Recent advances and remaining barriers to the development of electrospun nanofiber and nanofiber composites for point-of-use and point-of-entry water treatment systems

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List of Acronyms

2-CEES	2-chloroethyl ethyl sulfide
2-CP	2-chlorophenol
ANSI	American National Standards Institute
AOP	Advanced oxidation process
BPA	Bisphenol
CBZ	Carbamazepine
CIP	Ciprofloxacin
CMT	Cimetidine
CNF	Carbon nanofiber
CNT	Carbon nanotube
CTAB	Cetrimonium bromide
DOM	Dissolved organic matter
ENF	Electrospun nanofiber
EPA	Environmental Protection Agency
Fh	Ferrihydrite
GAC	Granular activated carbon
GFH	Granular ferric hydroxide
kV	Kilovolt
MB	Methylene blue
MCL	Maximum contaminant level
MF	Microfiltration
MOx	Metal oxides
NF	Nanofiltration
NFs	Nanofibers
NPs	Nanoparticles
NSF	National Science Foundation

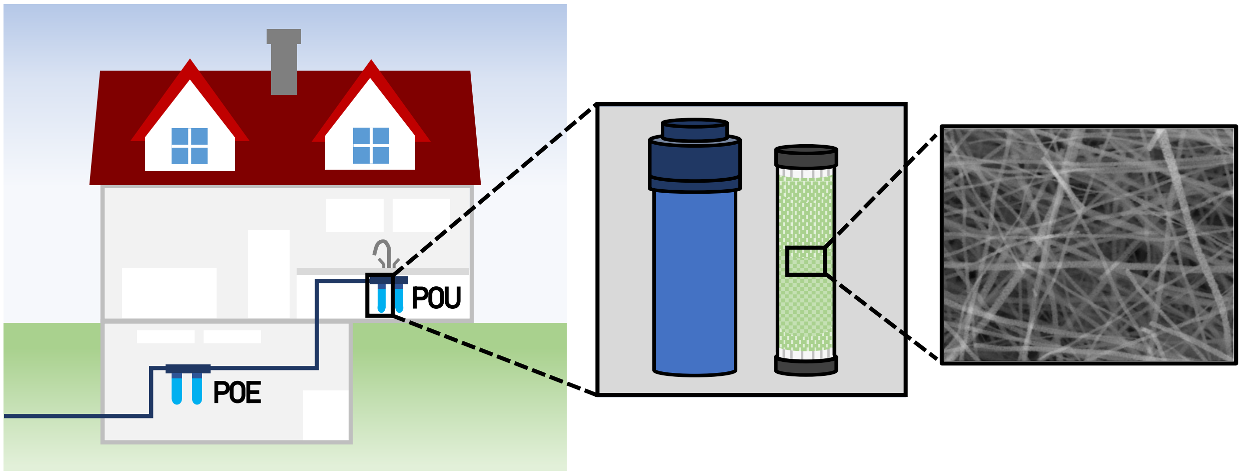
PAA	Polyacrylic acid
PAC	Powdered activated carbon
PAN	Polyacrylonitrile
PFAS	Per- and polyfluoroalkyl substances
POE	Point-of-entry
POU	Point-of-use
PVA	Polyvinyl alcohol
PVDF	Polyvinylidene fluoride
PVP	Polyvinyl pyrrolidone
QAS	Quaternary ammonium surfactants
RO	Reverse osmosis
SAC	Strong-acid cation
SBA	Strong-based anion
SDS	Sodium dodecyl sulfate
TBAB	Tetrabutylammonium bromide
TiP	Titanium isopropoxide
UF	Ultrafiltration
UV	Ultraviolet
WAC	Weak-acid cation
WBA	Weak-based anion
WHO	World Health Organization

Abstract

In this review, we focus on electrospun nanofibers as a promising material alternative for the niche application of decentralized, point-of-use (POU) and point-of-entry (POE) water treatment systems. We focus our review on prior work with various formulations of electrospun materials, including nanofibers of carbon, pure metal oxides, functionalized polymers, and polymer-metal oxide composites, that exhibit analogous performance to media (e.g., activated carbon, ion exchange resins) commonly used in commercially available, certified POU/POE devices for contaminants including organic pollutants, metals (e.g., lead) and persistent oxyanions (e.g., nitrate). We then analyze the relevant strengths and remaining research and development opportunities of the relevant literature based on an evaluation framework that considers (i) performance comparison to commercial analogs; (ii) appropriate pollutant targets for POU/POE applications; (iii) testing in flow-through systems consistent with POU/POE applications; (iv) consideration of water quality effects; and (v) evaluation of material strength and longevity. We also identify several emerging issues in decentralized water treatment where nanofiber-based POU/POE devices could help meet existing needs including their use for treatment of uranium, disinfection, and in electrochemical treatment systems. To date, research has demonstrated promising material performance toward relevant targets for POU/POE applications, using appropriate aquatic matrices and considering material stability. To fully realize their promise as an emerging treatment technology, our analysis of available literature reveals the need for more work that benchmarks nanofiber performance against established commercial analogs, as well as fabrication and performance validation at scales and under conditions simulating POU/POE water treatment.

1. Introduction

Because of high-profile instances of widespread industrial pollution (e.g., contamination from per- and polyfluoroalkyl substances or PFAS) and decaying water infrastructure (e.g., lead contamination in Flint, Michigan), the United States has witnessed a growing distrust over tap water quality, especially in communities of color.[1,2] A large number of Americans also rely on unregulated water sources (e.g., domestic or private drinking water wells), while many still lack routine access to a reliable piped water supply.[3] These challenges have led many consumers to seek out alternative commercial supplies (e.g., bottled water) and/or other means to improve the quality of water available to them.[4,5]



POU/POE device in home setting **POU housing and cartridge** **Electrospun nanofiber mesh**

Figure 1. Schematic of electrospun nanofiber membrane-integrated point-of-use (POU) and point-of-entry (POE) devices in a household. Insets include diagrams of the housing and cartridge of a POU device and an SEM of an electrospun nanofiber mesh for a potential nanofiber membrane-integrated POU cartridge. Adapted from Tap Score.[6]

When properly installed, operated, and maintained, decentralized technologies can play an important role in helping consumers secure safe and reliable drinking water. Point-of-use (POU) and point-of-entry (POE) devices are treatment technologies that treat water at the location of primary use (e.g., the tap) or upon entry into a building, respectively (**Figure 1**).[7]

With the noted challenges that many consumers still face in securing their drinking water supply, increasing reliability, affordability, and access to POU/POE devices could, in certain cases, address these concerns.[8] Widely used options for POU/POE treatment include activated carbon filters, ion exchange devices, and reverse osmosis membranes, with devices in the US certified by outside agencies to ensure a certain level of performance (e.g., National Science Foundation (NSF) International and the American National Standards Institute (ANSI) are two private organizations that issue standards to certify the efficacy of in-home water treatment devices).[9,10] Nevertheless, all commercially available POU/POE devices have limitations, including cost of acquisition and the need for routine maintenance and replacement to ensure performance. There are also persistent questions of equity and justice surrounding POU/POE devices; can those that most often need these technologies afford them?

Decentralized water treatment offers an opportunity for the integration of nanotechnology, potentially enhancing performance and lowering the cost of, and thereby increasing access to, POU/POE technologies. Engineered nanomaterials exhibit more surface area per unit mass than their larger-scale material analogs, which can equate to longer operational lifetimes and smaller technology footprints more suitable for in-home use.[11–13] The potential for greater inherent reactivity relative to larger-scale materials may also help with the removal of traditionally recalcitrant pollutant classes. However, nanotechnology-enabled water treatment devices are not without their own set of challenges.[14] It can be reasonably concluded that the use of nanoparticle suspensions or dispersions is simply not practical, especially in POU/POE treatment. Alternative reactor assemblies also suffer from limitations (e.g., packed columns of nanomaterials are likely infeasible due to large pressure head),[15–19] and all applications include the potential for unwanted release of nanomaterials into the treated supply.

Notably, to the best of our knowledge, no known commercially available POU/POE certified through NSF International or ANSI uses engineered nanomaterials or nanomaterial composites.

In this review, we focus on one particularly promising type of nanoengineered material for POU/POE applications: electrospun nanofiber composites. Electrospinning is an industrial viable fabrication route for the production of nonwoven membranes ideal for water treatment applications.[20–22] Critical evaluation of the literature shows that the laboratory performance of electrospun materials is well established (as evidenced by several recent reviews on different nanofiber applications),[23–27] and that they generally behave as would be expected from the wealth of complementary literature on more conventional membranes, engineered nanomaterials, and their composites. Thus, the primary goal of this review is to identify and establish the unique attributes of various electrospun nanofiber formulations that may make them suitable, if not preferred, alternatives to more traditional materials used in commercially available, certified POU/POE devices. We will focus on literature that has demonstrated the use of electrospun nanofibers and their composites for targets relevant to POU/POE treatment including regulated and emerging organic pollutants, distribution system derived metals, and typically recalcitrant oxyanions. Where possible, we will prioritize results from studies conducted using reactor assemblies (e.g., flow through devices) that are representative of POU/POE treatment, thereby allowing preliminary assessment of application performance and longevity. Finally, we will conclude by discussing some emerging areas of nanofiber application for water treatment, while also highlighting research needs and future challenges associated with translating promising laboratory studies into commercially available, nanofiber-enabled POU/POE technologies.

While there have been several recent reviews on various environmental applications of electrospun nanofibers,[23–27] to the best of our knowledge this is the first to critically evaluate the existing literature to assess their potential for the specific application of decentralized water treatment using POU/POE technologies and identify future research needs to realize their POU/POE application at scale. Because certification of POU/POE technologies requires strict adherence to water quality and testing conditions, while focusing on select pollutant targets most relevant to decentralized water treatment, we believe there is need for a review that focuses specifically on the existing state-of-the-art and future research priorities for this intended use application.

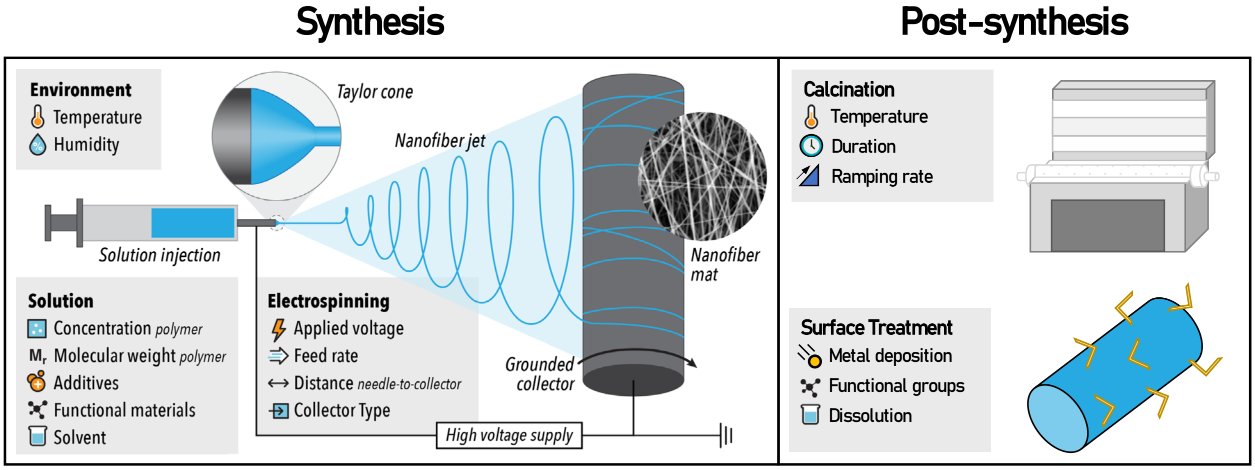


Figure 2. Schematic of electrospinning system and tunable parameters at various stages of synthesis that can be used to control nanofiber physical and chemical properties.

2. Overview of electrospun nanofibers for use in water treatment

Electrospinning is a versatile fabrication process that produces three-dimensional networks of nonwoven fibers with nm to µm size ranges in diameter.[28,29] During the synthesis process (**Figure 2**), a polymeric sol-gel precursor solution is ejected from a spinneret under a high potential (kV), and it is stretched and elongated under a whipping motion until reaching a grounded collector, forming fine fibers.[30] As shown in Figure 2, synthesis variables provide a high degree of tunability in the resulting fiber properties. Parameters including the concentration of the polymer in the sol-gel, the applied voltage, the feed rate of the syringe, and the distance between the spinneret and grounded collector all can be used to tune fiber morphology and properties. Additives including nanoparticles and surfactants can also be integrated into the sol-gel to alter fiber composition and impart new types of surface groups to influence performance. After synthesis, calcination can be used to transform organic polymer precursors into inorganic fibers, while various forms of chemical post-processing can also be used to tailor surface chemistry. Through careful control of these variables, electrospinning can be used to produce a variety of membrane architectures, from pure polymers to functionalized polymer-nanoparticle composites (**Figure 3**) that we and others have argued are ideal for use in hybrid filtration platforms capable of removing contaminants using both physical and chemical processes.

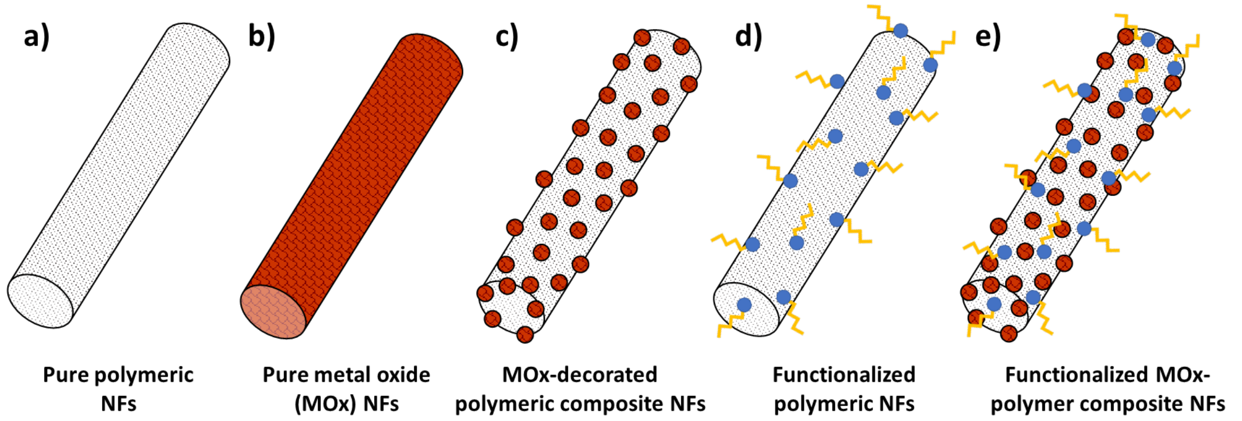


Figure 3. The evolution of electrospun nanofiber (ENF) materials from pure materials of either polymers or metal oxides (MOx) to integrated composites of these materials.

Initially, most applications of electrospun nanofibers for water treatment concentrated on the use of polymeric nanofibers, usually from precursors such as polyvinyl pyrrolidone (PVP), polyacrylic acid (PAA), and polyvinylidene fluoride (PVDF), for membrane filtration applications (Figure 3a).[24,31,32] Such work focused on identifying electrospun alternatives to polymers fabricated using more traditional routes for applications in low and high-pressure membrane filtration [e.g., microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO)].[33] These applications exploit the three-dimensional nanofiber network for physical separation processes to remove unwanted constituents from source water. At present, electrospun polymeric nanofibers represent viable membrane alternatives;[34,35] for example, nanofibers with high hydrophobicity, porosity, and an interconnected open pore structure have been shown to provide higher permeability compared to traditional polymeric membranes (i.e., analogous materials fabricated via phase-inversion approaches).[21,35–37]

Electrospinning can also be used for the production of pure metal oxide nanofibers (Figure 3b) and composites of polymeric nanofibers (Figure 3c) with embedded metal or metal oxide nanoparticles, thereby enabling chemical treatment applications including chemical oxidation (via photocatalysis), disinfection, sorption and ion exchange.[38–40] Pure inorganic nanofibers offer potential benefits analogous to ceramic membranes, including higher stability in the presence of strong acids and bases or active oxidants (e.g., ozone, hydroxyl radical, peroxyl radical) during advanced oxidation processes.[41,42] Popular examples include pure titanium dioxide and iron oxide (e.g., hematite) fibers,[43–45] which can be fabricated by transforming polymer fibers containing metal oxide precursors into corresponding oxides via high-temperature annealing in a controlled atmosphere. Similarly, carbon fibers can be produced from a polymer like polyacrylonitrile after thermal treatment in the absence of oxygen.[46,47] Extensive work with inorganic nanofibers has demonstrated that the same physical/(photo)electrochemical property variables desirable in discrete nanoparticles are beneficial in pure inorganic nanofiber performance;[48,49] for example, specific surface area can be manipulated by decreasing fiber diameter, thereby increasing the capacity of electrospun sorbents.[50]

Because pure inorganic and carbon nanofibers often lack the strength and durability of polymers, which may limit their practical viability, electrospun composites represent a possible compromise in reactivity and durability.[51–54] For these materials, (nano)particles of metals, metal oxides, and/or nanostructured carbon are integrated into polymer nanofibers to increase the membrane's functionality. Such composites are easily produced using electrospinning by simply dispersing nanoparticles into the polymer sol-gel precursor solution. Although the resulting nanoparticle-polymer composites overcome some of the material strength deficiencies encountered with pure inorganic fibers, they can suffer from more limited chemical reactivity as some reactive surface area on the integrated nanoparticles will be lost when embedded within the polymer.[55] Accordingly, there is much ongoing interest in exploiting the fabrication of these materials (e.g., increasing polymer porosity)[56–58] to increase the access of the embedded particles to solution during application and expose more reactive surface area.

Some studies have investigated surface modifications of polymeric nanofibers by means of grafting, blending, or coating to form functional groups (i.e., amine, thiol, carboxyl, carbonyl, etc.) on the fiber surface for specific applications and processes (Figure 3d). Through the addition of organic compounds, metal salts, surfactants, or co-polymers, these functionalized polymers can exhibit increased flux and higher surface area, leading to improved treatment performance compared to unmodified polymeric nanofibers.[23,59,60] Depending on the nature of the surface groups, they can also impart functionality including pollutant uptake through chelation or surface complexation processes.[61,62] Through imparting such surface groups, nanofibers are able to not just rely on physical separation processes common for more traditional membranes but also removal, ideally highly specific or targeted, for dissolved solutes.

A final modification of nanofibers combines the chemical functionality imparted by surface groups and nanofiber-nanoparticle composites (Figure 3e). Made either by post-processing to introduce surface functional groups to composites or through a combination of appropriate building blocks in the sol-gel precursor solution, these materials are promising for their potential multi-functionality.[25,63] Through simple additive performance from each component or possible synergies through interactions between the surface group and embedded particles, these materials help to increase the functionality of chemically reactive nanofibers without changing the physical footprint of the non-woven fiber membrane.

3. Applications of electrospun nanofibers relevant for POU/POE treatment

In the United States, NSF International and ANSI categorize POU/POE devices based on their treatment focus, which dictates device certification. These include devices specifically intended to improve the aesthetic quality of water (i.e., taste and odor), water softening devices (i.e., removal of hardness causing ions Ca^{2+} and Mg^{2+}), and various technologies to removal contaminants that pose risk to human health including organic and inorganic chemicals, emerging pollutants, and microbial pathogens. Here, we will highlight some of the most recent and promising research, development, and applications of electrospun technologies to meet these drinking water consumer needs. While we acknowledge the large group of recent reviews on electrospun fibers and their physical/chemical properties,[23–27] these have been broad in their scope of applications and not always highlighted the materials most promising for us in decentralized treatment based on material design and testing considerations most relevant to these applications

In Table S1, we have compiled an extensive list of published works looking at applications of electrospun nanofibers we have found most relevant to POU/POE applications. For each of the studies included, Table S1 summarizes key characteristics of the materials that were fabricated (elemental composition and nanofiber diameter), the experimental conditions used to test the performance of these materials (including the aquatic matrix and target pollutants), as well as the relative strengths of these works and the opportunities that exist to build upon the published findings to further advance POU/POE technology development with electrospun materials. As evaluation criteria for the relative strengths and opportunities identified in Table S1, we have selected the following criteria, which we contend are required elements for nanofiber research necessary for POU/POE technology development. They are:

- *Comparison to commercially available analog materials (CA):* An important performance benchmark for new materials is how their performance compares to analogous, commercially proven materials used in POU/POE treatment (e.g., activated carbon, commercial ion exchange resins, under-counter reverse osmosis membranes) or those materials that represent the industry standard for established full-scale treatment (e.g., Aeroxide P25 for photocatalytic processes). Such comparisons would ideally be integrated into all development and testing for materials intended for POU/POE applications.
- *Relevant pollutant targets for commercial POU/POE applications (RT):* We define relevant targets as contaminants of emerging concern such as unregulated organic contaminants (or models thereof), taste and odor causing compounds, regulated inorganic oxyanions (e.g., nitrate and arsenate), pathogens, and corrosion

derived metals (e.g., lead, copper), focusing on targets for which certified POU/POE devices exist.

- *Use of complex, drinking water relevant water quality in performance trials (WQ):* This includes considering the influence of aquatic matrix such as different pH values, temperature, and the influence of common co-solutes found in source and finished drinking water (e.g., co-existing CO^{2-}, NOM and other co-occurring contaminants).
- *Evaluation of material properties including strength, durability, and longevity (MS):* A critical aspect of materials used in POU/POE applications is their longevity and stability during application. Most certification processes are associated with expectations for performance lifetime. Moreover, for nanoengineered materials, there is need to demonstrate the integrity of the material during operation to ensure no release or leaching of unwanted materials into the treated supply. Materials must also be sufficiently flexible and strong to be engineered into appropriate reactor platforms for POU/POE use.
- *Performance testing that includes simulated POU/POE treatment (ST):* Finally, materials intended for use in POU/POE systems must be tested under conditions simulating such application platforms, which includes trials that extend beyond simple batch sorption studies (for equilibrium capacity) and also consider performance under more dynamic, likely kinetically limited, flow-through conditions. Of equal importance is demonstrating that the material can be translated from laboratory benchtop scale studies to technology demonstrations and prototyping at scales more representative of POU/POE applications.

We note that in our analysis of the appropriate literature summarized in Table S1, the extent to which certain of these factors were considered was somewhat variable. For example, a study may only have tested a nanofiber sorbent across different pH values, but not examined performance over more complex aquatic chemistries. Nevertheless, we categorized this study as having a strength related to WQ because it was at least considering how some aquatic variables could influence performance.

Thus, the intent of this contribution, which we believe distinguishes this work from prior reviews in this area, is to emphasize recent advances and emerging opportunities for the fabrication of chemically-active electrospun membranes specifically for decentralized water treatment. We will focus on work relevant to electrospun membrane applications in POU/POE technologies, which often are used as a final polishing step for a relatively clean source water matrix. This is an ideal application point for such nanofiber membranes so as to avoid their rapid fouling or clogging when applied to lower-quality source water with elevated suspended solids. For example, one can envision an application platform in which electrospun membranes can be integrated into a standard POU cylindrical filter housing, perhaps even supported by more traditional and widely used block carbon filters, to enhance device performance (Figure 4). It may even be possible to layer functionalized fibers or fiber composite, each designed to target specific constituents of concern in drinking water so that the overall treatment device exhibits multi-functionality. As illustrated in Figure 4, nanofiber membrane layers would be assembled in the same arrangement of the current footprint used in most POU/POE filtration devices, making integration into existing platforms highly plausible. It is with such application end point in mind that we critically review the available literature on the development of electrospun membranes for treatment of drinking water pollutants commonly targeted with current commercially available POU/POE devices.

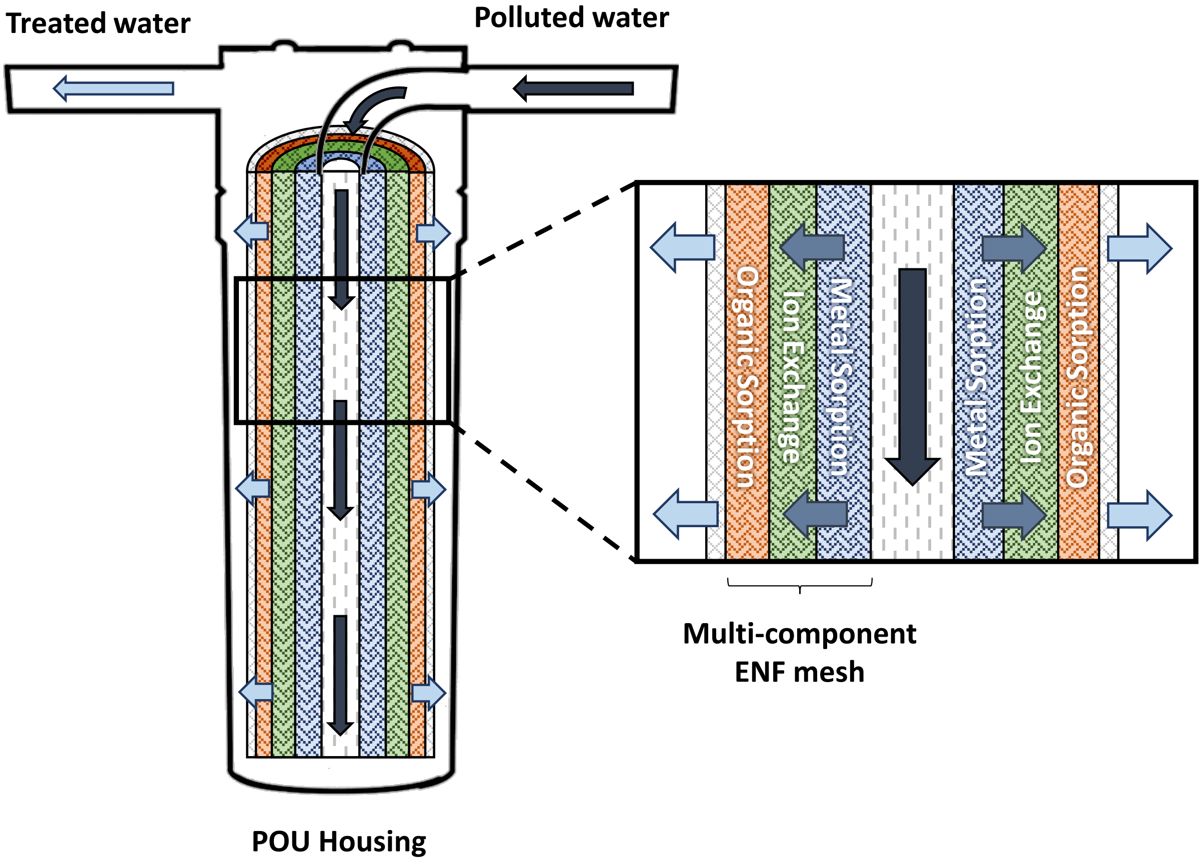


Figure 4. Schematic of a multi-layered, electrospun nanofiber mesh-integrated POU device, enabling sequential treatment processes in a single unit with small technology footprint.

3.1. Traditional membrane application including RO. Traditional membrane systems have been extensively studied for bacterial/viral removal, organic compound removal, and desalination, depending on the size requirement (i.e., microfiltration, nanofiltration, reverse osmosis). Since the earliest years of electrospinning, initial environmental applications revolved around polymeric nanofibers for filtration treatment.[37,63] Polymeric nanofibers membranes, often composed of polyacrylonitrile (PAN), polyvinylchloride (PVC), or polypropylene (PP) as a few examples, have been reported to possess good flexibility, high flux, and low transmembrane pressure,[61] all of which are desirable attributes for such performance applications. Additionally, electrospun membranes have shown great promise due to their higher interconnected porosity, larger effective surface area, and greater mechanical stability and hydrophobicity, markedly improving flux performance compared to traditional filtration membranes.[25]

Fouling is still a significant challenge facing the field of membrane filtration. To combat membrane fouling, many studies have demonstrated surface modification of electrospun nanofiber membranes via grafting, surface coating, and interfacial polymerization, leading to increased hydrophobicity and better flux throughput.[61] Others report the decoration of electrospun membranes with silver nanoparticles, a well-known antimicrobial agent, to alleviate biofouling.[64] Additionally, electrospun membranes require additional support for better mechanical strength and are usually manufactured or utilized in a hybrid system with a substrate.

3.2. Sorption of organic compounds. In designing a proper sorbent, broad spectrum activity, high surface area, and rapid rate of pollutant uptake are most desirable.[65] Specifically for sorption of organic pollutants, hydrophobicity is a common driver of pollutant uptake in traditional sorbents such as granular or powdered activated carbon (GAC and PAC, respectively).[66] More polar organic compounds tend to be susceptible to uptake via specific binding interactions. Accordingly, appropriate sorbents, including some forms of activated carbon, are designed to contain surface functional groups that may contribute to hydrogen-bonding and other types of chemical binding interactions for polar organic pollutant uptake. For POU water treatment, carbon block filters are popular in brands including the Amway eSpring™ water treatment system.[67] These carbon block filters are typically produced from carbon derived from charcoal, wood, or coconut and manufactured via either extrusion or molding.[68]

An appropriate electrospun analog to activated carbon is carbon nanofibers (CNFs) typically 200-500 nm in diameter due to their tunable chemical and material properties, high internal porosity, and large specific surface area.[47,49,69] The promise of CNFs is the ability to combine the application platform of a flexible membrane mesh with the high-capacity performance toward aqueous organic contaminants[70,71] of GAC and PAC. For example, Li et al. reported that the adsorption capacity of electrospun CNFs was significantly greater than PAC against a select group of emerging organic pollutants (i.e., ciprofloxacin, bisphenol, and 2-chlorophenol), attributing the enhanced performance to the greater surface area and pore size distribution of the fibers (**Figure 5**).[70]

Functionalized CNFs and CNF composites have often shown improved sorption performance relative to unmodified CNFs.[72,73] For example, integration of nanoparticles (e.g., CNTs, graphene oxides, metal oxides) and/or surface oxidation (via HNO_3, H_2SO_4) of CNFs can improve sorption performance, especially towards more polar organic compounds.[72,74,75] In one instance, Behnam et al.[72] reported on the fabrication of metal oxide-decorated CNFs (via nanoparticle integration into the precursor sol-gel) for the removal of Diazinon, a liquid-phase organophosphorus compound. Adsorption studies revealed a 4-fold improvement in the uptake rate due to the presence of embedded MgO and Al_2O_3 nanoparticles compared to the bare activated carbon nanofibers, which the authors attribute to the presence of surface hydroxyl groups on the integrated metal oxide nanoparticles (**Figure 6**).[72]

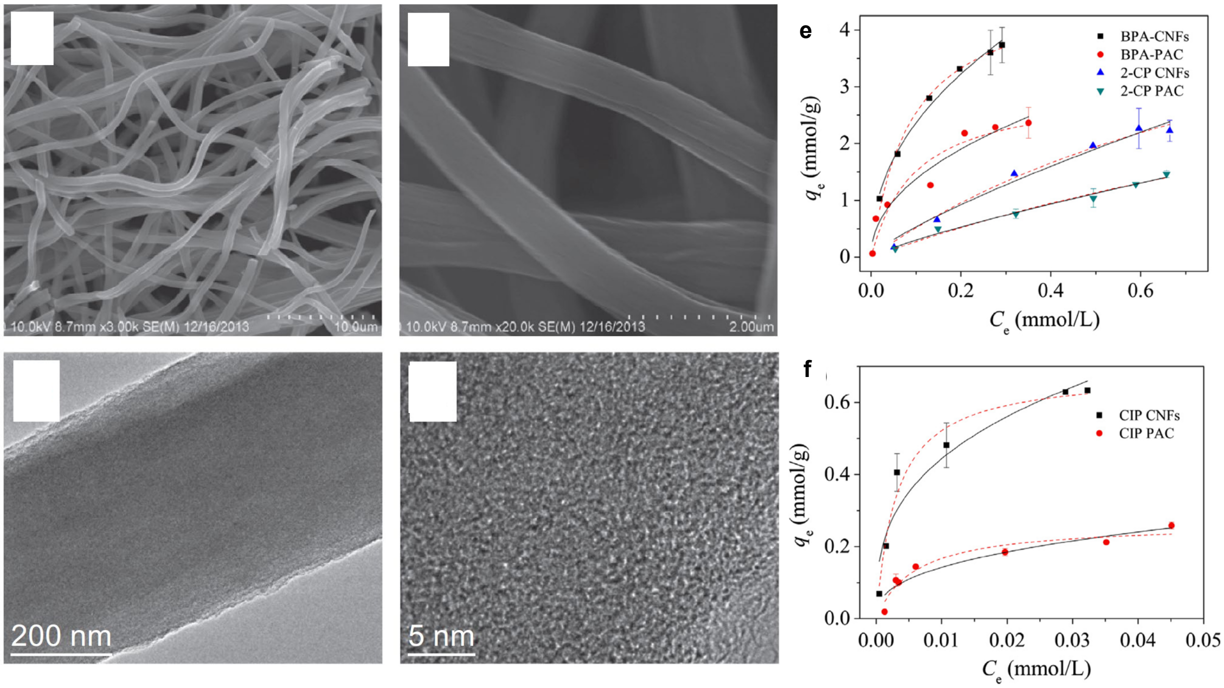


Figure 5. SEM (a, b) and TEM (c, d) images of carbon nanofibers (CNF) used for sorption of emerging organic pollutants. Also shown are sorption isotherms of the CNFs and powdered activated carbon (PAC) for e) bisphenol (BPA), 2-chlorophenol (2-CP), and f) ciprofloxacin (CIP) fitted with Langmuir model (red dotted line) and Freundlich model (solid black line), respectively. In all cases shown, CNFs resulted in greater organic pollutant uptake than PAC. Reproduced from Li et al.[70] with permission from Elsevier.

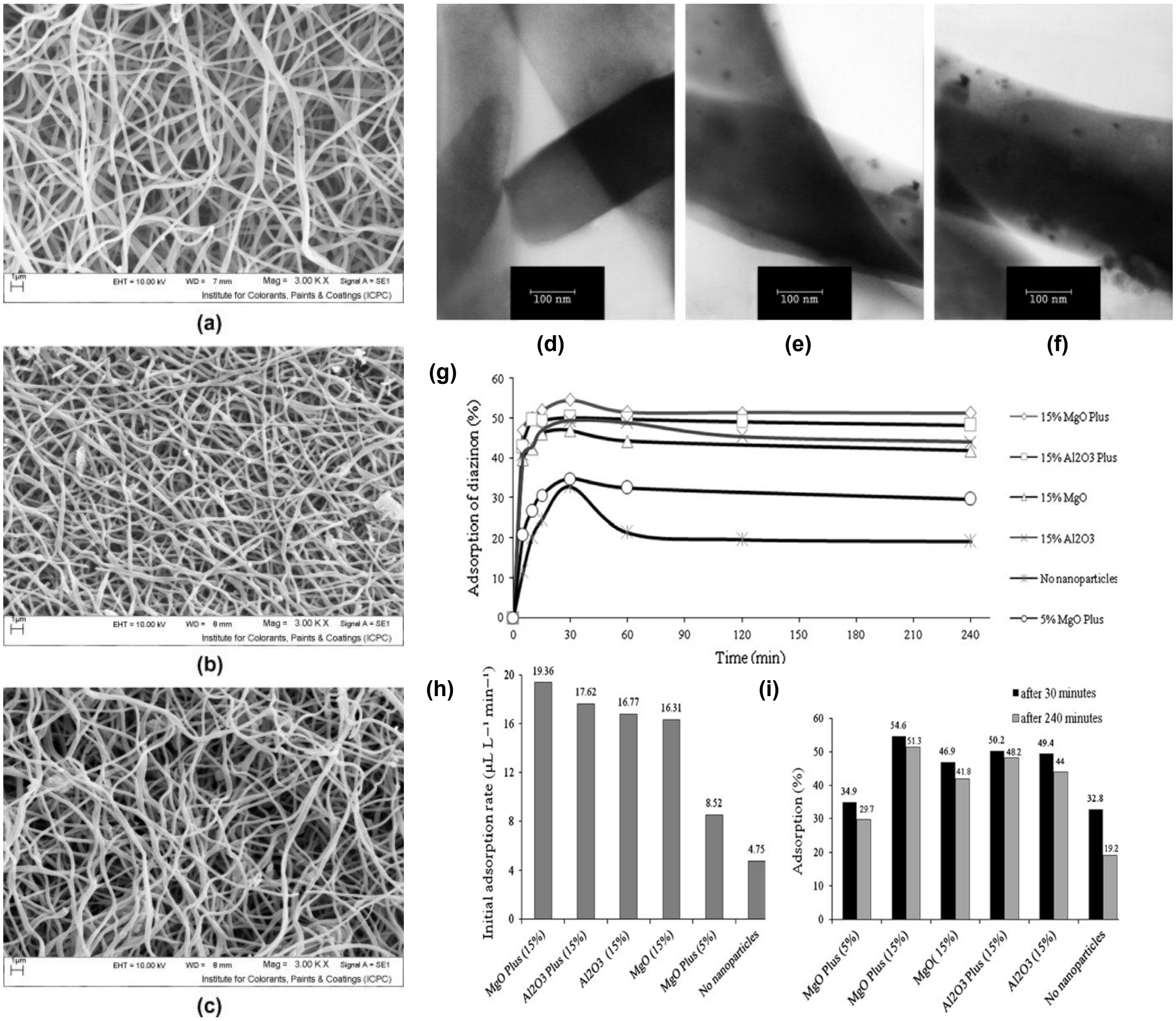


Figure 6. SEM and TEM images of ACNFs embedded with: (a, d) no metal oxide nanoparticles, (b, e) 10 wt.% MgO Plus, and (c, f) 10 wt.% Al_2O_3 Plus nanoparticles. Also provided (g) destructive adsorption of Diazinon, (h) initial adsorption rate of Diazinon after 5 min, and (i) the amount of adsorbed Diazinon after 30 and 240 min by ACNFs containing MgO, Al_2O_3, MgO plus, Al_2O_3, and no nanoparticles, with corresponding values indicated. We note that Plus grade nanoparticles were noted by the authors to have smaller crystal size and greater specific surface area compared to standard grade materials. Reproduced from Behnam et al.[72] and Dadvar et al.[74] with permission from Springer and Elsevier, respectively.

There remain practical challenges with the mechanical strength of CNFs, which may ultimately hinder their use in flow-through filtration systems.[76,77] However, recent advances in synthesis have led to more flexible nanofibers that can be more easily built into treatment systems. For example, we and others have found that the introduction of porosity (from sublimation of volatile phthalic acids) in CNT-CNF composite membranes increased membrane flexibility and durability during simulated testing in flow-through treatment systems.[75] We found that the porosity, in combination with embedded CNTs, produced a composite CNF with faster uptake rates and comparable surface-area normalized capacity to a commercial GAC for both atrazine, a widely used herbicide, and sulfamethoxazole, an unregulated emerging organic pollutant. Notably, porous CNFs without embedded CNTs exhibited limited uptake capacity toward these two compounds, suggesting that the embedded CNTs, accessible to solution through the induced porosity, were the primary sorbent driving pollutant uptake with these composites. Thus, these composites may represent a promising way to take advantage of CNTs in water treatment in a composite platform that limits the potential for their release into the finished water supply.

We note that polymeric nanofibers[34,78,79] and their composites[80] can also exhibit good sorption capabilities, but they typically fall short in terms of microporosity, and ultimately sorption capacity, when compared to their carbonized analogs.[69] In fact, the relative success of polymeric electrospun materials as sorbents will largely depend on the nature of the organic pollutant being targeted and the type of polymer or polymer composite being used for pollutant uptake. In our own work with polymer composites, we found that organic pollutant uptake did vary in response to the type of polymer (hydrophobic versus hydrophilic), as well as the nature of the chemical, in some instances.[80] Although potentially promising for hydrophobic compounds ($\log K_{\mathrm{ow}} > 4$), we would generally classify uptake as too slow (i.e., equilibrium on the order of several hours to one day) for moderately hydrophobic to polar organic compounds. This may limit the use of polymer sorbents in in-home treatment systems, which can be limited by residence time (e.g., POU filtration systems). As such, polymer-based sorbent materials are probably best suited for treatment systems with sufficient residence time to allow operation at or near equilibrium uptake (i.e., use in batch or semi-batch reactor systems).

3.3. Sorption of metals. Many commercially certified POU and POE treatment devices target metals including lead, copper, cadmium, chromium (hexavalent and trivalent), arsenic, and mercury.[81] Several of these metals such as lead, copper, and cadmium are those originating from corrosion of plumbing within centralized water distribution systems, premise plumbing on the consumer's property, or other forms of more decentralized drinking water infrastructure (e.g., private well components).[82–86] Because such corrosion-associated metals enter into the water supply solely through water delivery to the tap, POU treatment devices are relied on by many consumers to minimize exposure to these metals. POU devices are also frequently relied upon during public health crises associated with lead in drinking water exposure (e.g., Flint, Michigan).[84]

For many metals, particularly lead, activated carbon filters have often been the POU treatment of choice.[87] For example, both Pur (PPT111)[88] and Brita (e.g., Longlast+)[89] offer pitcher filters that are certified for removal of copper, cadmium, and mercury that rely on carbon filters, occasionally in combination with an ion exchange resin. Block carbon filters certified for lead removal under NSF/ANSI 53[90] are also common in many common bottle-filling devices marketed by companies including Elkay[91] and other vendors. Use of activated carbon for metals relies on the high surface area and large number of electron-rich sites on the carbon surface to produce a high-capacity sorbent for positively charged metal ions.[66] Beyond activated carbon, POU or whole-home reverse osmosis units are also effective for metal removal (e.g., Aquasana OptimH2O™ AQ-RO-3, among others[92]). In fact, membrane systems, while more expensive, may be preferred for lead removal, particularly given recent evidence of variable performance of traditional block carbon filters and granular carbon filters toward (nano)particulate lead species.[93]

Electrospun fibers can advance technologies for metal treatment through their ability to capture colloidal particulates via entrapment in pore spaces and remove dissolved metal species via sorption processes driven by electrostatic or more specific binding interactions. Electrospun membranes may also enable certain proven approaches for metal uptake to be more easily deployed at the POU scale; for example, granular ferric hydroxide (GFH), which is marketed as media for arsenic removal, is likely limited to whole home and larger treatment applications due to its grain size.[94–96] To the best of our knowledge, there is no iron oxide-based POU treatment analog for GFH, despite the well-recognized ability of iron oxides as sorbents for many dissolved metals.[97]

Several studies have demonstrated the immense potential for pure inorganic or ceramic electrospun fibers for metal uptake. These can be fabricated by electrospinning polymer sol-gels containing precursors that are then annealed into pure inorganic materials, typically metal oxide and carbon nanofibers, with relatively high surface area and porosity.[98–102] Examples of electrospun fibers used for metal uptake include oxides of iron (Fe_2O_3),[103,104] aluminum (Al_2O_3),[105] silica (SiO_2),[106] and cerium (CeO_2).[107] For instance, Ma et al. reported on the synthesis of electrospun silica fibers composed of a nonporous core and a mesoporous shell, exhibiting excellent adsorption of heavy metal cations (i.e., Pb^{2+}, Cd^{2+}).[106] Furthermore, post-process grafting of thiol functional groups on the silica fibers led to enhanced adsorption performance (**Figure 7**). Additional performance optimization has also been demonstrated through multi-component metal oxide nanofibers, most notably Al_2O_3-Fe_2O_3, which some have shown to exhibit greater adsorption capacity toward hexavalent chromium compared to either pure counterpart (**Figure 8**).[108–111] More generally, work to date with pure inorganic fibers has demonstrated that links between fiber performance and their physical properties follow expectations that are now well established for nanoparticle analogs (e.g., smaller particle size results in high specific surface area and, thus, more uptake per unit mass).[104]

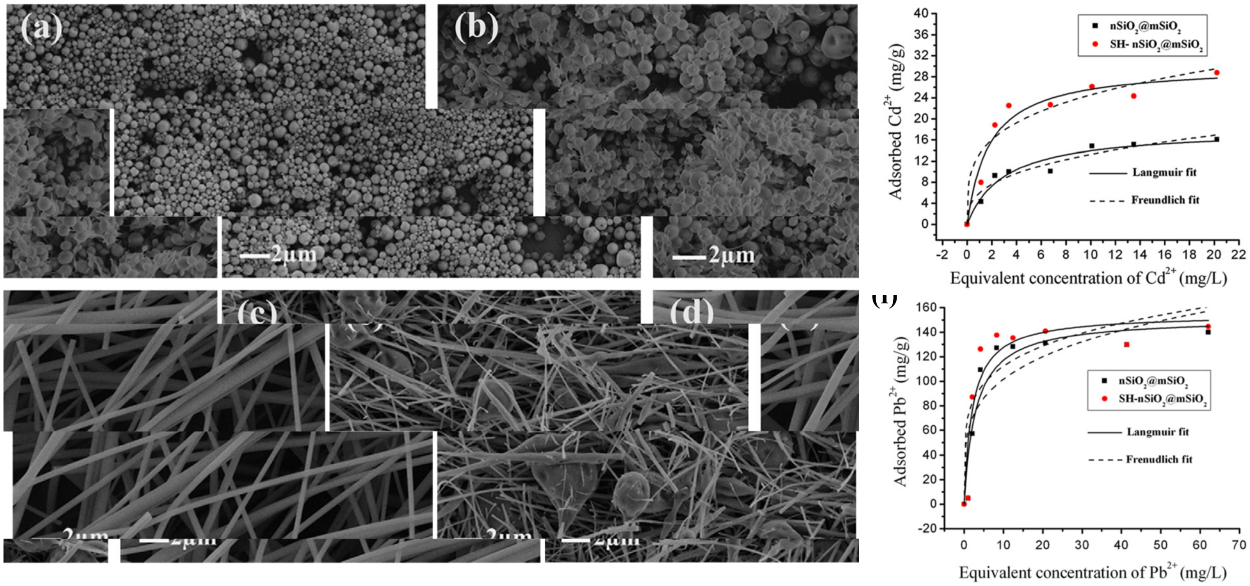


Figure 7. SEM images of electrospun silica nanofibers prepared using different hydrolyzing times: (a) 150 min, (b) 170 min, (c) 190 min, and (d) 210 min. Sorption isotherms of silica nanofibers ($nSiO_2@mSiO_2$) and thiol-functionalized silica nanofibers ($SH\text{-}nSiO_2@mSiO_2$) with corresponding fits (as indicated) for (e) cadmium (Cd^{2+}) and (f) lead (Pb^{2+}) uptake. Reproduced from Ma et al.[106] with permission from RSC.

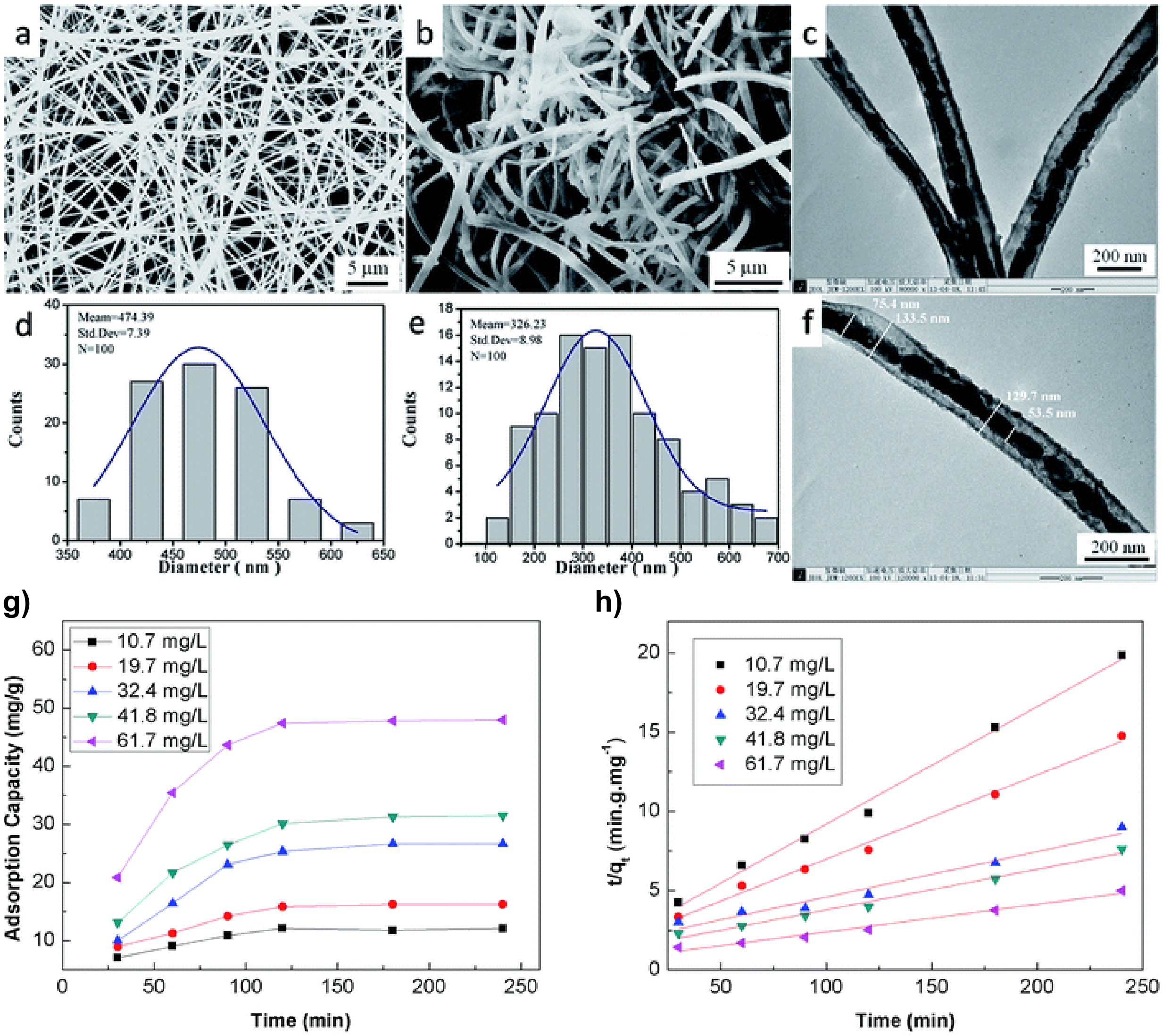


Figure 8. SEM images of (a) ammonium ferric citrate/polyvinyl alcohol (PVA) composite nanofibers and (b) α-Fe_2O_3–γ-Al_2O_3 core–shell nanofibers used for metal uptake. (c and f) TEM images of α-Fe_2O_3–γ-Al_2O_3 core–shell nanofibers. Also provided are the distribution of fiber diameters from (d) the ammonium ferric citrate/PVA composite nanofibers (470 ± 7 nm), and (e) α-Fe_2O_3–γ-Al_2O_3 core–shell nanofibers (330 ± 9 nm). For uptake of hexavalent chromium (Cr(VI)) on α-Fe_2O_3–γ-Al_2O_3 core–shell nanofibers, shown are (g) reported uptake curves over time for several different initial chromate concentrations and (h) the corresponding pseudo-second-order model-fit plots. Reproduced from Li et al.[110] with permission from RSC.

Although capable of achieving high removal of metals from water, there are considerable practical limitations to the use of pure inorganic fibers in POU and POE technologies. Like CNFs discussed earlier, pure inorganic fibers of metal oxides are typically very brittle and exhibit poor mechanical stability and flexibility, a result of morphological changes that occur while annealing (e.g., loss of mass due to the combustion of the polymer from the sol-gel).[13,112] Accordingly, without advances in fabrication to improve material strength, it will be difficult to envision technology applications for inorganic nanofiber materials.

A more promising approach for metal uptake may be polymer-metal oxide composites. Here, metal oxide nanoparticles are simply loaded into the sol-gel precursor solution prior to electrospinning. This approach allows high-capacity sorbent materials (e.g., metal oxide nanoparticles) to be deployed for POU and POE applications in a more flexible, polymer-based filtration platform. Because there has been extensive work with such polymer composites targeting a range of pollutants,[59,113–121] we have elected to focus on recent applications for uptake of common corrosion-derived metals (lead, copper, cadmium), as well as other metals of concern (As) that have existing POU/POE treatment options.

Perhaps the biggest challenge with such electrospun composites is preserving the sorbent activity of the metal oxide nanoparticles once embedded on or within the polymer nanofiber support, which will consume reactive surface area. To avoid this challenge, many, including our own research team, have used post-processing of nanofibers to surface enrich or decorate underlying polymers with high surface area metal oxide sorbents (**Figure 9**).[122] This produces surface-enriched or even core-shell composites that exhibit greater capacity (i.e., more sorbent accessible to solution) and faster rates of pollutant uptake (i.e., uptake can be slow on embedded particles due to necessary pore diffusion to access the sorbent surface) than traditional, homogeneously blended composites. We acknowledge, however, that additional post-processing to achieve such surface-enriched materials may be disadvantageous from the perspective of manufacturing and material production cost, thereby impacting the practical viability of such surface functionalized materials.

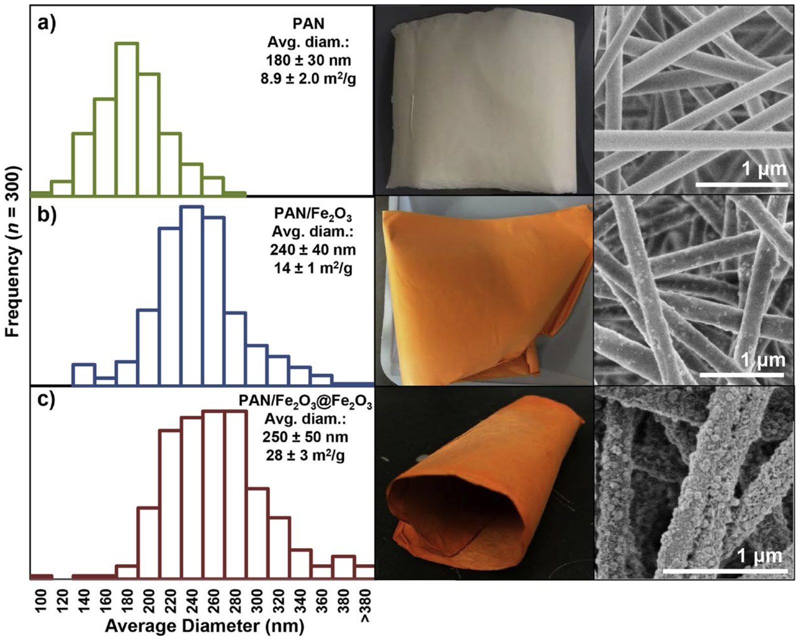


Figure 9. Size distribution histograms for (a) PAN, (b) PAN embedded with Fe_2O_3 that was prepared by blending the Fe_2O_3 nanoparticles into the precursor sol-gel (PAN/ Fe_2O_3), and (c) core-shell PAN/Fe_2O_3@Fe_2O_3 nanofibers that were prepared via the hydrothermal post-processing of PAN/ Fe_2O_3). In addition to nanofiber diameter distributions, which increase with composite and core shell materials, also shown are corresponding digital pictures and SEM images of the nanofiber mats. Reproduced from Greenstein et al.[122] with permission from Elsevier.

To avoid the issue of additional post-processing after electrospinning, we have discovered a unique approach to composite fabrication that exploits interactions between embedded nanoparticles and surface-segregating surfactants.[114,116] Surfactants are often included in precursor solutions to control surface tension.[123] We have found that certain surfactants with quaternary ammonium groups, functionalities often used in water treatment as coagulants and in ion exchange resins,[124,125] can also be used to impart chemical functionality to polymer nanofiber surfaces. We have fabricated polyacrylonitrile (PAN) nanofibers containing tetrabutylammonium bromide (TBAB), a common cationic quaternary ammonium salt (QAS), and also explored possible synergies arising from the inclusion of both QAS and iron oxide nanoparticles during electrospinning.[114] Our initial focus on QAS was motivated by prior observations that this class of surfactants is surface-segregating[126] TBAB accumulates at the polymer-air interface during synthesis, resulting in surface enrichment of positively charged, quaternary ammonium binding sites. Iron oxide nanoparticles (in this case, commercially available amorphous Fe_2O_3) were used because of their sorbent activity toward several species targeted with POU/POE devices, including Pb and As.[127,128] Notably, and as we recently detailed,[114] PAN nanofibers electrospun from solutions with QAS and iron oxide nanoparticles exhibit surface enrichment of not only cationic quaternary ammonium functionalities, but also the iron oxide nanoparticles; both species appear to co-locate to the polymer surface. This, in turn, produces a sorbent with higher capacity than expected based on the performances displayed by PAN modified with only TBAB or the iron oxide particles.

We have subsequently also observed and reported[116] on this surface-segregating behavior for anionic surfactants like sodium dodecyl sulfate (SDS) in combination with iron oxide nanoparticles. The combination of SDS and a commercial iron oxide nanoparticle during synthesis of electrospun PAN produces a composite enriched in iron oxide sites. Notably, we found that SDS is not retained in the polymer after synthesis, releasing into solution and leaving behind pores in the PAN (i.e., SDS acts as a porogen), which ultimately increases the capacity of the PAN/iron oxide composite. We contend that this unique, single-pot synthesis approach to produce nanofiber composites with surface-enriched binding sites may help streamline composite fabrication at the scale necessary for use in POU/POE devices.

3.4. Ion exchange for oxyanion removal. Ion exchange technologies use a functionalized stationary phase or resin. Typically, this resin consists of a cross-linked polymer matrix with covalently bonded functional groups that possess a fixed ionic charge able to attract dissolved ionic species.[129] Prior to application, these functional groups are initially saturated with a counter ion, which then is displaced or exchanged by the target ion when the pollutant is removed from solution.[130–132]

Ion exchange is commonly used in POU/POE technologies for the removal of common cations and anions. For example, many in-home water softeners rely on cation exchange resins, which are often certified to not only remove hardness-causing ions (Ca^{2+} and Mg^{2+}) but also regulated cations like barium and radium.[81] Similarly, Brita Standard Filters for pitchers and other water dispensers contain cation exchange resin for the purpose of removing contaminants like copper, zinc, and cadmium.[133] There are also a variety of more specialized treatment devices available commercially that rely on anion exchange for targets like nitrate, often in the form of POE devices for whole-home use.[134]

Because ion exchange most often relies on functionalized polymer resins, electrospun polymers with appropriate surface functional groups can be designed to function similarly in water treatment settings.[52] Design of such polymer fibers can be guided by chemistries successfully used in traditional materials. Specifically, there are four general types of conventional resins: strong-acid cation (SAC), weak acid cation (WAC), strong-base anion (SBA), and weak-base anion (WBA).[129] SBA resins, for example, have a quaternary amine group as the fixed charge.[129] For dilute aqueous phase concentrations, ion exchange resins tend to prefer the counterion of greater valence. For example, in the case of SBA, the selectivity preference follows: $PO_4^{3-} > SO_4^{2-} > Cl^-$. For monovalent anion exchange, the selectivity sequence is typically: $ClO_4^- > I^- > NO_3^- > Br^- > Cl^- > HCO_3^- > OH$.[129] Practically, this preference towards highly charged ions reduces removal efficiencies for some target pollutants (e.g., perchlorate preferred over nitrate).[7,129] Resins can also be produced so as to be selective to certain targets in order to overcome such interferences from ionic co-solutes. For example, commercially available nitrate-selective ion exchange resins rely on triethylamine or tributylamine functional groups,[135] and are popular because of their durability and selectivity of nitrate over other common anionic constituents in groundwater (e.g., sulfate, bicarbonate and chloride).

Motivated by the target application of POU/POE ion exchange, we[114] have designed polymer and polymer-composite nanofibers functionalized with cationic surfactants, such as cetrimonium bromide (CTAB) and tetrabutylammonium bromide (TBAB), for oxyanion removal from drinking water. These materials exploit the phenomenon of surface-segregation, in which surfactants like quaternary ammonium surfactants (QAS), preferentially migrate to the nanofiber surface during electrospinning to minimize the resulting free energy in the functionalized polymer materials (**Figure 10**). This produces a polymer mat consisting of nanofibers that are surface-enriched with positively charged, quaternary ammonium sites, enabling the removal of oxyanions (like chromate) from solution via ion exchange as confirmed by counterion release during oxyanion uptake. We have found that TBAB, which is a QAS with branched alkyl chains, is preferred for such applications over CTAB, which contains a linear alkyl chain, because it is better retained in the polymer (PAN) matrices during application for anion removal.

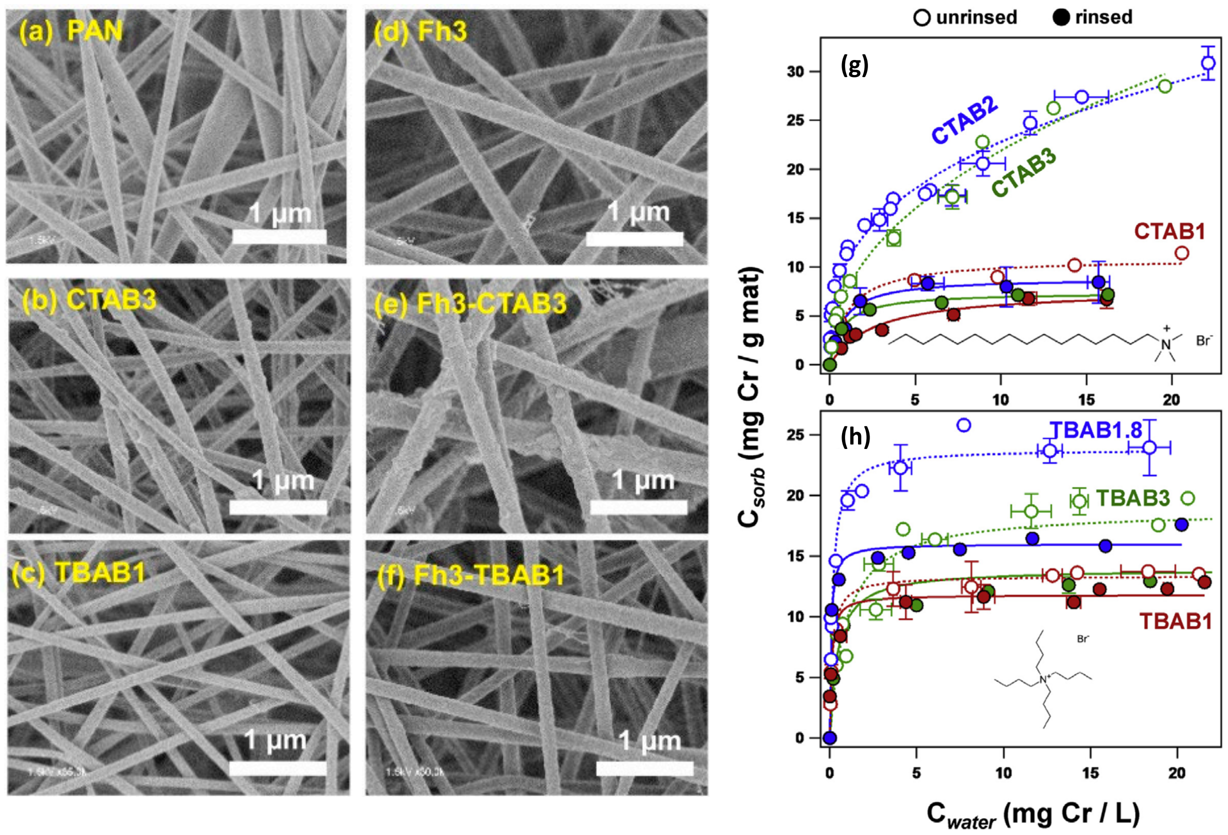


Figure 10. SEM images of (a) bare PAN fibers, (b) PAN fibers with CTAB, (c) PAN fibers with TBAB, (d) Fh-decorated PAN fibers, (e) Fh-decorated PAN fibers with CTAB, and (f) Fh-decorated PAN fibers with TBAB. Chromate sorption isotherms are shown as a function of (g) CTAB and (h) TBAB loading for materials that were used in uptake experiments as prepared (so-called "unrinsed" materials shown as open symbols with isotherm fits shown as dashed lines) and materials that were extensively washed with water prior to uptake experiments (so-called "rinsed" materials shown as solid symbols with isotherm fits solid lines). The decrease in performance observed between unrinsed and rinsed samples was attributed to the release of CTAB and TBAB (molecular structures shown in inset) from PAN during chromate uptake studies. Lines represent model fits to the Langmuir isotherm model for all composites except CTAB2 and CTAB3, for which data were best described by the Freundlich isotherm model. We note that in this work, composite mats were referred to as 'Fhx-CTABy' or 'Fhx-TBABz', where x, y, and z denote the ferrihydrite (Fh) nanoparticles, CTAB, and TBAB concentrations in the sol-gel, respectively, based on wt.% to the total sol-gel mass. Reproduced from Peter et al.[114] with permission from Elsevier.

This single-pot approach used in Peter et al.,[114] and in subsequent studies by our group,[136] represents a promising route to produce functionalized nanofibers suitable for ion exchange applications without any necessary post-processing of the polymer surface. There remain opportunities to expand this approach, including the use of alternative surfactants that may enable targeting of different pollutant types and represent more sustainable alternatives (e.g., QAS and concerns for their potential antimicrobial activity).[137] There is also a need to consider more closely the role of the polymer. Although surface-segregation has occurred readily in our work with polyacrylonitrile (PAN), there may be treatment applications where a hydrophobic membrane substrate (e.g., PVDF or polystyrene) is more beneficial, and exploiting surfactant-polymer interactions for such polymer materials requires additional investigation.

Ion exchange technologies, whether using conventional materials or electrospun polymers, are not without limitations. As with any technology relying on equilibrium partitioning, ion exchange systems require sufficient contact time for pollutant removal and exhibit limited capacity. They also must be regenerated, which often involves the use of concentration brine solution, and brine management is a well-documented challenge with such units.[138,139]

With more research and development, it is possible that electrospinning may enable new application platforms that overcome these traditional challenges of ion exchange while also increasing integration of ion exchange processes into existing POU/POE treatment systems. For example, block carbon filters are widely used in many in-home units (e.g., refrigerators). Amending block carbon with a functionalized polymer layer would allow ion exchange to be integrated into this existing POU architecture, thereby enabling removal of selected ions, like nitrate. However, this will require increases in the capacity and rate of ion uptake on such functionalized polymers to ensure sufficient removal during the contact time and lifetime afforded by such block carbon filters.

3.5. Organic chemical transformation. Recent advances in POU and POE technologies have led to the integration of ultraviolet (UV) lamps for disinfection.[140,141] Amway[142] and Aquasense,[143] for example, both offer in-home water treatment that relies on UV light to kill pathogens including bacteria and viruses. The emergence of UV light-emitting diodes (LEDs) has helped catalyze the growth of this approach in POU/POE devices.[144]

UV photolysis in POU systems could be exploited further to address the emergence of recalcitrant organic compounds by coupling them with photocatalytic materials to drive advanced oxidation processes (AOPs). AOPs are a group of treatment methods that utilize a combination of oxidative reagents to form highly reactive radical oxygen species (i.e., hydroxyl radicals) for organic pollutant degradation.[145,146] Some recent work has been conducted on the use of photocatalytic nanomaterials for POU applications, but specifically towards microbial disinfection rather than organic photo-oxidation.[147,148] Thus, there is much potential for the effective incorporation of UV-active photocatalytic membranes in POU/POE devices, particularly electrospun materials.

Titanium dioxide (TiO_2), considered the gold standard of photocatalysts, has garnered a great amount of attention in the environmental field for decades,[149,150] and thus, not surprisingly, has been heavily investigated as electrospun nanofibers,[151–153] as well as a slew of other nanomaterials.[154] These studies revealed the systematic control of physical properties (i.e., crystal phase, grain size, porosity, fiber diameter) of the nanofibers, which can be appropriately tuned towards optimizing photocatalytic activity. As a result, TiO_2 nanofibers with diameters that range from 30 to 300 nm have been investigated towards the degradation of pharmaceuticals such as phenol,[155] tetracycline,[156] carbamazepine,[157] and ranitidine,[158] as well as a host of dye contaminants.[159–161] For instance, Maeng et al. reported on the degradation of organic contaminants (i.e., cimetidine, carbamazepine, propranolol) by electrospun TiO_2 nanofibers and identified how the chemical moieties of the compounds interacted with variable aquatic conditions (i.e., DOM content, scavenger compounds, pH), ultimately affecting photocatalytic performance (**Figure 11**).[157] Moreover, TiO_2 nanofibers have been reported to outperform conventional nanoparticles based on photocatalytic activity, especially commercially available Aeroxide P25® photocatalyst.[155]

Zinc oxide (ZnO) nanofibers have also been investigated as potential photocatalysts for water treatment.[162–164] Although not as photocatalytically efficient as TiO_2, a benefit of ZnO is that it contains mid-band defect states (or quasi-stable energy states) and thus can be activated by renewable visible light, which comprises a major portion of the solar spectrum. Unfortunately, a major practical hurdle to the use of ZnO in treatment is its limited stability in water, making it highly prone to dissolution during application.

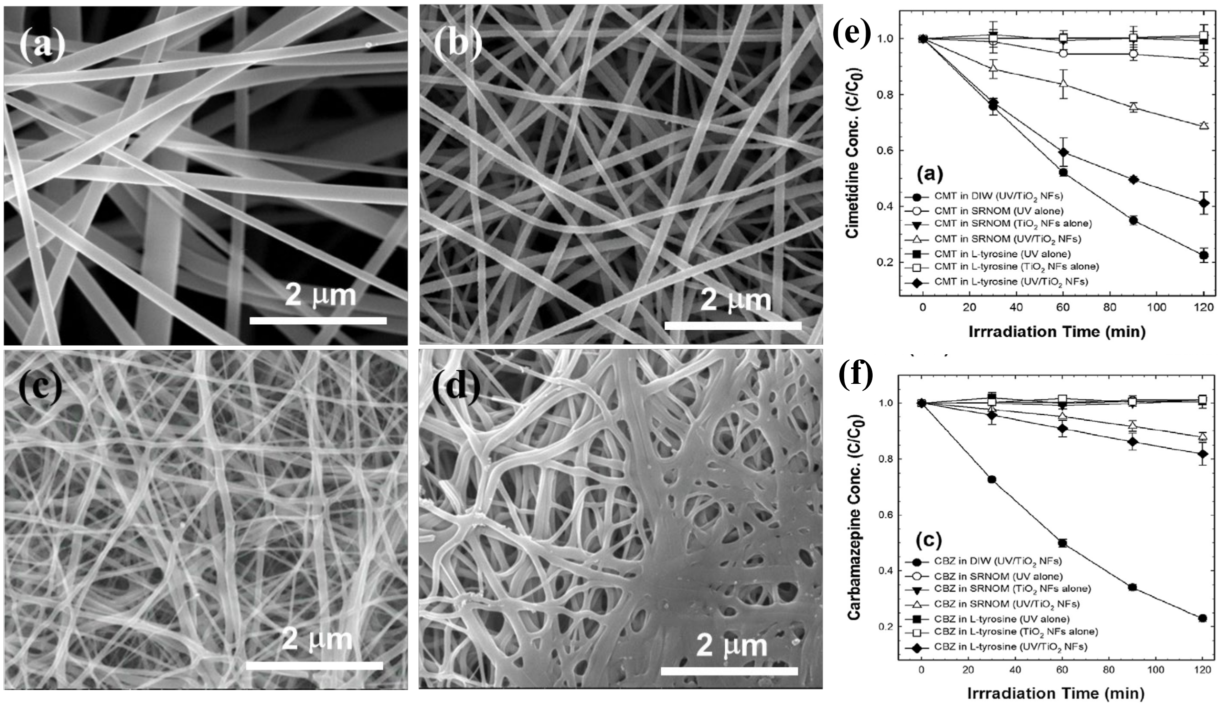


Figure 11. SEM images of (a) as-electrospun titanium isopropoxide-polyvinylpyrrolidone (TiP-PVP) composite nanofibers, TiO_2 NFs after annealing at 600 °C (b) without and with hot pressing at a pressure of (c) 15 MPa and (d) 20 MPa. Hot pressing was used to improve adhesion of the TiO_2 films to the quartz substrate for the purpose of increasing longevity. Reactivity studies show the effect of organic matter on the degradation of (e) cimetidine (CMT) and (f) carbamazepine (CBZ) for TiO_2 nanofibers under UV irradiation. Reproduced from Maeng et al.[157] with permission from Elsevier.

Furthermore, many strategies have been developed to enhance the photocatalytic activity of semiconductor-driven water treatment technologies due to the narrow window of the solar energy (i.e., UV light) necessary to trigger the photocatalytic activation of TiO_2. In fact, an advantage of electrospinning is that it makes integrating such strategies into semiconductor design simple via a single-pot sol-gel fabrication approach. The first method is by manipulating the composition of traditional UV-responsive semiconductors (e.g., TiO_2, ZnO) through doping and/or decorating with other materials to form hybrid nanofibers. Some examples of such hybrid nanofibers include TiO_2 with metals (e.g., Au, Ag, Cu)[165–172] and TiO_2 with other metal oxides (e.g., CuO, ZnO, SnO_2).[173–179] Results have shown that these decorated/composite materials outperform conventional pure TiO_2 nanofibers and nanoparticles, due to band gap engineering and/or surface plasmon resonance.[180–183] The second method is by altering semiconductor composition to enable photoactivation by visible light due to an intrinsically shorter band gap compared to that of traditional TiO_2 (i.e., < 3.2 eV).[184–188] Results show that towards pharmaceutical pollutants, these materials, which include bismuth vanadate ($BiVO_4$), bismuth tungstate (Bi_2WO_6), bismuth ferrite ($BiFeO_3$), tungsten trioxide (WO_3), and niobium oxide (Nb_2O_5), outperformed conventional titania nanoparticles and electrospun nanofibers when irradiated with visible light ($\lambda > 395$ nm).[189]

Perhaps the most mature approach for integrating electrospun photocatalytic materials into POU/POE devices is through the development and application of TiO_2-embedded composites.[190–193] For example, Lee et al. reported on TiO_2-embedded polymeric nanofibers composed of polyvinylidene fluoride (PVDF) or a blend of both PVDF and polyvinylpyrrolidone (PVP) for coupled sorption and photocatalytic destruction of organic contaminants (**Figure 12**).[193] Composite fibers were produced that were either nonporous (PVDF only) or porous

(PVDF/PVP blend), where pores to increase reactive surface area were generated via polymer blending and subsequent washing of PVP. Tested against a group of model and emerging organic contaminants (i.e., methylene blue, bisphenol A, 17α-ethynylestradiol), the porous composite fibers exhibited excellent sorption and photocatalytic degradation compared to both the bare PVDF fibers and the nonporous TiO_2-embedded composite fibers. The work of Lee et al. is notable for its demonstration of how electrospinning can be used to build multi-functional composite in a single, reactive filtration platform, assuming challenges in reactor design (e.g., integrating UV-light into POU/POE devices) can be overcome.

Similarly, composites of TiO_2 nanoparticles within carbon nanofibers have also been synthesized and subsequently tested against a suite of recalcitrant organic micropollutants in a UV-irradiated cross-flow filtration apparatus (**Figure 13**). Although the photoactive composite was only a few hundred microns thick, considerable micropollutant transformation (as much as 60-90% for inlet concentrations of 0.5 μM) was observed during a single pass through the filter at fluxes typically used in microfiltration applications. Tunable synthesis via electrospinning could be used to influence composite reactivity, with photocatalytic performance increasing with TiO_2 mass loading in the carbon nanofibers. Moreover, the performance of these composite materials was reasonably well maintained in complex matrices including co-solutes (e.g., carbonate) known to scavenge hydroxyl radical, the presumed oxidant responsible for micropollutant transformation. The mechanical flexibility exhibited by these carbon nanofiber composites coupled with the chemical activity of the TiO_2 photocatalysts could be another promising approach for integration into POU/POE devices.

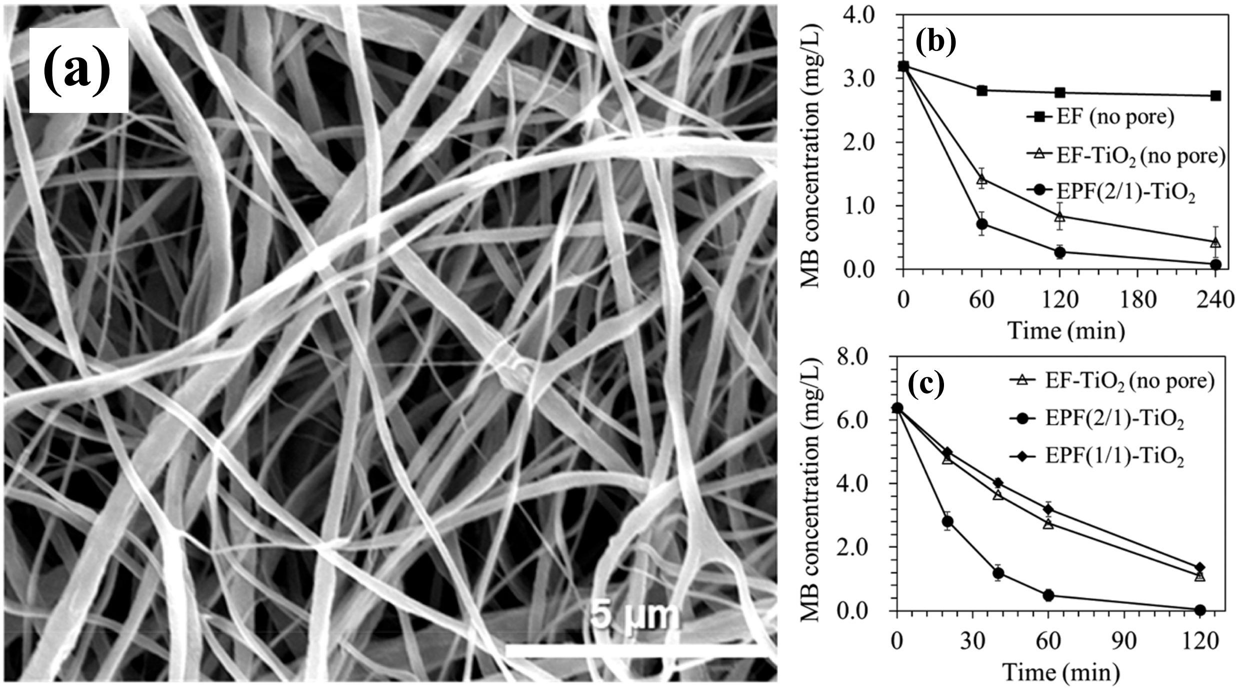


Figure 12. (a) SEM image of TiO_2-decorated polymeric nanofibers. (b) Adsorption of methylene blue (MB) under dark conditions using mats made with electrospun fiber (EF), electrospun nonporous fiber containing P25 TiO_2 (EF-TiO_2), or electrospun porous fiber containing TiO_2 (EPF(2/1)-TiO_2). (c) Removal of MB by concurrent adsorption and photocatalytic degradation under UVA irradiation using mats made with EF-TiO2 or electrospun porous fibers prepared with different polymer blending ratio (EPF(2/1)-TiO_2: PVDF(12%)/PVP(6%) and EPF(1/1)-TiO_2: PVDF(9%)/PVP(9%)). Reproduced from Lee et al.[193] with permission from ACS.

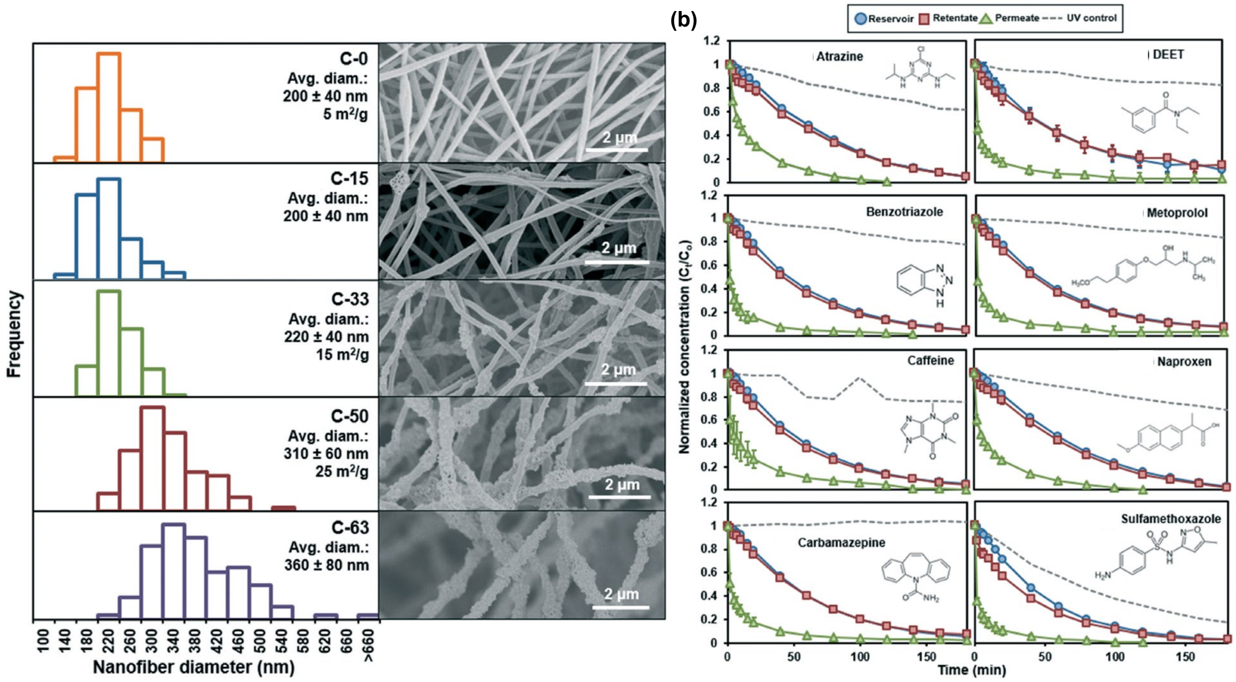


Figure 13. (a) Histograms and associated SEM images for carbon-TiO_2 nanofiber composites. Composites denoted as C-(wt%), where the TiO_2 weight percent is relative to PAN content in the initial sol-gel. All electrospun sol-gels also contained 2.5 wt% PTA, which was used as a porogen to increase composite flexibility. (b) Normalized concentration of model organic contaminants (name and structure in graph insets) over time in the reservoir, retentate, and permeate of the cross-flow, UV-light irradiated filtration apparatus. Reactivity data are shown for a C-50 nanofiber filter. UV controls, conducted without C/TiO_2 present, are shown by grey dashed lines. Reproduced from Greenstein et al.[192] with permission from RSC.

Overall, there are a large number of studies attempting to demonstrate how electrospun photocatalytic nanofibers may help to overcome practical challenges that have hindered the widespread application of photocatalytic nanoparticles for organic contaminant removal.[194] However, most of the work to date has not investigated nanofiber performance under conditions representative of water treatment and has also focused on targets and matrices that are not broadly relevant to POU/POE applications. For example, a good number have centered on the photodegradation of common dyes (e.g., methylene blue, methyl orange, rhodamine B) at relatively high concentrations (10-50 mg/L) while also lacking details on necessary water quality conditions (e.g., pH, buffer systems). Future development of photocatalytic nanofiber membranes, specifically polymer-based nanofiber composites, must work to conduct performance benchmarking and employ reactor systems that are most appropriate of simulated water treatment conditions and deployment at scales suitable for POU/POE treatment applications.

4. Opportunities for nanofibers to expand POU/POE technologies

With the heavy amount of research being conducted around electrospun fiber-enabled water treatment, there remain opportunities for such technologies to address emerging challenges in the decentralized water treatment space. Here, we highlight three areas where additional research and development could help electrospun materials address current and emerging water treatment needs of various consumers.

One area where there is both an existing need for treatment and a growing body of research with electrospun membranes is the treatment of uranium in drinking water. In many areas of the arid southwestern United States, naturally occurring uranium, as well as uranium contamination resulting from legacy nuclear development, has contaminated groundwater supplies. This problem is well documented among the Navajo Nation,[195,196] where uranium levels in groundwater supplies can often exceed 50 μg/L (the MCL set by the U.S. EPA is 30 μg/L).[197–199]

Electrospun materials have been popular choices for uranium removal through the ability to chemically process polyacrylonitrile (PAN) fibers to produce functional groups that are highly reactive and specific toward dissolved species of uranium. Most popular among this approach is the reaction of nitrile groups in PAN with a reducing agent (e.g., hydroxylamine) to yield amidoxime functional groups.[200–203] For instance, we have observed steady performance of amidoximated PAN fibers across matrices, including complicated solutions simulating drinking water sources (i.e., solutions with high hardness and ionic content) and even synthetic urine (for possible biomonitoring applications).[204] Other approaches have involved similar chemical functionalization of polymers to produce other types of uranium-binding sites including nitrogen (N) and phosphorus (P)-based binding agents.[201,205] There is a large body of literature on the use of such materials for the recovery of uranium from matrices like seawater, with growing interest in their application for water treatment.[206,207] There remain challenges: post-processing of fibers may increase production cost and time, which may make use of amidoxime or similar groups infeasible at the industrial scale needed for POU/POE device manufacturing. However, given the clear consumer need for reliable technologies for uranium treatment, development of high performance and cost effective electrospun fibers for uranium removal from drinking water should be viewed as a priority.

Another opportunity is expanding the use of antimicrobial fibers for applications in drinking water disinfection.[208] Even prior to the emergence of COVID-19 and the critical need it revealed for antimicrobial filtration materials,[209,210] there was a relatively deep body of research on antibacterial electrospun fibers, usually for use in bandages and antimicrobial surfaces.[211–215] These types of electrospun fibers are most typically produced through the integration of an antibacterial agent during synthesis.[216–218] Silver (Ag) has garnered much interest for use in antifouling and disinfection applications,[56] usually integrated into ceramic and polymeric substrates as nanoparticles to produce composite fibers. Given established antimicrobial activity of Ag particles, such composites are often touted as promising materials for point-of-use water treatment, especially for developing and underdeveloped countries.[219–227]

A persistent concern about such applications of biocidal-integrated materials is the release of the biocidal agent over time, potentially diminishing performance while also contaminating the finished water supply. Here, more work is needed, and a focus should be on exploring the fundamental interactions between biocides (e.g., Ag) and support matrices to optimize retention and minimize the release of these agents under simulated treatment applications at scale. For example, while more work is needed, some studies have shown that due to strong interaction between Ag nanoparticles and certain support materials (e.g., N-containing polyurethane), Ag loss during such filter applications can be minimized, resulting in Ag levels in the finished water that are below the current U.S. EPA and WHO limit for drinking water (0.1 ppm).[64,228,229] Beyond concerns over Ag release, the disinfection capacity of such Ag-impregnated filters will likely be exhausted over continuous operation because Ag ion resulting from nanoparticle dissolution is believed responsible for pathogen kill.[230,231] Further, as is observed for most reactive metal substrates, surface fouling by natural organic matter (NOM) or sulfide (which may be important during treatment of some groundwater sources) may compromise the efficacy of disinfection.[232] As such, practical studies evaluating performance and operational lifetime during simulated applications involving complicated matrices and conditions most representative of POU/POE devices are needed, perhaps even more so than additional, purely fundamental research studies with such biocidal materials.

Finally, another emerging opportunity relates to the use of electrospun fiber networks in electrochemical water treatment, especially with the extensive amount of work already conducted with electrospun carbon nanofibers (CNFs).[233–238] There is surging interest in the use of electrochemical approaches for water treatment because these approaches can be decentralized while being powered by renewal sources of electricity (e.g., solar).[239] Critical to recognizing the promise of electrochemical water treatment is the development of appropriate electrode materials for use in such applications. Here, electrospun CNFs may be useful due to their high surface area and tunable characteristics during synthesis. For example, higher carbonization temperatures can be used to increase their graphitic structure, and thus improve the conductivity of these materials.[240] Yet, higher temperatures can also result in more brittle materials due to extensive mass loss during carbonization, which may make the most conductive materials impractical for application with intentional efforts to increase their material strength.[241] Accordingly, as is the case with most other applications of electrospun CNFs, a priority for advancing their use in electrochemical water treatment applications will be finding the appropriate balance between high material strength and desirable attributes of electrode materials (e.g., low resistivity, high surface area).

5. Outlook and Recommendations

Our review of the literature yielded 59 studies, summarized in Table S1, with relevance to the application of electrospun nanofibers in POU/POE treatment. The results of our literature analysis are summarized in Table S2, where we aggregate these studies based on their identified strengths and the remaining research opportunities they present. From this analysis, we find that most of the work to date have focused on suitable targets for POU/POE applications (i.e., based on the contaminants that are frequently the focus of certification). Moreover, most studies have challenged their materials with various aquatic matrices to establish how performance varies with water quality. A number of investigations have also considered the strength and stability of the nanofibers to assess their integrity and potential for unwanted component leaching during application.

An important consideration is also benchmarking the performance of electrospun materials toward appropriate commercial analogs. We found that a considerable percentage of the studies considered herein did benchmark against a commercially relevant treatment, where the majority of these were instances of Aeroxide P25 being used as a basis of comparison for photocatalytic nanofiber performance. Studies that provided quantitative performance benchmarks (e.g., sorption capacities, rate constants) for nanofibers relative to commercial analogs tested under the same experimental conditions are summarized in Table 1. Generally, our analysis of the literature shows that electrospun nanofibers and nanofiber composites performed comparably to, and in some instances outperformed, their commercial analog counterparts. We note that we limited our comparison in Table 1 to those studies that tested commercial analogs toward the same contaminant target and using the same experimental conditions, otherwise differences in experimental conditions during performance testing may confound comparison to analogs.

Table 1. List of select nanofiber and nanofiber composite (NF) studies with performance comparisons with commercial analogs (CA).

Treatment	Materials[a]	Reactivity				Reference
Organic Sorption	NF: CNF CA: PAC	CNF	ciprofloxacin	q_m = 0.68 mmol/g		Li et al., 70
			bisphenol	q_m = 4.82 mmol/g		
			2-chlorophenol	q_m = 6.18 mmol/g		
		PAC	ciprofloxacin	q_m = 0.26 mmol/g		
			bisphenol	q_m = 2.98 mmol/g		
			2-chlorophenol	q_m = 5.37 mmol/g		
Metal Sorption	NF: PAN/Fe_2O_3 & PAN/Fe_2O_3@Fe_2O_3 CA: GFH powder	PAN/Fe_2O_3 NF	As^{5+}	q_m = 7.2 mg/g	q_{SA} = 0.10 mg/m^2	Greenstein et al., 122
			Cr^{6+}	q_m = 4.6 mg/g	q_{SA} = 0.06 mg/m^2	
			Pb^{2+}	q_m = 42 mg/g	q_{SA} = 0.59 mg/m^2	
			Cu^{2+}	q_m = 26 mg/g	q_{SA} = 0.37 mg/m^2	
		PAN/Fe_2O_3@Fe_2O_3	As^{5+}	q_m = 9.3 mg/g	q_{SA} = 0.18 mg/m^2	
			Cr^{6+}	q_m = 7.3 mg/g	q_{SA} = 0.14 mg/m^2	
			Pb^{2+}	q_m = 57 mg/g	q_{SA} = 1.1 mg/m^2	
			Cu^{2+}	q_m = 35 mg/g	q_{SA} = 0.67 mg/m^2	
		GFH powder	As^{5+}	q_m = 62 mg/g	q_{SA} = 0.13 mg/m^2	
			Cr^{6+}	q_m = 38 mg/g	q_{SA} = 0.08 mg/m^2	
			Pb^{2+}	q_m = 110 mg/g	q_{SA} = 0.25 mg/m^2	
			Cu^{2+}	q_m = 31 mg/g	q_{SA} = 0.07 mg/m^2	
Chemical Oxidation	NF: TiO_2 & Au-TiO_2 CA: Aeroxide P25	TiO_2 NF	phenol	k_{obs} = 0.07 min^{-1}		Nalbandian et al., 155
			atrazine	k_{obs} = 0.05 min^{-1}		
			carbamazepine	k_{obs} = 0.1 min^{-1}		
			DEET	k_{obs} = 0.06 min^{-1}		

			sulfamethoxazole	k_{obs} = 0.08 min^{-1}		
		Au-TiO$_2$ NF	phenol	k_{obs} = 0.65 min^{-1}		
			atrazine	k_{obs} = 0.35 min^{-1}		
			carbamazepine	k_{obs} = 0.8 min^{-1}		
			DEET	k_{obs} = 0.67 min^{-1}		
			sulfamethoxazole	k_{obs} = 0.4 min^{-1}		
		Aeroxide P25	phenol	k_{obs} = 0.15 min^{-1}		
			atrazine	k_{obs} = 0.1 min^{-1}		
			carbamazepine	k_{obs} = 0.25 min^{-1}		
			DEET	k_{obs} = 0.1 min^{-1}		
			sulfamethoxazole	k_{obs} = 0.15 min^{-1}		
	NF: TiO$_2$ CA: Aeroxide P25	TiO$_2$ NF	ranitidine	k_{obs} = 0.008 min^{-1}		Choi et al., 1578
		Aeroxide P25	ranitidine	k_{obs} = 0.011 min^{-1}		
	NF: TiO$_2$ & Ag-TiO$_2$ CA: Aeroxide P25	TiO$_2$ NF	phenol	k_{obs} = 0.097 min^{-1}		Nalbandian et al., 172
		Ag-TiO$_2$ NF	phenol	k_{obs} = 0.27 min^{-1}		
		Aeroxide P25	phenol	k_{obs} = 0.074 min^{-1}		
	NF: BiViO$_4$, Ag-BiViO$_4$ & Au-BiViO$_4$ CA: Aeroxide P25	BiViO$_4$ NF	phenol	k_{obs} = 0.0014 min^{-1}	$k_{obs,395\ nm}$ = 0.0011 min^{-1}	Nalbandian et al., 189
		Ag-BiViO$_4$ NF	phenol	k_{obs} = 0.0023 min^{-1}	$k_{obs,395\ nm}$ = 0.0016 min^{-1}	
		Au-BiViO$_4$ NF	phenol	k_{obs} = 0.0018 min^{-1}	$k_{obs,395\ nm}$ = 0.0022 min^{-1}	
		Aeroxide P25	phenol	k_{obs} = 0.074 min^{-1}	$k_{obs,395\ nm}$ = 0.0009 min^{-1}	
	NF: PVDF/TiO$_2$ CA: Aeroxide P25	PVDF/TiO$_2$ NF	bisphenol A	k_{obs} = 0.014 min^{-1}		Lee et al., 193
		Aeroxide P25	bisphenol A	k_{obs} = 0.003 min^{-1}		

Another important consideration is how material performance is measured relative to the unit mass or surface area of material (i.e., mass or surface-area normalized performance descriptors). For example, Greenstein et al.[122] found that their iron-oxide polymer nanofiber composites were less reactive than commercial granular ferric hydroxide (GFH) on a per unit mass basis but exhibited greater activity per unit surface area. Relative to GFH, their nanofibers exhibited greater external surface area that also produced faster uptake rates on the nanofibers.[122] We also note that some studies that did conduct performance benchmarking relative to a commercial analog are not included in Table S2 because a simple activity descriptor was not available to simply represent the performance comparison. For example, Peter et al.[75] conducted sorption isotherm and pH-edge experiments with their CNFs and with granular activated carbon (GAC), but the isotherms for both materials were non-linear and did not reveal a sorption capacity that could serve as a basis for performance comparison. As with Greenstein et al.,[122] these CNFs exhibited sorption of organic pollutants (atrazine and sulfamethoxazole) that was comparable to GAC on a surface-area basis, as well as faster uptake rates.

Our analysis of existing literature (Table S1) and summary analysis in Table S2 reveals some clear needs for future study around the use of nanofibers in POU/POE systems. There is a clear need to test the performance of nanofiber materials using reactors and conditions that better simulate POU/POE applications, including continuous flow-through or semi-batch systems. Many studies to date have utilized closed or no flow (batch) experimental systems, which tend to reflect performance under equilibrium-type conditions. In flow-through systems, which are most common for POU/POE treatment, kinetic limitations may constrain performance. Thus, more testing is needed to establish whether performance metrics established in batch systems are maintained under conditions more representative of treatment. Similarly, there are few studies that address the larger-scale fabrication of electrospun materials, prototyping of nanofiber-based POU/POE technologies, and longer-term performance trials of nanofiber-based treatment devices. This is a high priority need area for future development of POU/POE technologies leveraging these materials, and it will require research efforts that seek to identify how best to translate nanofiber fabrication from the laboratory bench to industrial scale (an issue discussed further below). Finally, more, if not all, studies should include appropriate analogs for benchmarking nanofiber performance. We acknowledge that in some instances, a clear analog for comparison may not be obvious (e.g., photocatalytic membrane systems[192]). Whenever possible, however, the value of electrospun nanofibers needs to be assessed relative to commercially available and widely trusted materials established in POU/POE treatment.

Beyond those identified from gaps in existing literature, there are additional advances that would also promote material and technology development. First, any nanofiber-enabled water treatment technology must be assembled from the right building blocks. When assessing the current state of the science and considering the balance in performance priorities of material reactivity, strength, and durability, the authors contend that the most promising route is the use of polymer-metal oxide composites. These materials hold the advantage of combining the beneficial performance properties that are already well-established for the selected building blocks for such composites. For example, electrospun polymer nanofibers are considered viable alternatives to more traditional polymer fabrication routes, while use of granular and powdered forms of carbon and certain metal oxides are widely used and accepted across water treatment. This should increase the likelihood that composite technologies using these building blocks gain certification and commercial acceptance for use in POU/POE devices because their non-electrospun analogs are already available through the commercial market. Nevertheless, there remain tremendous opportunities for fundamental discovery related to how the nature of the polymer (hydrophilic versus hydrophobic) influences composite performance. Moreover, more work is needed related to understanding and exploiting fundamental interactions between the immobilized or embedded material (e.g., metal oxide or carbon nanomaterial), base polymer, and other synthesis aids (e.g., surfactants), as these interactions have implications for material performance (e.g., controlling the available surface area of immobilized materials) and long-term stability (e.g., ensuring no release of the embedded materials).

To facilitate more prototype testing, another important area for future development relates to the reliable and reproducible industrial production of nanofibers that is necessary for their integration into POU/POE units.[25,35] While electrospinning is growing in commercial and industrial viability, there are still some fabrication obstacles facing electrospun membrane upscaling. This includes needle clogging, electric field interference, and, most importantly, property accuracy and repeatability, although some solutions to these problems are available (i.e., multi-needle spinnerets, needleless spinning, auxiliary electrodes).[20] Related to these issues, more work is needed to explore how synthesis conducted at laboratory scale, which often involves simpler (e.g., single-needle) configurations and lower production rates (e.g., sol-gel solutions electrified at a few tenths of a mL per min) translates to larger fabrication units (e.g., multi-needle rigs), emerging fabrication routes including needleless electrospinning, and higher production rates (where parameters like sol-gel viscosity will necessarily be different to accommodate larger-scale production). Moreover, for applications in water treatment, fabrication costs will need to be minimal to compete with market-accepted approaches like activated carbon. As such, identifying relatively simple fabrication routes to highly functional and high-performing materials will be paramount.

Finally, it is imperative to develop and grow our understanding of nanofiber and nanofiber composite stability under operation requirements of continuous flow systems (e.g., high mechanical strength, high permeability, low pressure drops). Indeed, to the best of our knowledge, there are no certified POU or POE technologies that leverage nanotechnology or nanoengineered materials. Thus, in developing nanofibers and nanofiber composites such as those described herein for treatment, special focus will be needed on generating datasets that demonstrate their performance and promise at scale, ideally under conditions that represent those used in certification testing. As a final consideration, there has been much focus on considering the sustainability of new water treatment technologies, particularly as it relates to new or innovative approaches and materials for application to persistent or emerging pollutants. Thus, while lifecycle analyses have been conducted on non-electrospun carbon nanofiber membranes and polymeric membranes to assess their environmental impact,[242,243] there persists a paucity of lifecycle data related to the fabrication and application of electrospun nanofibers for water treatment. We advocate for more analyses on the sustainability of electrospun nanofiber membranes, including associated lifecycle costs from raw materials and emissions associated with fabrication and device manufacturing, in complement to efforts to further develop and commercialize such technologies.

Overall, we find the outlook for electrospun technologies at the scale of point-of-use and point-of-entry treatment systems is positive. Although these are certain to be more costly materials than those that currently dominate the marketplace, their promise is in the versatility of their fabrication. This should allow new and different materials that are better suited to target certain pollutants and fill existing (e.g., iron-based POU analogs for As sorption) and emerging (e.g., microfiltration membranes for colloidal lead) needs in the certified technology market.

6. Conclusion

The relevant research literature shows that electrospun nanofiber materials can be effective alternatives for the removal of harmful contaminants in decentralized water treatment applications. Specifically, due to their high external surface area and application as filtration membranes, they are well-suited to advance nanotechnology-integrated POU/POE devices. In this review, we examined and scrutinized the current landscape of literature relevant to the application of electrospun nanofibers for POU/POE treatment, finding that:

- Use of electrospun fibers for traditional membrane separations is well-established, along with promising research illustrating nanofiber modifications to address issues such as fouling;
- Fibers used for organic sorption are primarily carbon-based due to their tunable properties including porosity and hydrophobicity, but opportunities remain to use

functionalization strategies to improve performance and target a wider range of organic contaminants;

- Fibers developed for metal sorption are primarily metal oxide-based, where functionalized polymer-metal oxide composites represent a particularly promising subset of materials for their ability to integrate high sorption capacity metal oxide nanoparticles;
- Fibers developed for us in ion exchange remain in their infancy, with relatively few examples in the published literature. There remain opportunities to integrate established functional groups used in commercial resins into polymeric nanofibers for such applications;
- There is a wealth of examples in the literature exploring photocatalytic fibers for use in chemical oxidation processes, but these materials require performance demonstrations under more appropriate water treatment conditions that use reactor configurations consistent with currently available UV-based POU/POE devices;
- Fibers may help address recognized needs in decentralized treatment for treatment of uranium, disinfection of pathogens, and electrochemical treatment approaches, which are gaining in popularity, and;
- Recommendations for future studies include a better understanding of interactions among material components, benchmarking to appropriate commercial analogs, reliable scaling in industrial production, and full-scale demonstrations with appropriate treatment conditions.

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