite membrane was 0.44 A cm⁻² at 120 °C and 40% RH (figure 7(D)). The maximum power density was 170 mW cm⁻², which is 2.4-fold than the

![Figure 6. Thermal shrinkage images of PP (a) and (d), PEK (b) and (e), PEK-Al₂O₃ (c) and (f) separators at 25 °C and 150 °C after 1 h, and the corresponding discharge cycling performance. Reproduced from [35] with permission of The Royal Society of Chemistry.](image-url)
one of recast Naion film (see in figure 7(E)). Moreover, self-crosslinking of polymers offers additional benefits. Carlo Boaretti et al embedded pre-crosslinked sulfonated SPEEK (SPEEK_CX) electrospun nanofibers in an Aquion casting matrix to form composite membranes for PEMs application [90]. The crosslinking led to the high stiffness of porous SPEEK nanofiber. Compared SPEEK_CX/Aquion membranes with Aquion membranes and non-crosslinked sulfonated SPEEK/Aquion membranes, the water uptake and dimensional swelling properties (figure 7(F)) demonstrated that SPEEK_CX/Aquion membranes possessed similar water uptake. However, the area swelling and volume swelling properties of SPEEK_CX/Aquion membranes were superior to others with the non-crosslinked component, which indicated that the incorporation of crosslinked SPEEK highly limit the swelling of membranes. As depicted in figure 7(G), the stabilized SPEEK_CX/Aquion membranes also exhibited a significant improvement in mechanical properties.

Enlightened by the intrinsic blocking effect of inorganic additives, a novel type of SSi-GO/SPEEK composites membranes were prepared via coaxial electrospinning technology by Wu Yao et al [77]. For conventional fuel cells fueled by hydrogen and methanol, some of the fuel can penetrate into the PEM to reduce the performance of the cell. However, SSi-GO/SPEEK PEM showed a lower hydrogen permeation current of density due to the good steric hindrance of the SPEEK, the wrinkled and cambiform-like microstructure of SSi-
GOs. Specifically, it is 39% hydrogen permeation compared to commercial Nafion 115. Meanwhile, the covalent electrospun membranes also possessed lower methanol permeability at $6.8 \times 10^{-5}$ S cm$^{-1}$ m$^{-1}$. This is about 11 times higher than Nafion 115. The novel confinement strategy combined the intrinsic blocking effect and tough mechanical properties of SPEEEK by multifunctional via designing of core-shell structures. And has been demonstrated to be a potential high-performance PEM. In addition, quantum dots, an ideal candidate for hybridization, has also been utilized for PAEs-based nanofibers to investigate the high-performance application of PEM. Li Ping et al in virtue of the advantages of QDs and PAEs to fabricated a novel type of QDs/SPEEK hybrid nanofiber membranes [25]. QDs enhanced the through-plane and in-plane proton conduction of the hybrid membranes. Especially, high conductivity of through-plane reaching 456 mS cm$^{-1}$ at 90 °C. And the amino and imino groups hanging on filled QDs combining with -SO$_3$H groups in SPEEK resulted in acid-base pairs, which further resulted in low-barrier pathways for protons. As a thermostable matrix with $T_g$ around 220 °C, SPEEK also endowed the hybrid nanofiber membranes with the potential to develop as a favorable elevated-temperature hydrogen fuel cell.

In total, the current evaluations have led to the conclusion that PAEs-based electrospun nanofiber membranes are a novel type of alternative PEMs materials. The advantages of high thermal resistance and mechanical properties belonging to PAEs can solve the stability of the battery in high temperature conditions to a certain extent. Nevertheless, some requirements including wettability and ionic exchange capacity need PAEs attached with some hydrophilic functional groups which lead to a loss of stability. Especially the sulfonated PAEs have lower methanol permeability and high ionic exchange capacity but are limited by the remarkable dimensional. Some efforts have been done, however, the invention of comprehensive PAEs-based electrospun nanofiber membranes with high efficiency and dimension stability still needs to be further explored for the battery field.

3.2. Durable membranes for wastewater treatment

Electrospun polymer micro/nanofiber membranes are natural candidates for wastewater treatment applications due to the advantages brought by large porosity, high surface-to-volume ratio, interconnected pore structures and also cost-effectiveness [69, 71]. With the incorporation of additional functionalities, PAEs-based electrospun nanofibers featuring thermal-chemical stability, durability and nano-porous structures became high-performance materials suitable for harsh conditions [22].

3.2.1. Oil/water separation membranes

The selection of electrospun membranes in oil/water separation applications depends on their wettability [16]. PAEs-based electrospun membranes are naturally hydrophobic due to the elementary chemical structure rich in aromatic rings and ether bonds. The OTS/PES nanocomposite electrospun membranes mentioned before are superhydrophobic membranes with potential applications in oil/water separation [21]. The hydrophobicity of PES was further reinforced by OTS, which resulted in a large water contact angle (CA) at 159.2° and a low threshold sliding angle (TSA) of 7.8°, with no reduction in thermostability and corrosion resistance. In oil/water mixtures and emulsions separation experiments, OTS/PAES electrospun membranes showed a gravity-driven flux of 7260–8720 L m$^{-2}$ h$^{-1}$ with a super large water rejection over 99%. By the same strategy, Shuangjiang He et al grafted hydrophobic TiO$_2$ nanoparticles on the electrospun nanofibers of PEN [70]. The obtained TiO$_2$/PEN showed much higher CA at 165 ± 2.4° and water rejection at 99.75 ± 0.12% than single PEN and super high separation flux of 6850 ± 270 L m$^{-2}$ h$^{-1}$ and 3530 ± 230 L m$^{-2}$ h$^{-1}$ for surfactant-free emulsions and surfactant-stabilized emulsions, respectively. Above all, PEN endowed the composite separation membranes with outstanding stability against physical damages, extreme temperatures, corrosive atmospheres and some other harsh environments. Moreover, Zengduo Cui et al employed fluorinated silica pellets compositied with PEK to fabricate novel super-hydrophobic membranes, maximum CA at 157.0 ± 4.3° [36]. Remarkably, the composite membranes exhibited high durability and self-cleaning ability combined with an efficient separation ability through a 200 h water flushing test.

On the other hand, modified PAEs-base electrospun membranes will turn to be hydrophilic materials while maintaining the initial comprehensive performance. Yingqing Zhan et al prepared PEN nanofiber membranes coating with halloysite nanotubes intercalated graphene oxide (GO) furtherly modified with polydopamine (PDA) to be used as a novel sewage treatment separator [22]. The composite membrane with super-hydrophilic feature and underwater super-oleophobicity was well-endowed by porous structure and polydopamine coating (see in figure 8(A)). Therefore, PEN composite membrane system possessed permeate flux of 1130.56 L (m$^{-2}$ h$^{-1}$) and a preferable oil rejection ratio over 99%, as well as antifoaming property for various oil/water systems. As shown in figure 8(B), reusability studies further demonstrated that the membrane can serve at high temperatures (up to 90 °C) and in strong acid and alkali solutions, which shows the ability of durable and reusable.
3.2.2. Pollutant removal membranes

PAEs-based electrospun membranes have remarkable adsorption properties due to their large specific surface area and the possibility of specific binding. This superior adsorption capacity gives them unique advantages in terms of metal ions, dyes and microorganisms absorption. Functionalization approaches mainly focus on surface modification and physical blending. Chuanyan Lv et al reported a facile strategy to fabricate positively-charged PES electrospun nanofibers membranes for bacteria and cationic dyes removing \cite{91}. As shown in figure 9, poly(2-(methacyloyloxy)-ethyl trimethylammonium chloride) (PMETAC) was chosen as antimicrobial cationic polyelectrolyte component to endowed PES with positively-charged surface and bacterial growth inhibition properties. After in situ crosslinking polymerization between PES and PMETAC, the composites solution was directly used for the electrospinning process. The easily obtained positively-charged PES membranes showed high absorption capacity and selectivity toward anionic dyes in wastewater (see in
also toxic toward pathogens. Aloe vera regeneration. On account of the complexity of biological applications study, there corresponding PAEs-based reached a very high value of 208 mg g$^{-1}$. Meanwhile, PMETAC modified PES membranes possessed high bacteriostatic performance towards E. coli and S. aureus with the maximum antibacterial activity of nearly 99% (see in figures 9(C)-(D)).

Making composites of hierarchically structured membranes is an efficient method to overcome the shortages of commercial ultrafiltration devices such as fouling and low flux during long-time service. In this concern, electrospun nanofibers become prior chosen materials regarding their possible internally connected porous structures and high surface area than ultrafiltration membranes, which are fabricated by phase inversion with dense layer structures. Yingqing Zhan et al designed PEN/bioinspired polydopamine (PDA) coated GO composite (PEN/GO-PDA) as a hierarchically structured electrospun membrane for durable and efficient anionic dyes separation [20]. Taking advantage of the hydrophilicity of PDA and GO components and of mechanical-thermal stability of the PEN electrospun skeleton, the prepared PEN/GO-PDA demonstrated high mechanical strength (28.1 MPa) in addition to high-temperature resistance. Hierarchically structured membranes exhibited a good rejection rate (92.6%) and high permeate flux (141.5 L (m$^2\cdot$h$^{-1}$)) towards Direct Blue 14 at 90 °C. Meanwhile, the reusability of prepared membranes was satisfying with a flux recovery ratio of 95% after recycling 3 times for 9 h. Besides, Yingqing Zhan et al also designed another efficient composite electrospun membrane for hexavalent chromium (Cr(VI)) absorption and removal [69]. The core-shell membrane was made of a PEN core and a polypropylene (PPy) shell that is combined using electrospinning technology followed by in situ polymerization. PEN/PPy nanofiber membranes presented a high (Cr(VI)) adsorption capacity of 165.3 mg g$^{-1}$ with good reusability by 4 times adsorption/desorption cycles.

Undoubtedly, the PAEs-based electrospun nanofiber membranes have shown excellent reusability in the wastewater treatment field, especially in harsh conditions including extreme temperatures and corrosive surroundings. So even though the oil/water emulsion permeate flux and pollutant adsorption capacity of modified PAEs-based electrospun nanofiber membranes are not yet comparable to that of other desirable materials, such as polyimide (PI) [92], polyacrylonitrile (PAN) [93] and cellulose materials [11], the relative researches are still going on. The future investigation should focus on the enhancement of performance of filtration and adsorption, and also advanced multi functionalities, for example, self-healing and self-cleaning ability.

3.3. Biocompatible scaffolds for tissue regeneration
In recent years, PAEs have been testified as biocompatible with tissue-like mechanical strength and have further been applied in several biological areas, such as tissue biology, bio-imaging and drug delivery [5, 80, 95]. The PAE-base nanofiber membranes with large specific surface area and porosity can form a three-dimensional structure and provide a stable biocompatible scaffold. Thereinto, PES has been applied in bone tissue engineering (BTE). PES membrane scaffolds are designed for cell attachment, anchorage, differentiation, proliferation and further functionalities due to their hydrolytic properties, mechanical-thermal stability and oxidative stability [96]. Taking advantage of these characteristics, Mahshid Sadat Kashef-saberi et al co-electrospun PES, poly(vinyl alcohol) (PVA) and platelet rich plasma (PRP) as PRP/PES/PVA blend scaffolds [97]. After systematic experiments, the osteogenic differentiation of human adipose-derived mesenchymal stem cells showed high proliferation and adhesion on PRP/PES/PVA blend scaffolds. Meanwhile, PEK is known as a type of implantable artificial bone because of its elastic modulus tailored to match natural cortical bone [98]. To overcome the weakness that is the surface bio-inertia of PEEK, Fan Zhao et al reported a novel flexible electrospun membrane made by blending of sulfonated PEEK (SPEEK) and polycaprolactone (PCL) [99]. The hydrophilicity and tensile strength were enhanced by the introduction of the PCL component so that protein adsorption has been improved compared to the one of SPEEK electrospun membrane. In addition, the results of alternated dipping mineralization showed that SPEEK/PCL composite possessed a much higher capacity of mineralization than SPEEK. Flexible SPEEK/PCL composite electrospun membrane, allowing protein attachment and mineral compounds generation, demonstrated their great potential in BTE applications. In addition to in vivo BTE, applications of SPEEK scaffold are also explored for in vitro and in vivo skin cell proliferation. Rajalakshmi Ekambaram and Sangeetha Dharmalingam fabricated an electrospun biomimetic aloe vera/SPEEK nanofiber scaffold [100]. The in vitro analysis evidenced that aloe vera/SPEEK composite is non-toxic for skin tissues, while in vivo assays in the Wistar rat model revealed that aloe vera/SPEEK had healing properties with the ability to close wound completely and without scars. These properties were attributed to the presence of $-$SO$_3$H groups on the SPEEK molecular skeleton and to the therapeutic bio-activity of aloe vera. The $-$SO$_3$H groups endowed the membrane with a higher capacity to absorb moisture and pus from wounds, and are also toxic toward pathogens. Aloe vera/SPEEK is therefore a promising nanofibrous scaffold for skin wound regeneration. On account of the complexity of biological applications study, there corresponding PAEs-based electrospun membranes still have a wide range of fields based on the tissue-like mechanical strength to explore.
3.4. High-sensitive fibrous sensors

PAEs-based electrospun membranes have unique superiority in sensors applications \[101\]. Advantages of excellent mechanical-thermal stability, chemical resistance and large specific surface area make them ideal frameworks to hold responsive materials. Moreover, their high porosity can improve signal transmission \[102\]. Mengzhu Liu et al took carboxylic acid-functionalized PEK as templates to form 3D copper oxide (CuO) network membranes \[38\]. Benefitting from the electrochemical properties of CuO and the enlarged surface area of the PEK skeleton, the composite membranes were efficient tools for non-enzymatic glucose detection. Compared with traditional CuO based glucose sensors, this 3D network structured sensor demonstrated higher sensitivity, better anti-interference ability and faster response.

PAEs modified with –SO3H groups also have a potential application for humidity detection \[103\]. Jonghyun Choi et al first reported a novel humidity sensor based on SPEEK electrospun nanofibers with different sulfonation degrees \[104\]. The results showed that water absorption and desorption are related to the sulfonation degrees. At the lowest sulfonation degrees, SPEEK performed at relative higher sensitivities (Log Rdc (\(\Omega\))/RH (%) = −0.06952 and Log Ca(F)/ RH (%) = 0.07078) with linear response to changes in RH between 30% and 80%. Moreover, the SPEEK humidity sensor also showed a fast response (75 s), as well as flexibility, durability and wireless abilities. Xifang Li et al further made SPEEK electrospun nanofibers with PVB to modify the morphology of the nanofibers \[40\]. Figures 10(A)–(B) illustrated the performance and response-recovery behaviors of the sensor. The purpose is to fasten the response and recovery time of SPEEK-based humidity sensor by the mean of water modules and protons control. Moreover, hydrogen bonds formed between –OH of PVB and –SO3H of SPEEK accelerate proton transfer and also improve long-term service stability. Thanks to the wide working range of RH, small hysteresis of 2.68%, fast response time below 1 s and recovery time below 5 s toward humidity, the SPEEK/PVB composite nanofibers are good candidates for high-performance breath monitoring applications. Although PAEs-based electrospun nanofibers sensor applications are rare, the study of employing them as responsive components or scaffolds for various sensor devices with thermostatility and durability is imaginative and fascinating.

4. Conclusion and future scope

In this review, we provided a summary of the recent progress on PAEs-based electrospun nanofibers. The above content is divided into two parts: first, various chemical/physical modifications that bring new functionalities to PAEs processed by electrospinning technology, and second, the typical high-performance applications in technological fields. Generally, two methods, molecular designing and physical blending are employed to fix the double-edged sword of PAEs, superior mechanical-thermal stability accompanied by extreme solubility associated with crystallinity. Introducing non-coplanar and twisted monomers in structures can improve the processability of PAEs without a significant decrease in mechanical-thermal stability. Grafting chemical groups on the polymer chain backbone is another convenient way to improve solubility and forbid crystallization in condensed phase while also providing anchoring points for further functionalization. Moreover, the diameter of PAEs electrospun nanofibers being in nano-micrometers scale, the bonding force between the fibers is weak, and fibers easily slip from each other and their system deform upon stretching. Then, PAEs nanofibers’ mechanical...
strength is usually unsatisfying regarding potential applications. The most reported method is post-thermal treatment during which stacks of fibers can melt slightly at high temperatures and adhere together. This is efficient but comes at a cost of porosity. Cross-linking reactions have also been reported as a post-crosslinking treatment on prepared nano fiber membranes. In situ cross-linking reinforced the dimensional stability but processing can be complicated. All the efforts aimed to combine the high-performance of PAEs with the advantages of electrospun fibers. At present, PAEs-based electrospun nanofibers have been widely studied in the field of energy, environment and biology. The reports mentioned above have demonstrated that PAEs-based electrospun nanofibers are expected to be efficiently used as: a potential alternative separator of LIBs with superior anti-shrinkage property, a promising kind of membrane for PEMs with low methanol permeability and satisfying mechanical-thermal stability, a durable high efficient filter for oil/water treatment in the harsh environment, a biocompatibility scaffold for tissue bioengineering, as well as a reusable and sensitive sensor device.

Despite numbers of advanced reports, PAEs-based electrospun nanofibers still belong to a new field with a lot of ongoing studies related to various field applications. First, after dealing with spinnability, the decline in dimensional stability due to fibrosis is still a problem of PAEs-based electrospun nanofibers. We suggest paying more attention to the innovation of electrospinning technology and post-treatment, for the better combination of high-performance of PAEs and advantages of electrospun nanofibers. Second, at present, most applications are still at the primary research stage. The preliminary evaluations of ions battery separators and fuel cell PEMs applications are quite positive. However, their commercial applications are still a long way off. The next priority is to open a new phase to realize high-performance in actual applications in the clean energy field. For oil/water treatment membranes, there needs to enhance the performance of filtration and absorption. And also, a way need be found for the efficiently and rapidly produce filter membrane on a large scale under the premise of functionality maintaining. For biological and sensor applications, the relevant studies are relatively few and more creative explorations of PAEs electrospun nanofibers possibilities are needed. The biodegradability of PAEs-base bioengineering materials needs to be improved, and reusability and hand-held design as sensor device also need to be investigated. The future prospect of the above was also presented in figure 11. PAEs-based electrospun nanofibers represent a new class of high-performance nano-fibrous materials. We believe that the previously present study will be concretized into fascinating potential applications and ultimately lead to successful commercialization with subsequent efforts.

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