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Perspective

3D-Printed Materials for Wastewater Treatment

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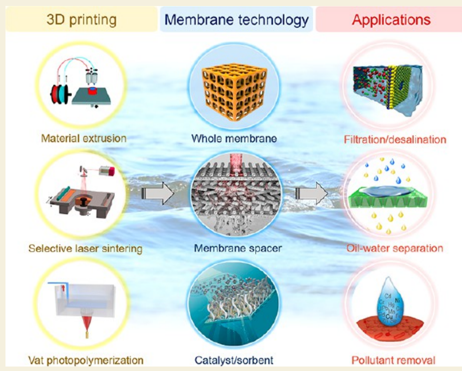
ABSTRACT: The increasing levels of water pollution pose an imminent threat to human health and the environment. Current modalities of wastewater treatment necessitate expensive instrumentation and generate large amounts of waste, thus failing to provide ecofriendly and sustainable solutions for water purification. Over the years, novel additive manufacturing technology, also known as three-dimensional (3D) printing, has propelled remarkable innovation in different disciplines owing to its capability to fabricate customized geometric objects rapidly and cost-effectively with minimal byproducts and hence undoubtedly emerged as a promising alternative for wastewater treatment. Especially in membrane technology, 3D printing enables the designing of ultrathin membranes and membrane modules layer-by-layer with different morphologies, complex hierarchical structures, and a wide variety of materials otherwise unmet using conventional fabrication strategies. Extensive research has been dedicated to preparing membrane spacers with excellent surface properties, potentially improving the membrane filtration performance for water remediation. The revolutionary developments in membrane module fabrication have driven the utilization of 3D printing approaches toward manufacturing advanced membrane components, including biocarriers, sorbents, catalysts, and even whole membranes. This perspective highlights recent advances and essential outcomes in 3D printing technologies for wastewater treatment. First, different 3D printing techniques, such as material extrusion, selective laser sintering (SLS), and vat photopolymerization, emphasizing membrane fabrication, are briefly discussed. Importantly, in this Perspective, we focus on the unique 3D-printed membrane modules, namely, feed spacers, biocarriers, sorbents, and so on. The unparalleled advantages of 3D printed membrane components in surface area, geometry, and thickness and their influence on antifouling, removal efficiency, and overall membrane performance are underlined. Moreover, the salient applications of 3D printing technologies for water desalination, oil–water separation, heavy metal and organic pollutant removal, and nuclear decontamination are also outlined. This Perspective summarizes the recent works, current limitations, and future outlook of 3D-printed membrane technologies for wastewater treatment.



KEYWORDS: *Additive manufacturing, 3D printing, membrane technology, membrane modules, wastewater treatment*

1. INTRODUCTION

Wastewater treatment has become a global concern fueled by pollutants from households, industries, and agricultural practices. Approximately 71% of the earth's surface is covered by water, and only a minuscule fraction of less than 1% is readily available as drinking water. It has been projected that water demand will rise by 55% in 2050 due to population growth, while the global water deficit is expected to reach 40% by 2030.[1] As per World Health Organization (WHO) reports, one out of four individuals lack access to safe drinking water, and the absence of this essential resource contributed to 6% of deaths in 2017 in low-income countries.[2] Therefore, it is highly imperative to ensure the sustainable utilization of water, which could be accomplished by developing effective wastewater treatment methods to meet the escalating demand. Although conventional wastewater treatment strategies such as flocculation, precipitation, ion exchange, and adsorption have shown promising results in treating wastewater, these techniques are often associated with several limitations. For instance, using inorganic flocculants often leads to the generation of significant amounts of sludge, which presents a major environmental issue. Additionally, precipitation leads to the generation of toxic byproducts like H_2S fumes and other colloidal sulfides. On the other hand, ion exchange-based treatments are only suitable for recovering metals at high concentrations. While adsorption is a cost-effective process for removing pollutants from wastewater, it requires thorough pretreatment of sorbents to achieve higher water treatment efficiency. Even though the aforementioned procedures reduce the concentration of contaminants in wastewater, they are not suitable for achieving the necessary regulatory standards for drinking water. More-

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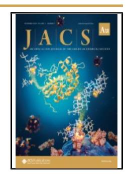

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over, these techniques are energy intensive and cannot eliminate toxic pollutants, including heavy metals, phosphorus, and nitrogen.[3]

In recent years, membrane-based techniques have gained substantial attention for wastewater treatment due to their high contaminant removal efficiency, compact modular design, and selective separation. However, their utilization is typically impeded by challenges related to membrane fouling and the inability to remove dissolved inorganic substances, such as salts and small ions, from water. To address these challenges, ongoing research and advancements are focused on developing new frontiers for achieving highly effective water remediation. In this context, one such ground-breaking technology is three-dimensional (3D) printing, which has revolutionized design, prototyping, and manufacturing. By leveraging the capabilities of 3D printing, novel membrane designs with enhanced efficiency and performance can be developed. The flexibility of 3D printing allows for the creation of complex geometries and customized structures, enabling the production of membranes tailored to specific wastewater treatment needs.[4] The conventional methods for membrane fabrication include thermally induced phase separation (TIPS), vapor induced separation, and nonsolvent induced phase separation (NIPS) which require large quantities of solvent, toxic monomer materials, and high carbon footprint and generate large amounts of waste leading to adverse effects on human health and environment.[5,6] On the other hand, 3D printing being a sustainable method eliminates the need for solvent discharge, offering a more environmentally friendly approach and allowing the development of innovative membranes with diverse features by choosing from a wide range of materials.[4,7] The application of 3D printing technology in wastewater treatment can be traced back to 2016 when researchers from the University of Bath utilized a 3D printer to fabricate a membrane module for ultrafiltration.[8] Since then, various kinds of 3D printing techniques, such as selective laser sintering (SLS), fused deposition modeling (FDM), solvent-based slurry stereolithography (3S), inkjet printing, and digital light processing (DLP), have been utilized for fabricating whole membranes as well as their module parts for water treatment.[4,5,8] In a typical 3D printing process, computer models prepare and direct ink to transform into 3D objects. This layer-by-layer fabrication approach involves printing or adding one layer at a time to create a 3D structure based on a 3D computer-aided design (CAD) model. During the process, the model is sliced into hundreds or thousands of layers employing a slicing software/additive manufacturing system which is typically dedicated to a specific 3D printer, and several printing parameters such as printing speed and pressure, layer height (resolution), infill pattern, and infill density are optimized. Postprinting, the 3D-printed item, typically requires postprocessing to remove unnecessary excess material and stabilize curing, the extent of which depends on the printer and printed structure type. 3D printing offers numerous advantages, including facile process, accuracy, uniformity, sustainability, adaptability, and cost-effectiveness.[9−11] Importantly, 3D printing allows the fabrication of complex geometrical structures, which is crucial for precisely manufacturing membranes and membrane modules for wastewater treatment. Although the implementation of 3D printing technology in water treatment systems commenced with membrane module research, the manufacture of membranes is an emerging area of investigation. Until now, 3D printing membranes have been explicitly used for industrial waste treatments, especially for the degradation of organic pollutants, oily effluents, and heavy metals generated by manufacturing and pharmaceutical industries.[12,13] The existing methods for industrial wastewater treatment are based on physiochemical and biological methods which are energy intensive, have poor permeability, require complicated preand post-treatments, and lead to clogging.[12] Not only for industrial waste, 3D printing has also been used for domestic water treatments and desalination applications for recycling and reusing undrinkable water.[14,15] In this regard, 3D printing offers a sustainable solution for domestic as well as industrial wastewater treatment by enacting a low-waste generating method with negligible chemical exposures in addition to being economical and precise, thereby addressing the gaps with existing wastewater techniques. Notably, 3D printing allows for the fabrication of even more resilient and ultrathin membranes with homogeneous pore diameters.[16−18] In addition, the layer-by-layer nanoscale printing of membranes would also enable well-organized pore sizes with robustness and preparation of intricate membrane structures with improved mass transfer, minimal fouling, and lower pressure loss.[19−22] Therefore, this integration of 3D printing technology to manufacture a variety of membrane modules offers a pathway toward improved wastewater treatment solutions and holds great potential in meeting the demands of sustainable water management.

The advantageous nature of 3D printing strategies has led to immense utilization for wastewater treatment, as evidenced by the increasing number of publications in the literature. Various reviews highlighting the applications of 3D printing in wastewater treatment have also increased in the past decade. For instance, Lee et al. gave a substantial overview of the production of spacers and membranes using various types of 3D printers.[23] Similarly, Yanar et al. broadly described the applications of 3D printing in the fabrication of membrane types such as ceramics and polymers and spacers. The authors also discussed the prospects and limitations of 3D printing in wastewater treatment. However, their studies did not thoroughly examine different module types and their specific applications in wastewater treatment.[24] Thus, to address this gap, the present Perspective presents a comprehensive analysis and delves into the utilization of 3D printing technology in the manufacturing of whole membranes and their modules, such as spacers, biocarriers, and sorbents.[25−32] This paper also highlights the different 3D printing techniques that have been widely employed for 3D printing of membrane structures and then moves on to the application of these modules in a wide range of fields, including water desalination, oil−water separation, water filtration, separation processes, and heavy metal removal. Finally, this paper aims to provide an extensive study of the advancements, challenges, potential applications, and future perspectives of 3D printing in these membrane-based systems.

2. OVERVIEW OF 3D PRINTING TECHNOLOGIES USED FOR WASTEWATER TREATMENT

With the advent of Industrial Revolution 4.0, there has been a paradigm shift in the manufacturing industry from conventional subtractive manufacturing toward advanced additive manufacturing, i.e., 3D printing. This technology involves fabricating the desired product computationally in CAD software, slicing it into 2D layers (G-code), feeding the proper raw materials into a 3D printer, and finally building the object from the virtual file layer-by-layer. Recently, different 3D

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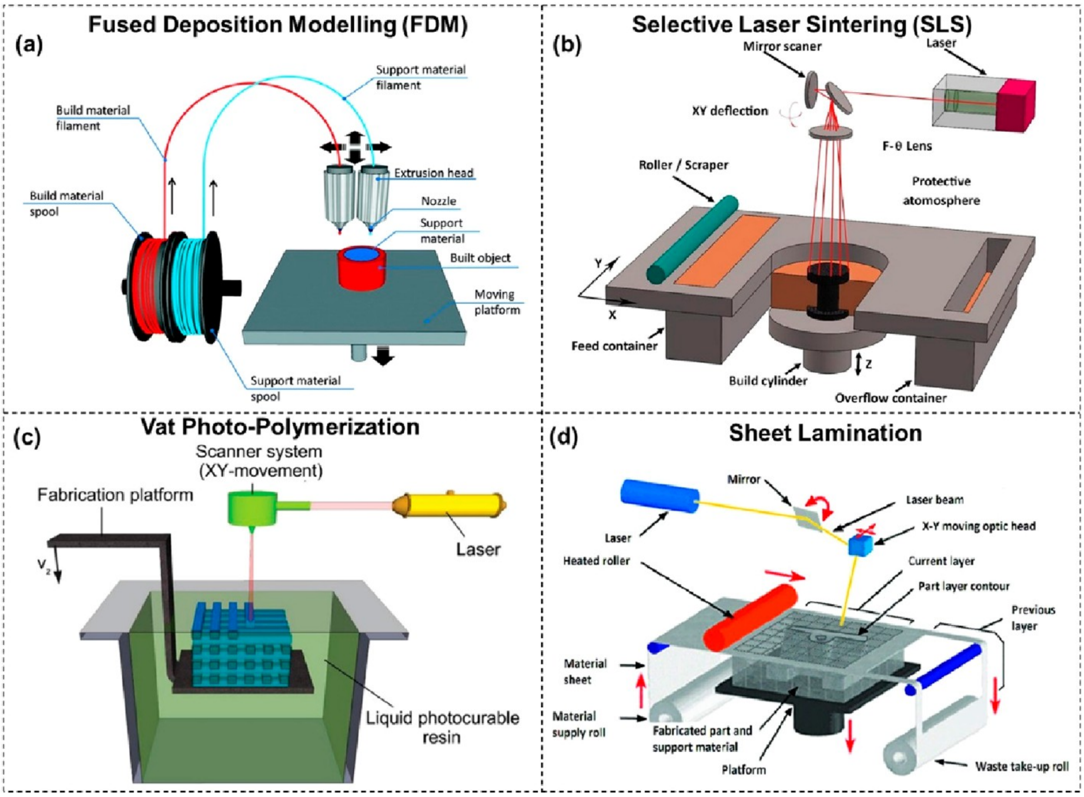

Figure 1. Different types of 3D printing technologies are employed for wastewater treatment. (a) Fused deposition modeling (FDM). Reproduced with permission from ref 25. Copyright 2016 Royal Society of Chemistry. (b) Selective laser sintering (SLS). Reproduced with permission from ref 26. Copyright 2014 Elsevier. (c) Vat photopolymerization. Reproduced with permission from ref 47. Copyright 2012 Elsevier. (d) Sheet lamination. Reproduced with permission from ref 59. Copyright 2020 John Wiley & Sons.

printing technologies have been studied for developing customized membranes that provide scalable and high-efficiency solutions for wastewater management. Moreover, 3D printing offers great flexibility in terms of design, a wide variety of material choices, minimal energy consumption, negligible material wastage, the least byproducts, and far less carbon footprint relative to subtractive processes, which have been employed for preparing novel membranes, module spacers, catalysts, and adsorbents for carrying out wastewater filtration and desalination, thus making it a lucrative strategy for water treatment. Until now, printing approaches based on extrusion, SLS, and Vat photopolymerization have been extensively employed for fabricating membranes for wastewater treatment. A detailed discussion of each technique is provided in the following subsections.

2.1. Extrusion-Based Printing

Extrusion-based printing relies on the application of continuous pressure to direct the material onto the printing platform from the print head, followed by its solidification. Of all material extrusion strategies, FDM has been widely explored owing to its simplicity in operation, flexibility in material selection, ability to produce durable components, and easy tunability of morphological properties of the printed membranes. As shown in Figure 1a, in this process, the molten thermoplastic polymer is extruded out of a thin nozzle under high pressure and deposited after solidification on a build platform in a layer-wise fashion.[25] The polymers being used as filaments must have an appropriate melt viscosity to extrude from thin nozzles. Commonly used materials for extrusion-based printing include polycarbonate (PC), poly(lactic acid) (PLA), acrylonitrile butadiene styrene (ABS), and poly(ether ether ketone) (PEEK), with or without additives. Particularly for water treatment, FDM has widely been utilized to design membrane modules such as feed spacers, which aid in maintaining membrane integrity and providing fluid flow. Until now, different geometries of feed spacers, such as ladder-like, herringbones, and helices, have been fabricated using FDM for wastewater treatment.[8,20,33−36] Polymeric feed spacers have been widely developed using FDM-based 3D printing, which have exhibited superior membrane performance, particularly in the case of membrane fouling. For successfully preparing polymeric modules using FDM, it is imperative to regulate the parameters related to polymer rheology and the gelation mechanism. Recently, FDM has also opened new avenues for printing composite spacers based on TiO_2,[37] PLA, and polymer-zeolite composites for multifunctional filtration membranes. Although FDM-based 3D printing has demonstrated promising applications in wastewater treatment, it is also associated with several limitations. One of the major limitation includes the low mechanical strength of printed parts due to the anisotropic properties caused by layer-by-layer deposition. Another drawback is related to the resolution, which is limited by the diameter of the nozzle (50−200 μm) being used[38] and the presence of cavities on the surface due to heterogeneity between layers.[39] To address these limitations, efforts are being dedicated to changing the nozzle geometry, which could allow for precise printing of membrane module parts with widths < 100 μm without clogging, improving the overall filtration performance.

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Another commonly used extrusion process for membrane technology is direct ink writing (DIW) which prints polymers from solutions, usually in the form of gels or pastes that further undergo solidification due to gelation, evaporation, or solvent-driven events.[40] This approach has been used to print intricate ceramic and polymer membranes with improved stability for applications in caustic environments.[41] For printing structures with high integrity using DIW, the precursor ink must exhibit excellent rheological behavior regarding apparent viscosity, yield stress under shear, and viscoelastic properties. Interestingly, DIW was also d to print carbon-based aerogels-based membranes, which exhibited excellent solar wastewater remediation performances, which have not been reported using any other 3D printing strategies.[42]

2.2. Selective Laser Sintering (SLS)

SLS is a powder-based 3D printing technique that fabricates 3D structures by sintering a thin layer of powder with the help of high-energy lasers. As shown in Figure 1b, the critical components of this 3D printing technology include a build platform, powder chamber, printing chamber with powder bed, and laser beam source.[26] Typically, the printing chamber and powder chamber are heated to a temperature below the melting point of the material, and then, the powder material is selectively fused into the predefined shape by sintering the powder bed using a laser source. Subsequently, to cover the sintered area, a fresh layer of powder is refilled onto the top surface of the powder bed, which is consistently repeated until the required 3D structure is obtained. Different materials, including polymers, ceramics, and composites in powder forms, have been used as printing materials in the SLS-based 3D printing approach. For water treatment, SLS was the first ever 3D printing technique to fabricate membrane spacers in the 2000s. Then, novel spacers with intricate geometries, such as modified filaments, twisted tapes, and multilayered structures, were fabricated using SLS. Because the SLS utilizes the powder bed as the support material for printing and does not require additional secondary support, this technique has become extremely popular for printing complex membrane geometries for wastewater treatment. The ability of SLS to process a wide variety of materials ranging from alloys to polymers and ceramics also provides unparalleled advantages for membrane fabrication. Moreover, this technology can achieve a moderate resolution of 80−250 μm and results in denser membranes with improved surface roughness.[43,44] Various studies have reported that the quality of the printed membranes as well as membrane modules such as feed spacers is substantially influenced by the particle size of powder, scan speed and spacing, laser power, sheet width, and energy density. To date, membrane modules based on polymers such as polypropylene (PP), polyamide (PA), and metal−organic frameworks (MOFs) have been printed by using SLS for water purification. Recently, apart from printing intricate structures for feed spacers, SLS has also extended to print advanced membrane modules, especially photocatalysts with different morphologies for wastewater treatment. Although SLS-based 3D printed parts provide numerous advantages for wastewater treatment, the final structures printed are not very sharp, have lower mechanical strength, and often necessitate postprocessing steps for their usage which limits their applications.[45,46] Further, longer printing times and the requirement for expensive equipment also pose significant challenges with SLS technology.

2.3. Vat Photopolymerization

This 3D printing process uses a light source to drive polymerization reactions in photosensitive materials in a controlled manner. During the process, a small dose of photoinitiator is blended with the liquid resin to generate free radicals and initiate the photoinduced polymerization process (Figure 1c).[47] Based on the type of light source and mechanism of polymerization used, vat photopolymerization is classified into subcategories: digital light processing (DLP), photopolymer jetting, stereolithography (SLA), and continuous liquid interface production (CLIP). DLP utilizes a UV light source from a digital projector or micromirror device (DMD) for inducing instantaneous polymerization. The light source is shined over the entire surface of the vat in a single projection, thereby curing one layer at a time, as a result of which DLP provides a faster printing speed compared with other 3D printing techniques. Irrespective of the layer number and complexity of the structure to be printed, DLP can typically achieve a resolution of about 1 μm.[48,49] Various reports have suggested that the printing resolution of DLP can be improved by incorporating light absorbers and materials for accelerating the polymerization reactions. The significant difference between DLP and SLS is the type of light source employed with SLS utilizing a UV laser instead of a projector source. In the SLS technology, the build platform is placed at a distance equivalent to the one-layer height from the liquid resin surface composed of photosensitive polymer inside the vat. Then the UV laser is used to cure and solidify the liquid resin layer by layer selectively. As per the predetermined path, the laser source controlled by the computer scans over the resin surface to cure it to a defined depth, followed by raising the build platform to facilitate the recoating of the photocurable resin, and this process is repeated until the final 3D structure is obtained. This technique can achieve a resolution of ∼10 μm, lower than the DLP-based 3D printing approach.[43] In recent years, CLIP technology has emerged as the fastest type of vat photopolymerization strategy, which can print structures with printing speeds that are 100 times faster compared to DLP and SLA by facilitating continuous printing with oxygen as polymerization inhibitors.[50−52] Regardless of the type of photopolymerization technique used, the quality of the printed structure is known to be influenced by tailoring the polymerization parameters, including UV power intensity, scanning speed, layer thickness, and exposure time. Owing to its high accuracy in printing slices down to microscale, vat photopolymerization-based approaches have been widely used in membrane filtration, particularly for designing feed spacers. Among all, DLP technology has shown promising results in developing highly complex ceramic spacers with microsized patterns, which have, in turn, contributed to reducing energy consumption and improving the flux of membrane-based filtration processes.[53] On the other hand, SLA presents limitations in membrane fabrication due to the decreased mechanical strength and robustness of the structures when placed in water over time. Even though photopolymerization techniques can generate high-resolution membrane modules, the requirement of postprocessing steps to improve the mechanical properties of the printed parts is a significant roadblock for practical applications. In addition, unlike extrusion-based printing, DLP and SLA techniques cannot execute multimaterial printing.

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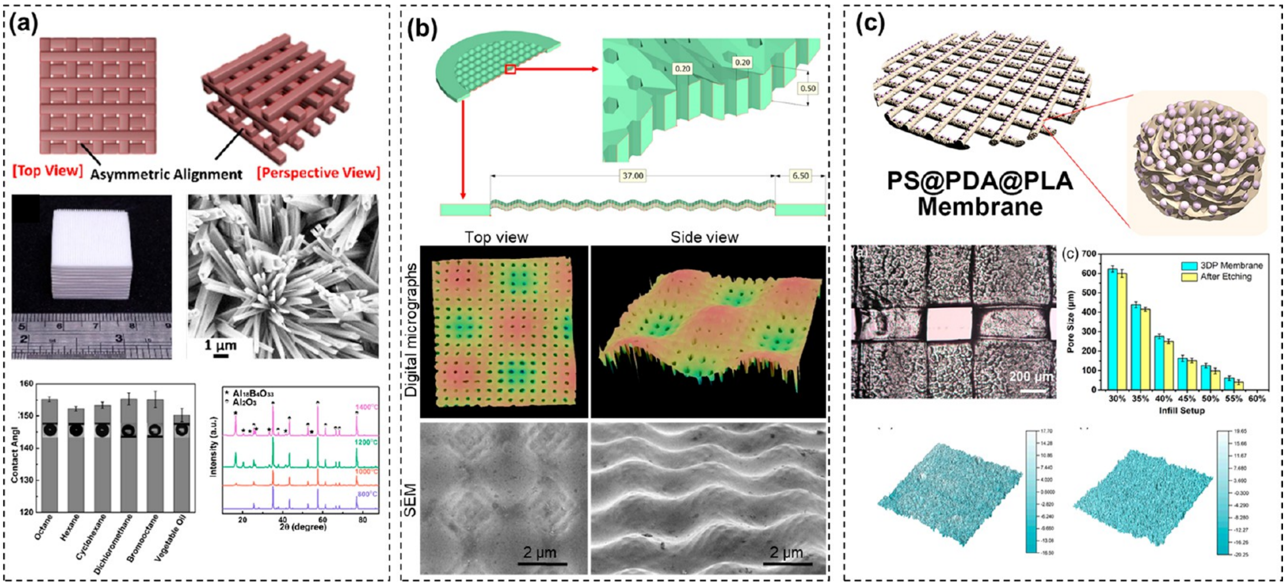

Figure 2. Fabrication of various types of whole membranes for wastewater treatment. (a) Aluminum borate whisker-based ceramic membrane meshes with high super hydrophilicity fabricated using 3D printing. Reproduced with permission from ref 71. Copyright 2019 Elsevier. (b) Flat and wavy structured ABS composite membranes designed using multijet 3D printing. Reproduced with permission from ref 72. Copyright 2019 Elsevier. (c) Superhydrophobic polymer porous membranes based on PLA with bioinspired lotus leaf-like structures fabricated using FDM. Reproduced with permission from ref 77. Copyright 2018 John Wiley and Sons.

2.4. Others

Other printing modalities that have also been used for fabricating membrane components for wastewater treatment include polyjet printing, sheet lamination, and other emerging techniques. A polyjet printer uses multiple jet heads to deposit polymer over a platform, where it gets cured with UV, and hence the final product has a much smoother surface finish compared to other methodologies, such as the FDM-based modules.[54] Moreover, polyjet printing involves lower costs and higher spacer resolution than other 3D printing techniques.[55] Polyjet printing has been widely used to fabricate spacers and biocarriers for membrane filtration and ultrafiltration applications. Most of the printed structures are based on polymers, such as PA, ABS, PP, and PLA. A literature survey reveals that the printing orientation, *y*-axis spacing, postprocessing steps, and aging time greatly influence the characteristics of the membrane modules printed using polyjet printing.[56−58] In addition, sheet lamination is another 3D printing technique used to print metal-based membrane structures. As depicted in Figure 1d, it involves a heat-activated sheet that is allowed to bond over the substrate via the pressure generated by the heat roller. According to the CAD model, the laser beam patterns each layer, and the thermoplastic adhesive between the layers helps bind the layers with the previous ones.[59] The primary advantage of this technique is its ability to generate flat sheets and hollow fibers that could potentially be used for different water purification modules.[60] However, rough surface finish and poor resolution are the major limitations restricting its adoption for wastewater remediation. Recently, other membrane fabrication techniques such as electrospinning and electro-spraying have also been regarded by some researchers as 3D printing approaches owing to their principle of creating objects by layer-by-layer and bottom-up deposition from the bulk layer.[61,62] These techniques were typically used to prepare highly uniform polymeric membranes for wastewater treatment.[61−64] For instance, Chowdhury et al. combined electrospraying with 3D printing for the first time to fabricate an ultrathin composite membrane for desalination applications.[61] Specifically, the integrated platform was used to develop the polyimide selective layer of the membrane where the monomers were deposited on the substrates to form the polyimide layer, and the 3D printing was used to control the thickness and roughness of the layers in order to obtain thin membranes of 15 nm. In another study, Su et al. prepared the superhydrophobic polymeric membranes coated with silica nanoparticles which are intercalated within the polymeric matrix by using electrospinning/spraying with direct ink writing 3D printing techniques for distillation.[64] Compared with conventionally prepared hydrophobic membranes, the electrospinning/spraying membranes were mechanically robust with higher wettability and high vapor permeability.

3. 3D-PRINTED MODULES FOR WASTEWATER TREATMENT

Recent improvements in 3D-printed materials and techniques have greatly aided the manufacturing of numerous modules for various applications involving water treatment. Innovative filtration and desalination membranes, as well as their module components, such as feed spacers, adsorbents, biocarriers, and catalytic structures displaying enhanced membrane performance and efficiency, have been designed using precise and adaptable 3D printing approaches.

3.1. Whole Membranes

Owing to its high efficacy and cost-effectiveness, membrane technology has been widely utilized for water treatment. Typically, the employed membranes are polymer or ceramic-based and are fabricated using[65] conventional techniques such as phase inversion, hollow fiber spinning, stretching, electrospinning, solvent casting, phase separation, extrusion.[66,67] However, these traditional manufacturing techniques have several constraints that could affect their efficacy and productivity. The limitations of the fabrication processes and

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the resulting membrane structures generally bring these constraints. These restrictions include nonuniformity in pore sizes and distributions, which may cause alterations to permeability and selectivity, impacting overall performance. Similarly, manufactured structures with reduced structural integrity can suffer from membrane degradation, distortion, and a reduction in the lifespan of their membrane structures. And a significant disadvantage of traditional approaches is membrane fouling, which occurs due to simplistic surface shape and constrained design flexibility.[68] To overcome these challenges, 3D printing approaches have emerged as promising alternatives that possess the potential to fabricate complex membranes with hierarchical structures. Compared to conventional processes, 3D printing enables the design of membranes with high degrees of freedom, high precision, better resolution, and low cost and offers excellent control over thickness and porosity. Until now, 3D printing technologies have been majorly used to develop ceramic-based membranes with complex internal structures for water filtration and oil−water separation. For instance, Hwa et al. developed 3D-printed porous ceramic membranes using Kanakra clay powder for water filtration.[69] Similarly, Akowanou et al. fabricated 3D-printed ceramic-based water filters using natural clay containing quartz and kaolinite.[70] The ceramic paste was prepared by mixing rice husk, acting as a pore-forming agent. 3D printing of the filter was carried out using a modified 3D printer with a nozzle diameter of 1 mm and a 100% infill density. The printed structures were left for drying at room temperature, followed by sintering at 900 °C. Further filter performance was analyzed using surface water treatment, revealing good removal of turbidity and organic matter with reduced fouling. For water−oil separations, Chen et al. designed the 3D printed super hydrophilic ceramic meshes combined with in situ grown aluminum borate whiskers (Figure 2a).[71] The whiskers were designed into shapes, including honeycomb meshes, sponges, and scaffolds, and displayed high durability against high temperatures and corrosive conditions.

In addition, various studies have reported the potential of 3D printing strategies for fabricating components of composite membranes, especially their substrates. Al Shimmery et al. employed 3D printing for designing the ABS-based support layer with flat and wavy surface structures for composite membranes and evaluated its filtration as well as antifouling performance, as shown in Figure 2b.[72] The membrane supports (50 mm) were using a multijet-based 3D printer, and the PES selective layers were deposited onto the printed support layers via vacuum filtration. The design specifications for the 3D printed supports were optimized based on the mechanical properties of the materials, the resolution of the 3D printer, and the turbulence generated by the features. While the resolution of the 3D printer dictated the pore size and interpore distance, the mechanical properties of the materials significantly influenced the thickness of the 3D printed support layers. The obtained results revealed that the rate of water permeability and the permeance recovery ratio of the membranes with wavy 3D printed supports was 30% and 52% higher than their flat counterparts, respectively, hence demonstrating its antifouling behavior.[73] In another study, Yuan et al. prepared polysulfone membranes using 3D printing technology with a switchable wettability for gravity-driven oil−water separations.[73]

Although the research related to the development of complete membranes by 3D printing technologies is limited, recently, there have been few reports suggesting the utilization of 3D printing for polymeric membranes. For the first time, Lv et al. in 2017 reported the development of 3D printed superhydrophobic porous membranes based on nano silica-functionalized polydimethylsiloxane (PDMS) ink using an extrusion-based homemade 3D printer consisting of computer-driven 3-axis movement platform and a micronozzle (diameter: 150 μm).[74] The 3D printing method enabled the filamentary extrusion of the ink from the micronozzle, which resulted in the layer-by-layer deposition of PDMS filaments onto the porous structures followed by thermal curing, thereby overcoming the challenge related to weak interface adhesion in membranes fabricated by traditional approaches. Moreover, the superhydrophobicity of the polymeric membranes was obtained due to the ability of the 3D printers to create surface roughness in the order of submillimeters, thereby eliminating the requirement of high loading density of silica nanoparticles. The as-printed porous polymeric membranes displayed a pore-size dependent oil−water separation efficiency with the highest efficiency of 99.6% achieved at a pore size of 0.37 mm.[75] Another study employed the liquid-based DLP approach based on Schwarz-P triple periodic minimal surface (TPMS) design to directly print PDMS membranes with high gas permeability.[76] Moreover, novel 3D-printed polymeric membranes and bioinspired features were fabricated for water−oil separation. Inspired by a cactus plant, Shin et al. 3D printed a mold that acted as a template for fabricating PDMS sponge membranes with porous structures.[77] Another study reported the fabrication of lotus leaf-based bioinspired 3D printed superhydrophobic poly(lactic acid) (PLA) membranes for oil−water separations (Figure 2c).[78] Further, to develop extremely thin membranes, a CLIP- based 3D printing approach was utilized, which could continuously print objects by enhancing the printing speed, in contrast to the previously reported layer-by-layer strategies. Even though the reported 3D printing strategies have shown promising features for water treatment, their applications are limited in printing smaller pore sizes with better surface finish, which are highly desirable for advanced water filtrations such as ultrafiltration and nanofiltration modules.

3.1.1. Membrane Spacers. Instead of constructing whole membranes, increasing efforts are dedicated to fabricating membrane module parts using 3D printing for wastewater treatment. One such important membrane module is channel feed spacers, which are highly essential in establishing continuous fluid flow and mixing. Typically used in spiral wound membranes (SWM), feed spacers ensure that the fluid passes at a constant feed, prevent impairment of the active layer of the membrane, increase turbulence, and reduce fouling. The configuration and orientation of placing the feed spacers between the membranes are crucial in determining the performance of the membrane. During the fabrication, the membrane is first folded in half and faced inward, followed by introduction of the feed spacer between the folded membranes. A key requirement for designing the channel feed spacers is to limit the dead zone formation to reduce particle deposition, leading to fouling. Although conventional techniques such as heat extrusion, vacuum forming, or molding have been used to design feed spacers, they cannot fabricate spacers with complex designs and geometries. Therefore, owing to its design freedoms, 3D

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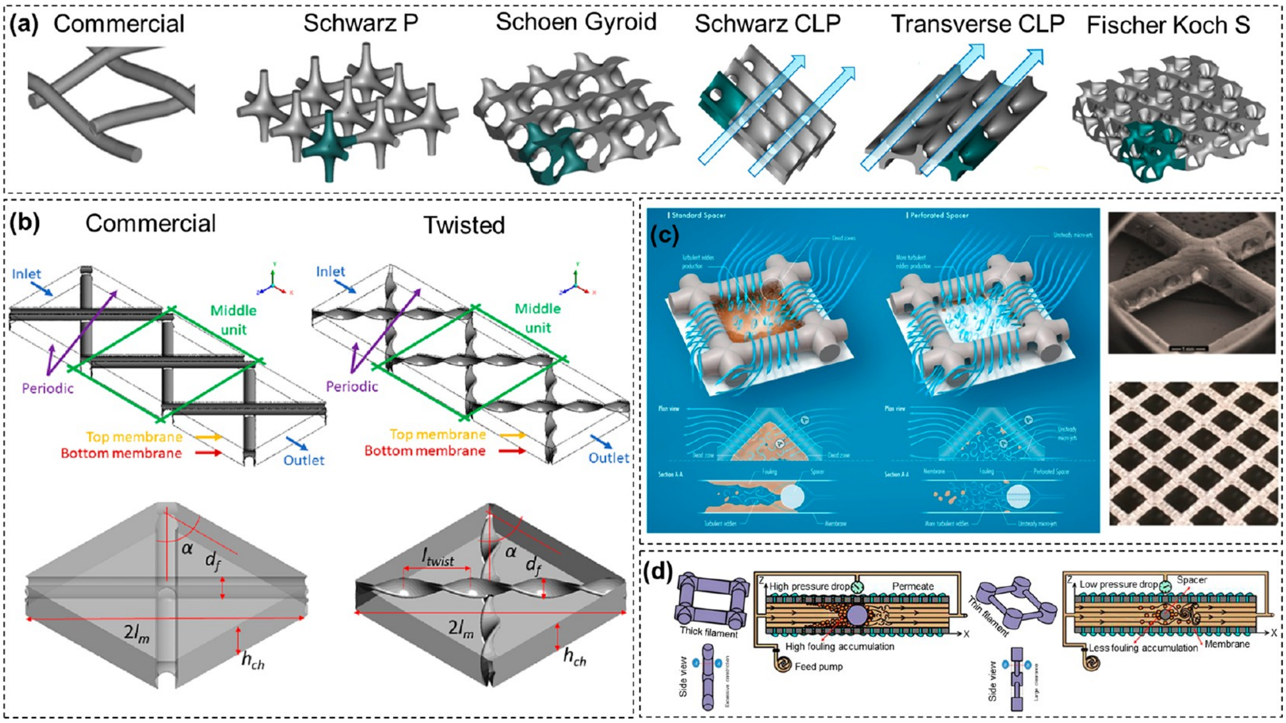

Figure 3. Different designs of the membrane spacers were prepared by 3D printing. (a) SLS printed TPMS spacers with five different topologies. Reproduced with permission from ref 19. Copyright 2018 Elsevier. (b) Structural comparison between 3D-printed twisted membrane feed spacers and conventional ones. Reproduced with permission from ref 80. Copyright 2023 Elsevier. (c) Perforated cylindrical column spacers with different hole geometries designed by DLP. Reproduced with permission from ref 81. Copyright 2018 Elsevier. (d) DLP-based distinct cylindrical spacers for enhancing the specific energy consumption of membranes. Reproduced with permission from ref 82. Copyright 2019 Elsevier.

printing technologies such as SLS, DLP, FDM, and polyjet techniques have widely been used to design spacers in various configurations, including multilayered structures, triply periodic minimal surface (TPMS), twisted tapes, helices, and ladders. For instance, Sreedhar et al. employed the SLS-based 3D printing approach for designing TPMS spacers for reverse osmosis (RO) and ultrafiltration (UF) processes. The obtained results depicted that the SLS-printed spacers significantly increased the permeability of the flux by 15.5% for RO and 38% for UF and decreased the pressure drop inside the filtration channels, compared to the commercially available spacers.[79] Apart from filtration, 3D-printed TPMS feed spacers have also been utilized for membrane-distillation-based water treatments. During the distillation process, channel feed spacers act as turbulence promoters for amplifying the mass transfer for better separation by suppressing the polarization-driven boundary buildup between the membrane layer and fluids. Thomas et al. fabricated the TPMS spacers with five different designs, as depicted in Figure 3a, for carrying out membrane distillation by using SLS-based 3D printing.[19] As compared to commercial net-type spacers, the as-fabricated 3D printed spacers demonstrated better fluid flux, higher heat transfer coefficient, and overall better performance in treating solutions with high fouling susceptibility. Importantly, the authors suggested that the utilized 3D printing technology enabled the fabrication of spacers in a single step, unlike the conventional strategies.[19] Later, Li et al. pioneered the manufacturing of 3D-printed spacers with twisted tapes using the SLS-based 3D printing approach, which resulted in enhanced mass transfer due to the generation of longitudinal vortices by the helical filaments.[80] In another study, Chong et al. in 2023 suggested that the better performance of the twisted tape spacers compared to the other configurations is owing to their significantly higher Sherwood number and decreased friction factor, which minimizes the stagnant zone formation and results in vortex generation as indicated in the previous studies (Figure 3b).[80] Consequently, net-type feed spacers have also been reported to be prepared by the SLS method. Interestingly, the authors observed that the energy density parameters, such as the laser power, scanning distance, and speed used during printing, were directly related to the mechanical properties of the printed spacers. With the increase in the energy density, the Young's modulus of the printed spacer increased, which resulted in the enhanced tensile strength of the spacer.

In addition to developing spacers with high mechanical strength, it is imperative to reduce the concentration polarization in membranes to alleviate membrane fouling issues for filtration applications. To address this, Kerdi et al. designed modified cylindrical filaments by introducing perforations in the spacer design via the DLP-based 3D printing approach, as shown in Figure 3c.[81] The effect of hole geometries (1, 2, or 3 holes) on pressure drop, permeate flux, and membrane fouling was assessed. The 3-hole spacer showed the most effective energy consumption and hydraulic resistance with a 60% pressure drop, while the 1-hole spacer showed maximum (75%) flux recovery with the least fouling on the membrane.[82] Furthermore, the performances of membranes equipped with spacers fabricated by different 3D printing technologies, including SLS, polyjet, and FDM, were also compared. It was revealed that irrespective of the printing method, all the 3D printed spacers displayed enhanced mass transfer compared to the conventionally fabricated membranes. Subsequently, Ali et al. fabricated cylindrical membrane spacers

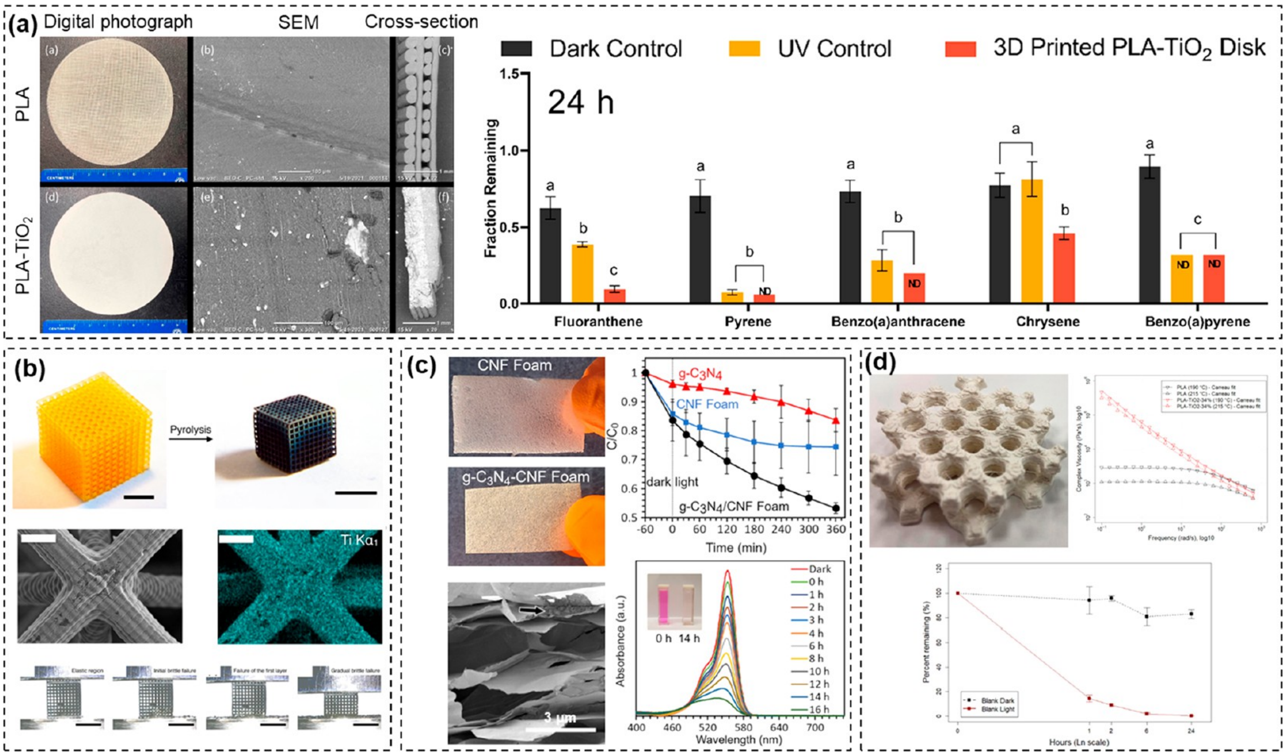

Figure 4. Fabrication of 3D-printed catalytic membrane modules for wastewater purification. (a) PLA-TiO_2 disk photocatalyst printed using a twin-screw extruder for degradation of polycyclic aromatic hydrocarbons using 3D printed PLA-TiO_2 structures. Reproduced with permission from ref 93. Copyright 2022 American Chemical Society. (b) Titanium photocatalyst printed using stereolithography for household water disinfection. Reproduced with permission from ref 95. Copyright 2018 Elsevier. (c) 3D printed floating photocatalyst modified with cellulose nanofibers (CNFs) and graphitic carbon nitride (g-C_3N_4) for rhodamine dye removal. Reproduced with permission from ref 96. Copyright 2020 American Chemical Society. (d) Composite photocatalyst based on TiO_2 prepared using fused filament fabrication (FFF) technology for microcystin (MC) toxin elimination. Reproduced with permission from ref 99. Copyright 2023 Elsevier.

in 2019which diminished the unsteady behavior of the flow by enhancing the clearance between the membrane and filament, which further led to reduced pressure drop and specific energy consumption (SEC) of the membrane filtration systems (Figure 3d).[82,83]

3.2. Photocatalysts

Photocatalysts which can get activated in the presence of a light source have emerged as prominent candidates for the oxidation of organic contaminants for their removal. In the presence of a light source of a particular wavelength, photocatalyst absorbs photons, and electron−hole pairs are created, which causes the catalyst surface to react with water and dissolved oxygen, thus leading to the generation of ROS species, hydroxyl (·OH) and oxide radicals (O^{2-}), via redox reactions. Conventionally, photocatalysts have been synthesized by various strategies such as reverse micelles, sol−gel process, metal−organic chemical vapor deposition (MOCVD), aerosol synthesis, wet synthesis by precipitation of hydroxides from salts, and microemulsion.[84−87] However, the existing methodologies are often challenged by disadvantages, such as smaller surface area and easy detachment of catalytic materials from the substrates. To overcome these constraints, 3D printing has been introduced as an effective and low-cost alternative for the development of photocatalysts due to its potential to fabricate structures with complicated geometries, which offer a high surface area to volume ratios and high porosity with the ability to finetune the surface of the photocatalysts. Until now, 3D printing technologies, including laser metal deposition (LMD), material jetting, sheet lamination, binder jetting, material extrusion, FDM, and vat photopolymerization have been widely explored for preparing photocatalysts such as TiO_2,[88] titania, ZnO,[89] Fe_2O_3,[90] C_3N_4[91] and bismuth-based[92] semiconductors for wastewater treatment applications. Among these, TiO_2-based photocatalysts have been widely fabricated using 3D printing approaches, owing to their outstanding electronic and optical properties, low toxicity, and high chemical stability. For instance, Sangiorgi et al. employed the FDM-based 3D printing strategy for fabricating TiO_2 nanoparticles immobilized onto PLA, which were used for developing photocatalytic filters for degrading methyl orange (MO).[93] By optimizing the printing parameters, such as filter diameter, infill, and scaffold height, extremely uniform 3D surface geometry was obtained, which led to 100% degradation of MO after 24 h of light irradiation.[93] Similarly, McQueen et al. fabricated 3D-printed TiO_2 composites, as depicted in Figure 4a, for the photocatalytic degradation of polycyclic aromatic hydrocarbons (PAHs) in sediment-contaminated water.[93] To prepare the composites, TiO_2 was mixed with PLA and printed using a benchtop extrusion-based 3D printer to create simple disk geometries with a 50% infill pattern and 0.2 mm layer height, ultimately leading to the generation of a durable structure with the increased surface activity of the photocatalysts. The degradation study showed that the 3D printed TiO_2-embedded composite polymer not only efficiently degraded PAHs within 48 h of treatment but also increased the degradation kinetics of 4−6 rings PAHs by subsequently generating ·OH radicals.[94] In another study, Skorski et al. treated wastewater by 3D printing TiO_2- ABS

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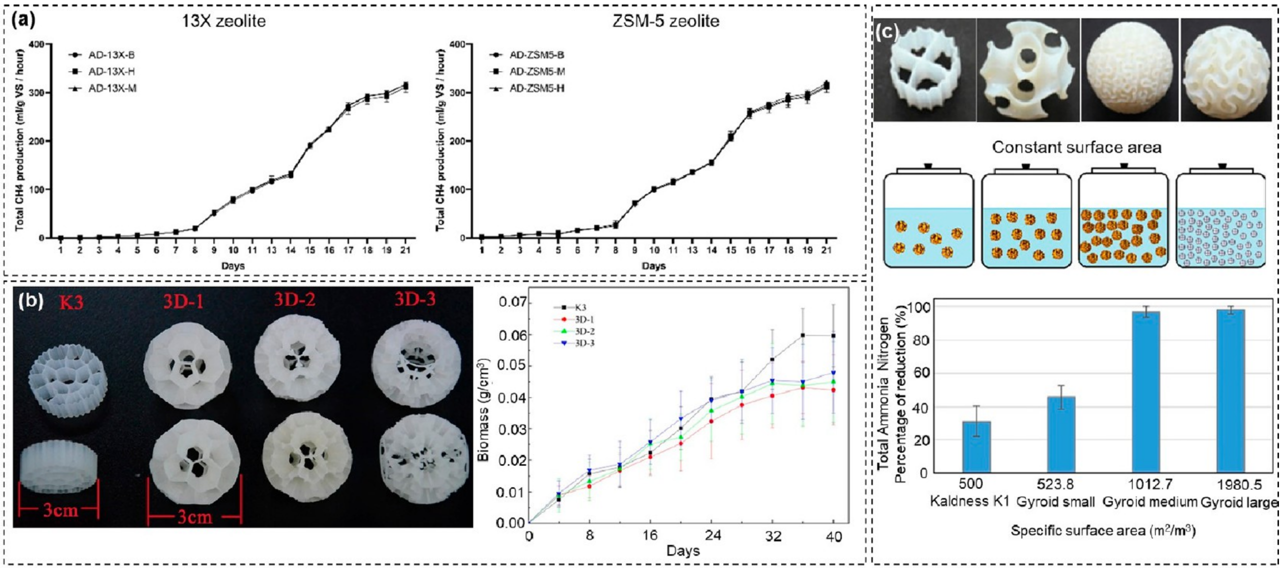

Figure 5. Development of a variety of biocarriers for the treatment of wastewater. (a) Novel biocarrier based on zeolites printed using DIW 3D printing technology and functionalized with inorganic composites. Reproduced with permission from ref 103. Copyright 2022 MDPI. (b) Fullerene-type nylon-based biocarrier fabricated with the SLS 3D printing technique with increased hydrophilicity and mechanical strength. Reproduced with permission from ref 104. Copyright 2015 Nature. (c) Polyjet-printed gyroid biocarriers in spherical morphology with different surface areas for nitrification processes. Reproduced with permission from ref 106. Copyright 2020 PLoS One.

nanocomposites in dogbone and cylinder shapes.[94] Interestingly, a transparent titanium-based photoresist was also fabricated by employing a stereolithography-based 3D printer by Greer and co-workers for household water disinfection under solar light (Figure 4b).[95] This layer-by-layer assembly was prepared by sequentially exposing the first layer to UV for 14 s, then the next four for 9 s, and the last layer for 3.5 s. The authors suggested that the mechanical strengths of 3D-printed titanium structures were several times higher than those of the commonly used titanium foams, which ensures that the photocatalyst is not leaked into the treated water. Even though the aforementioned photocatalytic systems obtained satisfactory results, most of the photocatalysts do not float, which limits their practical application.[95] It is noteworthy that floating photocatalyst, which has a relatively lower density than water, is crucial for oxidation processes in water, because it can enhance the availability of photocatalytic surface-to-light irradiation, resulting in the effective removal of surface pollutants. As shown in Figure 4c, a buoyant pickering photocatalyst carrier was created by Anusuyadevi et al. using green cellulose nanofibers (CNFs) packed with graphitic carbon nitride (g-C_3N_4).[96] To create wet-stable nanocomposite foam, researchers synthesized g-C_3N_4 by thermally polymerizing urea at 600 °C. This was followed by the insertion of g-C_3N_4 in CNFs. Rhodamine was employed as a model contaminant for the photocatalytic investigation, and the findings revealed that nanocomposite foam successfully removed dye.[97] In another study, Darkhosh et al. developed floating photocatalysts from expanded perlite (Ep) containing delafossite ($CuFeO_2$) as a semiconductor photocatalyst. The one-pot solvothermal approach was used to produce the photocatalyst $CuFeO_2$@Ep. Under visible light irradiation, a nanocomposite photocatalyst demonstrated 99% elimination of methylene blue (MB).[97] Similarly, Khan et al. produced a floating photocatalyst from Ep-doped with cadmium sulfide (CdS) via liquid phase deposition. Rhodamine was degraded by the nanocomposite catalyst by 70% when exposed to visible light.[98] Additionally, Kennedy et al. developed a composite photocatalyst for the elimination of the microcystin (MC) algal toxin by immobilizing photocatalytic TiO_2 in a biocompatible thermoplastic polymer, PLA, as shown in Figure 4d.[99] Using fused filament fabrication (FFF), the PLA-TiO_2 composite was further 3D printed into lattice and disc designs. The starting bed temperature was set at 70 °C, and the extrusion temperature was set at 215 °C with an infill density of 34%. Over 24 h, a photocatalytic degradation study revealed decreased MC content at low pH.[100] In another study, de Vidales et al. utilized the FDM-based 3D printer (extruder temperature: 200 °C, printing process speed: 1000 mm min−1) for printing TiO_2 anatase phase on low-density polyethylene (LDPE) substrates for efficient removal of CECs from wastewater.[100] To improve the catalytic activity, TiO_2 filaments were printed as meshes to increase the surface area of the catalyst compared to the traditional plate-based TiO_2 photocatalyst. After 30 min of UV exposure, the as-printed floating photocatalyst degraded MB and ofloxacin by 2% and 0.8%, respectively, significantly higher than the traditionally available photocatalyst.[101] Although the aforementioned studies highlight the potential of using 3D printers for developing photocatalysts, they are sometimes challenged by the issues related to the substrate polymers, such as low thermal stability, low surface areas, and the requirement for photosensitive materials. Hence, in the future, it is suggested that photoactive materials can be directly loaded onto the 3D-printed substrates.

3.3. Biocarriers

Recently, bioreactors have been extensively employed for wastewater treatment for degrading organic pollutants, including phenols, hydrocarbons, pesticides, and medication residues. One of the key constituents of bioreactors is the biocarriers which are porous materials that can provide a surface for microorganisms to grow and form biofilms, thereby enhancing the rate of pollutant degradation.[102] The mechanism of action of biocarriers in wastewater treatment involves

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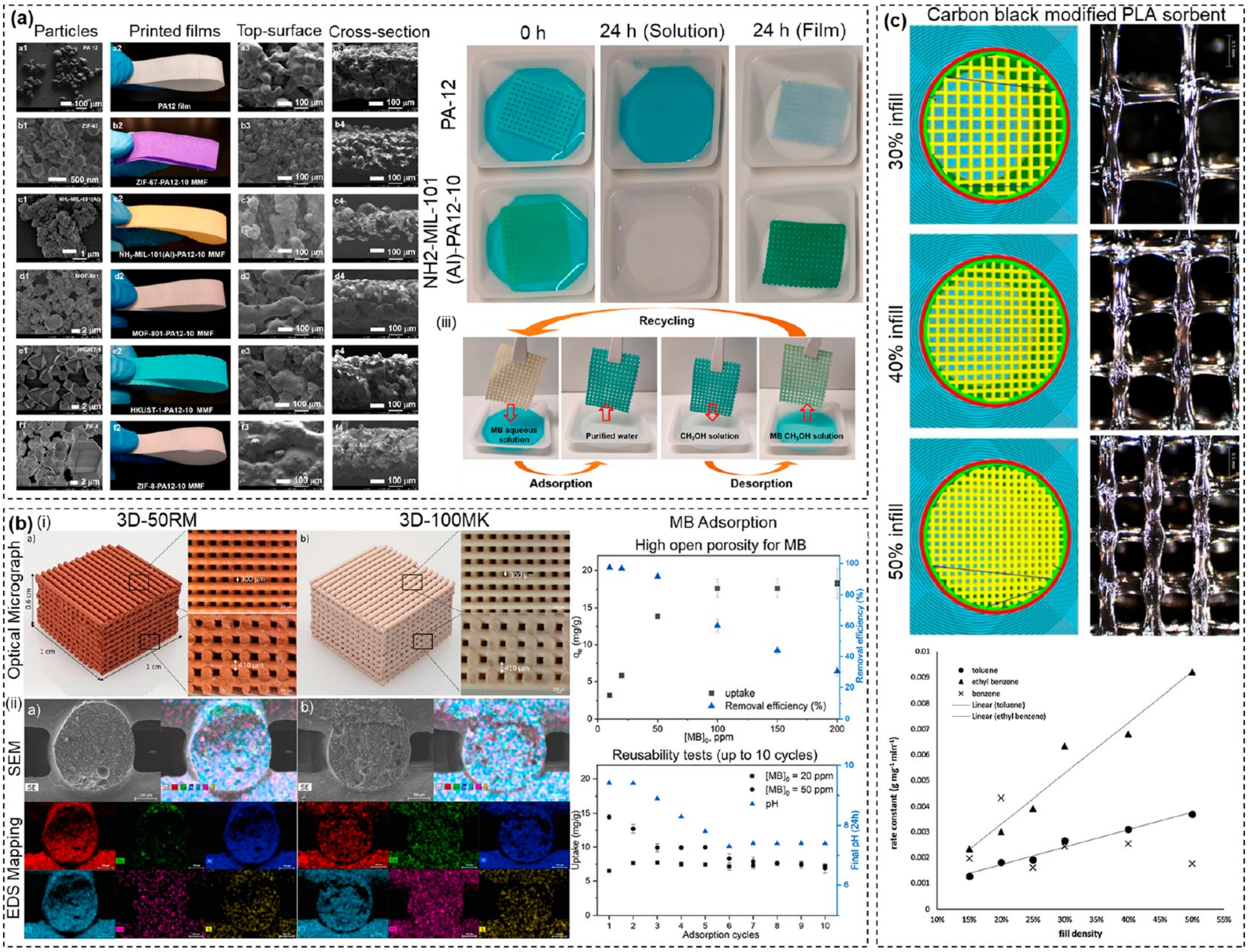

Figure 6. Manufacturing various sorbents using 3D printing technology to soak contaminants from wastewater. (a) Different MOF-based particles embedded in polymer matrices were prepared using the SLS technique for MB degradation. Reproduced with permission from ref 108. Copyright 2019 American Chemical Society. (b) 3D-printed geometric structures of red mud using DIW for adsorbing MB. Reproduced with permission from ref 110. Copyright 2023 Elsevier. (c) PLA sorbents designed by 3D printed in cylindrical structures for removing organic contaminants from wastewater. Reproduced with permission from ref 111. Copyright 2020 Elsevier.

the attachment of microorganisms to the surface of the carrier, followed by the formation of a biofilm, which provides a protective layer for microorganisms, allowing them to degrade pollutants[103] more efficiently, even under adverse conditions such as high pollutant concentrations, low dissolved oxygen levels, and fluctuations in pH and temperature. Moreover, biocarriers have a density relatively lower than that of water, which allows them to circulate effectively within the bioreactor system. An ideal biocarrier should have a larger surface area, high durability, high porosity, and high surface roughness and should allow for efficient mass transfer. Traditionally, manufacturing techniques such as cutting and molding are used to develop biocarriers based on polymeric materials such as polyethylene and PP. However, their utilization is limited due to their ability to generate only limited structures and designs. On the other hand, the inherent prospects of 3D printing technologies to produce biocarriers with better mechanical strength and durability are making them popular choices for biocarrier fabrication. Recently, Chioti et al. synthesized a novel biocarrier (24 × 14 × 7 mm) based on zeolites (13X and ZSM5) by using the DIW-based 3D-printing approach, as shown in Figure 5a. The zeolites were mixed with various inorganic composite materials such as bentonite, montmorillonite, or halloysite nanotubes, along with an organic binder, forming a resultant ceramic printing paste. The results revealed that compared to the commercially available carriers, the as-printed carriers exhibited relatively low methane production, high chemical oxygen demand, P (phosphorus), N (nitrogen), NO_2, and NO_3 reduction, and reduced biofilm formation.[103] In another study, Dong et al. designed a fullerene-type nylon-based biocarrier with the help of the SLS 3D printing technique (Figure 5b). Compared to the commercial polyethylene K3 biocarriers, the 3D-printed nylon biocarriers exhibited higher surface roughness (70 μm), mechanical strength (tensile: 30 MPa, bending: 20 MPa and impact: 3 kJ/m^2), and hydrophilicity.[104] Moreover, the biocarriers were suspended in the bioreactors, which allowed them to move freely in the presence of flowing water, thus enabling increased mass transfer. These remarkable surface characteristics, along with the specialized hollow structures obtained via 3D printing, resulted in higher microbial activity, which was 60% higher than commercial carriers.[104] Beyond the traditional bioreactors, 3D printing approaches have also been employed to design the carriers for moving bed biofilm reactors (MBBRs), which are widely used for wastewater treatment due to their simple operation and high efficiency.

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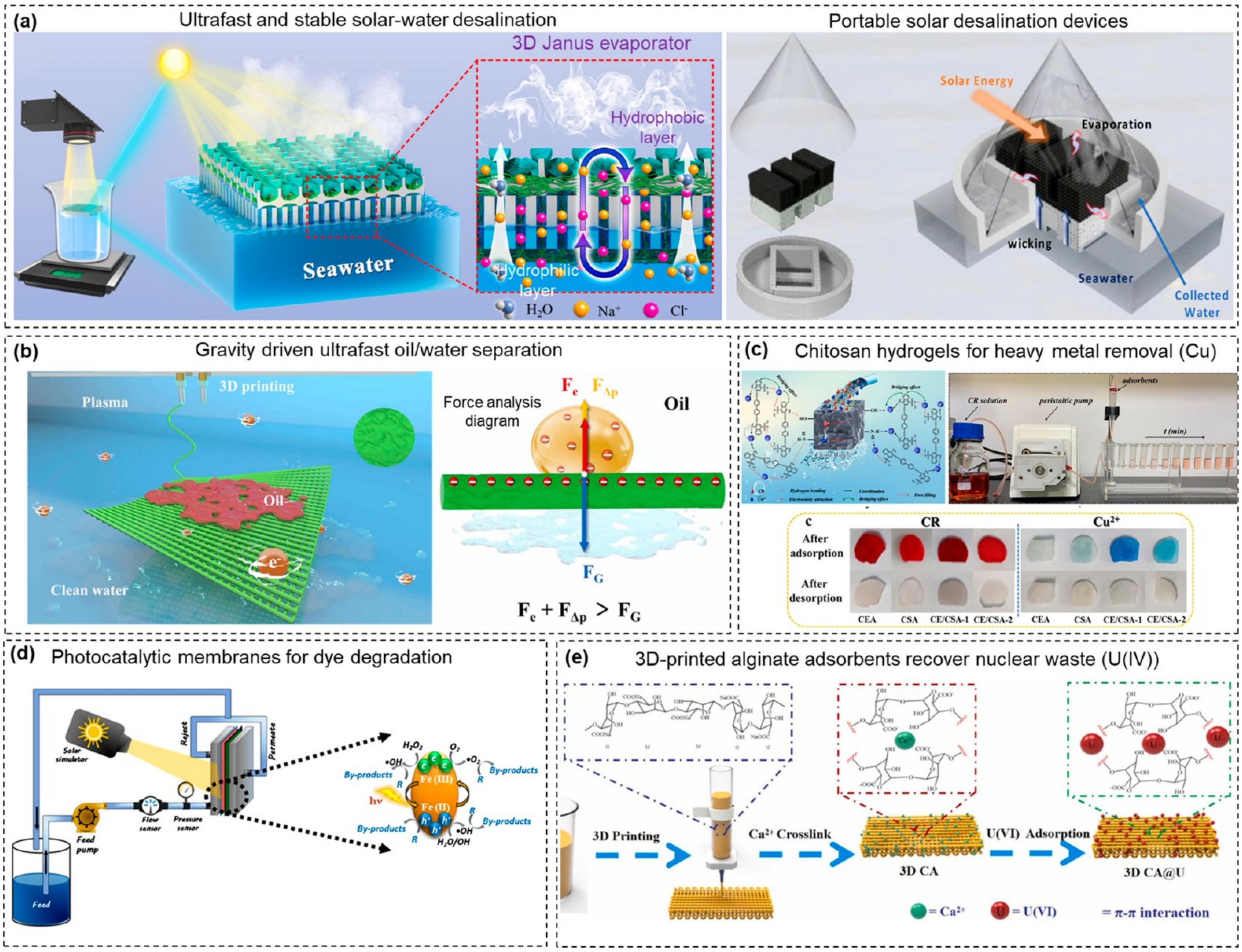

Figure 7. Application of 3D-printed membrane modules in wastewater treatment. (a) Solar-energy-activated desalination devices manufactured using additive manufacturing for ultrafast seawater purification. Reproduced with permission from ref 112. Copyright 2023 Elsevier. (b) 3D-printed plasma-modified ABS membrane for oil−water separations. Reproduced with permission from ref 113. Copyright 2023 Elsevier. (c) 3D-printed porous chitosan hydrogel-based membranes for the adsorption of heavy metal ions. Reproduced with permission from ref 114. Copyright 2023 Elsevier. (d) Photocatalytic feed spacers with nanorod morphologies for degradation of dyes. Reproduced with permission from ref 115. Copyright 2022 Elsevier. (E) Illustration showing the printing route and mechanism of the 3D printed CA for U(VI) absorption. Reproduced with permission from ref 116. Copyright 2022 Elsevier.

For MBBR, it is extremely important to have an optimized design for the biocarrier that would enable the attachment of the bacteria to the maximum possible surface area for receiving good exposure to nutrients, resulting in highly stable biofilm formation and, in turn, influencing the overall performance of the reactors. To fabricate biocarrier media with complicated designs for MBBR, Elliot et al. employed polyjet 3D printing to develop gyroid-shaped carrier media in spherical morphology with a than that of the commercial carrier (baseline K1 Kaldnes).[105] Consequently, Proano-Pena et al. also developed the 3D-printed gyroid-shaped biocarriers with different specific surface areas ($\sim 21.9 \times 10^{-4}$–82.9×10^{-4} m^2) for nitrification processes for wastewater treatment.[106] The results indicated that biocarriers with large and medium surface areas showed the highest nitrate production and performed better than the commercially available carrier (baseline K1 Kaldnes). The best-performing design for constant ratio was found to be a large SSA gyroid with an efficiency of ammonia conversion of 99.33%, for constant total surface area was medium SSA gyroid with an efficiency of 94.74%, and for constant biocarrier media count was also a large SSA gyroid with an efficiency of ammonia conversion of 92.73% after an incubation of 8 h (Figure 5c).[106] The aforementioned studies signify the role of 3D printing technologies as novel tools for developing intricately structured biocarriers with increased surface areas that can activate the diverse microbial community for wastewater treatments.

3.4. Sorbents

Adsorption of organic contaminants such as ammonia, heavy metals, and volatile organic contaminants (VOCs) by adsorbents has proven highly efficient for wastewater treatment.[107] Such adsorption processes utilize a sorbent that can selectively adsorb molecules or ions from a liquid or gas phase for its removal and mainly determine the adsorption efficiency. Owing to their economic feasibility and easy availability, natural materials, including carbon-based materials and industrial wastes, are widely used as absorbents, typically prepared using coprecipitation, thermal decomposition, microwave irradiation, chemical reduction, microemulsion, and arc discharge.[107] However, most of the developed adsorbents are mechanically unstable and possess low flexibility, and their separation processes are extremely complex. On the other hand, 3D printing technologies have turned out to be excellent alternatives for fabricating structured sorbents with high mechanical strength, controllable porosity, high stability, outstanding efficiency, and the ability for mass production. Until now, different configurations of packed bed spherical sorbents such as porous, monolithic, cylindrical, open, and periodic inner structures have been reported to be fabricated by using 3D printing approaches. Among all materials, MOFs are extensively prepared by 3D printing for adsorption in wastewater treatment, especially for removing organic dyes due to their larger surface areas, distributed metal sites, and porous crystalline structures. For instance, as shown in Figure 6a, Li et al. 3D printed the MOF-polymer matrices with thermoplastic PA12 powder as a matrix substrate using the SLS technique to remove MB dye.[108] Among all MOFs, NH_2-MIL-101(Al) crystals with the smallest pore size displayed the highest flexibility, maximum MB adsorption rate (152 mg/g), and good recyclability (~81.3%) after being stirred at 800 rpm for 30 min in the absorption experiments. To further improve the flexibility of the MOF-based sorbents, ABS-based polymeric substrates were printed by using 3D printing and coated with Cu-benzene tricarboxylic acid MOFs for MB removal. The as-printed MOF composite sorbents were uniform in nature with miniature hill-like morphology and displayed increased surface wettability, which contributed toward improved adsorption of dyes.[109] In another study, Liu et al. designed Cu-MOFs-based composite sorbents with PLA films as the matrix substrate layers using 3D printing to remove malachite green (MG).[109] Within 10 min, 3D printed Cu-MOF/PLA sorbents could remove 90% of the MG, observed after reusing the films five times. In another report, Gonçalves et al. used the DIW-based printing technique for preparing red mud-metakaolin with different geometrical structures (parallelepipeds and cylindrical) by manipulating the nozzle diameter and extrusion speed, as shown in Figure 6b.[110] Each structure consisted of 20 layers, with each filament inclined 90° to the previous ones. The as-designed sorbents showed about 99% dye-removing efficiency after 72 h of immersing the printed structures in MB solutions. Moreover, few studies have suggested that 3D-printed PLA sorbents with specialized monolithic structures have demonstrated enhanced adsorption performance for dye removal.[111] Further, 3D printing approaches have also been utilized to develop cylindrical sorbents, since they can control the dimensions, orientations, and geometries of sorbents in porous beds. For instance, Lagalante et al. designed cylindrical and conductive PLA sorbents using 3D printing (nozzle diameter: 0.4 mm and extrusion temperature: 215 °C) for removing VOCs, such as benzene, toluene, and ethylbenzene from wastewater (Figure 6c).[111] The authors anticipated that the infill density could be directly correlated with the removal performance, with the highest removal of VOCs (benzene: 50.6%, toluene: 81.3%, and ethylbenzene: 92%) obtained at 50% infill density after 5 h of treatment due to the increase in the effective surface area.[112]

4. APPLICATION IN WASTEWATER TREATMENT

3D printing technologies present tremendous potential in fabricating unique structures with intricate geometries, which manifest different developments in water treatment. Owing to their rapid fabrication rates, cost-effectiveness, and simplicity of the process steps, 3D printing-based techniques have surpassed conventional water treatment approaches. Until now, 3D-printed membranes and their modules, adsorbents, and biocarriers have been widely explored for water treatment applications, including desalination, oil–water separation, water filtration, and heavy metal and organic contaminants removal as well as nuclear decontamination. Particularly for water desalination, 3D printing strategies offer advantages over conventional manufacturing techniques as they can be used to fabricate not only whole membrane systems and module components but also energy management components such as solar collectors and evaporators (Figure 7a).[112] Out of all 3D printing technologies, DLP is the most widely used approach for thermal and membrane desalination because of its excellent flexibility, high accuracy, and diverse material compatibility. To obtain structures with specific functionality for water desalination, material-jetting-based polyjet printing has also been employed to deposit functional materials on the surface of membranes. In addition, material extrusion-based 3D printing approaches have been reported to enhance the heat transfer ability of membranes, which is vital for water desalination. Nonetheless, most of the reported studies have focused on utilizing 3D printing for designing spacers for desalination rather than whole membranes. These printed spacers generate higher shear stress for fluid flow, enhancing the flux and reducing fouling during desalination. In addition, due to the recurrent disposal of industrial pollutants into water bodies, oil–water separation techniques are becoming increasingly critical for mitigating water pollution. For carrying out the oil–water segregations, porous membranes fabricated by conventional thermal and vapor-induced separation approaches lack selectivity and permeability and use large volumes of solvents. Therefore, using 3D printing approaches for oil–water separations has gained tremendous attention due to their superior characteristics such as cost-effectiveness, design flexibility, solvent-free process, lower energy consumption, etc. Most of the studies have utilized 3D printing techniques to design superhydrophobic and superoleophilic micro/nano hierarchical structures with excellent wettability for oil–water separation membranes to reduce membrane fouling and increase the lifetime of membranes (Figure 7b).[113] To further mitigate oil-spill accidents, specialized 3D-printed porous membranes and meshes are also developed, which serve as replacements for traditional oil skimmers and sorbent materials relying on their ability to remove oil and thereby reduce the overall operational cost-effectively. The increasing toxicity concerns related to heavy metals, including mercury, lead, and cadmium contamination in wastewater, have also driven the utilization of advanced manufacturing techniques such as 3D printing for designing complex hydrogels and sorbents with different shapes and sizes for removing toxic metals from the wastewater. Importantly, 3D printing enables the design of unique adsorbents with larger specific surface areas for adsorption processes, which is highly crucial for heavy metal removal. Efforts are also being dedicated to utilizing 3D printing for fabricating bioinspired "green" sorbents for heavy metal removal due to their low-cost, nontoxicity, and biodegradation. One such example is the class of chitosan-based sorbents, which possess a larger surface area, porous structure, and high reusability when developed using 3D printing to remove heavy metal ions, such as Hg^{2+}, Cd^{2+}, Pb^{2+}, and others. Subsequently, chitosan-based hydrogels with larger surface areas were also prepared using extrusion-based 3D printing approaches in a solvent-less manner for efficient adsorption (Figure 7c).[114] The 3D-printed chitosan hydrogels also displayed the potential to respond to external stimuli with

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abrupt change in volume changes abruptly. To further improve the adsorption efficiency, 3D-printed materials were functionalized with heavy metal binding sites by chemical or nanostructural modifications for specific metal ion attachment for its removal. Not only heavy metals but also the advantage of 3D printing to generate structures with tunable surface area, porosity, and roughness have been shown to improve the removal efficiency of complex organic dyes, such as malachite green, MB, rhodamine dye, and other dyes, as compared to adsorbents prepared by conventional techniques. Functional adsorbents have been developed by 3D printing technologies for dye removal, which generate flexible and porous structures with high surface areas for the highly efficient adsorption of organic dye molecules. Of all materials, 3D printing has been widely used to design MOF-based structures for dye degradation owing to its intrinsic larger surface area, porous structures, open channels, and different surface functional groups. To improve the mechanical stability of sorbents for long-term dye removal applications, adsorbents are activated by coating with modifiers, including biological molecules, CNTs, etc. Apart from sorbents, 3D printing has recently been used to design photocatalysts for dye removal due to their high efficacy and cost-effectiveness (Figure 7d).[115] Typically, photoreactors have been 3D printed and functionalized photocatalysts such as graphitic carbon nitride (GCN) and ZnO for the degradation of dye molecules, which displayed high degradation efficiency and recyclability. The remarkable ability of 3D printing in designing complex geometrical structures has also paved the way for treating highly dangerous radioactive or nuclear waste, such as cesium-137, strontium-90, and tritium, in wastewater (Figure 7e).[116] 3D-printed materials were used as active supports and were coated with functional materials such as MOFs and zeolites for removing nuclear waste in the long run with high efficiency.[117]

5. SUSTAINABILITY, PROCESS ECONOMICS, AND SCALE-UP POTENTIAL

3D printing is a highly sustainable manufacturing technique that supports sustainability by reducing waste generation, energy requirements, and carbon emissions. 3D printing offers the possibility of designing membranes and their modules with a minimal amount of chemicals and recycling the same materials for consecutive cycles of production. Importantly, 3D printing utilizes the layer manufacturing principle rather than cutting parts from larger sections usually done by conventional techniques, which reduces waste.[1] 3D printing does not generate scrapes and material loss due to defects since the principle is based on AM.[1,2] The materials used for printing also influence the sustainability of the process. For instance, natural materials such as cellulose, algae, etc. are being used as 3D printing materials for more sustainable production. Moreover, the process of 3D printing is rapid and direct which reduces the overall energy demands and CO_2 emissions.[3] In order to reduce carbon emissions, one of the novel 3D printers, i.e., Eco Printing is proposed for water treatment which uses waste polymers as printing materials and a solar charging battery for power. The ability to perform on-site printing also reduces the carbon emissions associated with shipping because consumers and manufacturers can produce the materials from digital files. From the aforementioned examples, it can be validated that 3D printing would be a big step toward a circular economy due to its potential to manufacture with negligible waste and unnecessary use of added chemicals

Since the utilization of 3D printing technologies for wastewater treatment still remains in the initial stage, its scalability for practical applications is one of the important aspects to be explored. However, the scale-up potential of 3D printing might be impeded due to a number of limitations such as cost of production, the rapidness of the process, difficulty in mass production, and maintaining the same functionalities at a large scale.[50,55,118] For instance, the investments in 3D printers and the complicated postprocessing steps especially with polymer-based techniques contribute significantly to the overall production cost for wastewater treatment applications.[118] Another major challenge for commercialization is to print flat larger sheets (10×5 m^2) which is difficult to achieve with the available 3D printers given their printing width (1 m).[12] In addition, the software used by 3D printers requires high levels of expertise to print, thereby obstructing its scale-up for practical applications. To address these challenges, in recent years, the adaptation of large-sized and powerful printers with vast types of feedstock materials and integration with surface functionalization platforms such as polymerization, etching, wettability, etc. have shown promising advantages for wastewater treatment at industrial scales.[61,119] The overall production and operational costs associated with transportation, maintenance, and supply chains could also be reduced by printing membranes on-site. To print larger structures in order to meet industrial demands, printers with multiple nozzles are being employed to produce ceramic membranes for water treatment.[120] In addition, researchers are also working toward developing user-friendly software that can be universally interfaced with all 3D printers for standardizing the production process.[112,118] Based on the aforementioned considerations, it can be anticipated that 3D printing technologies are getting ready for industrial applications but mostly for generating parts that are otherwise highly expensive to prepare using other manufacturing techniques. For complete adaptations, serial productions and 3D printing farms would be a stepping-stone in the manufacturing process; however, the process economics and complexities will play a major role in determining their success.

6. CHALLENGES AND FUTURE PERSPECTIVES

Several challenges must be resolved to enable more widespread adoption of the 3D printing technology for large-scale production. One of the most fundamental challenges in 3D printing is the resolution, which depends on many variables, including nozzle diameter, printer type, material utilized, and, most crucially, printing technique.[9] Defects such as voids or gaps between consecutive layers of material during the printing process are another significant drawback of 3D printing. The resultant product might become more porous, which could negatively impact its strength and durability because of poor interfacial adhesion between printed layers.[121] The layer-by-layer printing approach used in AM introduces variations in the material's microstructure within each layer and at the interfaces between layers, impacting the mechanical properties of printed parts.[122] Additionally, 3D printing employs CAD software to build intricate digital models. However, due to errors made during the tessellation process, printed objects may not reproduce as expected in the digital model.[123] Another issue is that 3D printing is sometimes less economical than conventional manufacturing methods due to its

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significantly slower printing speed and higher production costs.[124] Collectively, industrial scale-up for 3D printing remains challenging. This constraint has prompted research into combining 3D printing with other methods to develop a more effective and economical production process.

A promising method for improving the performance of 3D-printed materials in water treatment applications is surface functionalization, which involves modification of the surface characteristics of the printed material, such as surface charge, hydrophilicity, and affinity toward certain contaminants, to increase adsorption, catalysis, and separation of water pollutants.[125] In this regard, integrating 3D printing with additional manufacturing procedures to create an integrated multiprocess system is one potential future approach to combat the current constraints of the technology and create more intricate and sophisticated structures by merging 3D printing with other production techniques[125] such as atomic layer deposition (ALD), stretching, vapor phase deposition (VPD), track-etching, and electrospinning. For example, Kozior et al. fabricated a 3D-printed composite filter from PLA and thermoplastic polyurethane (TPU) filaflex, which was further coated with polyacrylonitrile (PAN) nanofibers by electrospinning.[125] Results indicated electrospun PAN nanofibers had strong adhesivity when deposited on a TPU-based scaffold, resulting in a mechanically stable composite filter with a nanofibrous surface.[126] Another significant benefit of post-treatment or surface functionalization, from an industrial perspective, is the ability to achieve the desired surface properties. This process enhances the performance of the overall printed material, providing additional advantages. One such promising approach is to use various deposition methods, such as ALD to functionalize the 3D-printed surface by incorporating nano- or microscale topographies.[127] It is a cutting-edge deposition process that uses successive, self-limiting surface reactions to produce highly conformal, pinhole-free thin films on substrates created by 3D printing. Additionally, it enables change of membrane attributes, including film thickness, surface charge, pore size, and chemical composition, enhancing the membrane's capacity for separation. Ji et al. fabricated polyoxometalate anions (POM)-doped 3D-printed ABS-based highly porous adsorbent for transition metal removal.[127] In this study, POM was utilized as a heavy metal binding site, which was immobilized on the ABS surface by strong hydrogen bonding, thus improving the hydrophilicity and overall adsorption potential of the printed adsorbent. Heavy metal adsorption studies indicated that POM-modified adsorbent {PW9}@ABS-15 showed remarkably higher heavy metal removal efficiency (100%) as compared to pure ABS-5 (~8.8%) and ABS-15 (~12.4%).[90]

The evolution of 3D printing has accelerated research toward the incorporation of a new dimension, "time", which has led to the emergence of a novel technology known as 4D printing, where the 3D printed structures evolve in terms of shape, property, and functionality with respect to time, upon providing an external stimulus like heat, light, pH, magnetic field and water.[128] This provides a strong advantage over the traditional 3D printing process, where printed objects remain static, whereas 4D printing allows self-assembly and shape transformations of the printed objects, making it a promising approach toward water treatment applications. However, 4D printing technologies are relatively unexplored, often require the use of high-cost printers, and are limited by the range of printed materials that can undergo subsequent changes in morphology and/or properties, which limits their applicability. Design-encoded fabrication is essential to harnessing the potential of 4D printing, which can further add to the complexity and cost of fabrication. Another major drawback of 4D printing is its inability to fine-tune the thermomechanical properties other than what is possible with available commercial resins. Thus, more work is warranted to establish the utility of 4D printed materials for water filtration.

7. CONCLUSIONS

The technology of AM has undergone remarkable advancements in recent years, supported by increased funding and extensive research and development activities worldwide. These factors propelled the transition from traditional manufacturing methods to the widespread adoption of 3D printing for a plethora of applications. This Perspective highlights the recent advancements in 3D printing for various wastewater treatment applications, which is one of the most explored fields. Owing to its numerous advantages such as design freedom, flexibility, customization, personalization, fabrication, reduced material waste, on-demand production, quicker prototyping, and the capacity to print intricate structures with a higher resolution, 3D printing has emerged as a promising platform for printing membranes and membrane modules such as spacers, biocarriers, and sorbents for wastewater treatment. The as-developed 3D printed materials have been widely used for water desalination, oil—water separation, water filtration, separation processes, and heavy metal removal. Compared to the conventional water treatment techniques, 3D printing-based approaches decreased membrane fouling, enhanced the degradation efficacy for heavy metals, and resulted in higher removal efficiency, thereby bridging the existing gaps. In addition, 3D printing also enabled the fabrication of reusable catalysts and adsorbents, which reduced energy demands and made the manufacturing process more environmentally friendly. Overall, the novel field of 3D printing is a major stepping stone toward obtaining sustainable manufacturing solutions in wastewater treatments.

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Notes

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