

The implications of 3D-printed membranes for water and wastewater treatment and resource recovery

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Abstract

It is widely acknowledged that three-dimensional (3D) printing or additive manufacturing will revolutionize many industries. However, the broad implications of 3D printing on water treatment membranes are not appreciated. 3D printing will transform the traditional membrane fabrication methods, reducing costs and industrial waste from manufacturing processes, with substantial benefits to treatment performance. In particular, 3D printing provides a high potential for radical decentralization. Remote communities, hospitality resorts, military bases, and oil and gas extraction operations could significantly benefit from the on-site fabrication of membrane units tunable to their specific wastewater challenges. Acute drinking water contamination events, like those associated with toxic by-products from algal blooms, chemical spills, forest fires, and extreme weather, cause adverse health effects on humans and shutdowns of piped water supply. 3D printing of customized membranes provides an opportunity for a fast response to such disastrous events. These membranes could be ready for installation within hours, with the vendor's role more akin to a software company installing a patch than the traditional approach, with major considerations for hardware availability, timeline, and supply chain. Despite these clear and potentially transformative advantages, 3D printing of membranes is not a panacea; countless aspects need to be taken into consideration for the successful implementation of this emerging technology. The full deployment of 3D-printed membranes in water and wastewater treatment can be achieved by extending the variety of printable materials, improving the speed and resolution of printing, creating nanoscale pores, reducing the fabrication costs, and improving the mechanical properties of the resulting membranes.

KEYWORDS

3D printing, additive manufacturing, decentralized wastewater treatment, drinking water contamination, membranes fabrication

[Correction added on 2 August 2022, after first online publication: Middle initial of João B. P. Soares was missing in the original published article and has been added in this version.]

1 | INTRODUCTION

With the world's population growth, change in lifestyle, and industrialization, water resources are severely contaminated, causing serious problems for public health and the environment.^[1] Membrane separation is one of the fastest emerging water treatment technologies due to its high removal capability and energy efficiency. However, some disadvantages of conventional membranes, including unreliability, high costs, complex fabrication procedures, and imprecise membrane structure, have restricted the widespread application of membrane technology.^[2] Therefore, new fabrication processes are needed to mitigate these challenges and optimize membrane structure. Three-dimensional (3D) printing (additive manufacturing) is a layer-by-layer fabrication method used to fabricate objects with geometrical restrictions and complicated designs.^[3] Recently, 3D printing has been employed in membrane separation technology to make membrane materials and membrane element parts. The studied 3D-printed membranes perform exemplarily in water and wastewater treatment and possess remarkable advantages compared to conventional membranes.^[4–7] 3D printing technology allows the fabrication of membranes with complicated designs in a single-step process. The ability to precisely control the membrane structure and pore geometry leads to uniformity, improved filtration, and cost reduction.^[8] In addition, 3D printing fabrication techniques are environmentally friendly with the minimum use of solvents.^[9,10] Despite the remarkable reduction in solvent consumption compared to conventional methods, solvent baths are sometimes used during the fabrication process to remove the supports or improve the surface quality. Besides, alcohols and propylene carbonate may be used to remove residual resins or polish the pores after the printing process.^[11,12]

So far, most of the developments in this field have been on the fabrication of the module parts, including spacers,^[13–17] static mixers,^[18,19] and spinnerets.^[20] In the last 10 years, membrane printing using ceramic and polymeric compounds has been increasingly studied.^[21–23] However, the use of 3D-printed membranes has just recently emerged, and its applications are restricted to only a few contaminations, such as oil/water separation. However, the fabrication of 3D-printed membranes is a cutting-edge technology that has only recently emerged. The application of current 3D-printed membranes is restricted to the separation of a few contaminations, primarily oil, from wastewater, most likely due to technical difficulties in making nano/angstrom selective pores/voids present in ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) membranes. Also, upscaling and mass production of membranes by 3D printing, while

being a hot topic research area, is still time-consuming and costly.^[24–26] Moreover, the 3D printing of some widely utilized materials in conventional membranes like polyvinylidene fluoride (PVDF) and polyethersulfone (PES) is not yet feasible.^[3,27,28] Figure 1 shows the schematic timeline of the evolution of 3D printing, the application of 3D printing in membrane technology, and the number of publications by searching for keywords '3D printing', 'water', and 'membrane' in the last decade. The data shows more than 160 publications in the field of 3D-printed membranes for water treatment applications in the last 10 years.

Many review papers dealing with 3D printing technology for membrane separation have focused on using 3D printing materials for membrane fabrication and modular elements. Design, fabrication protocols, and various applications of 3D-printed membranes for water treatment have been well studied in several recent publications.^[3,29–32] The scope of some reviews has even become more limited to one particular application, such as desalination.^[9,33] The environmental friendliness of 3D printing as an alternative method to current membrane fabrication techniques was studied as well.^[34] In this review paper, we emphasize the recent progress of 3D printing technology for membrane separation, focusing on water and wastewater decentralized systems.

The wastewater decentralization system is one of the applications that could be tremendously impacted by the introduction of 3D-printed membranes. Membrane-based decentralization is a practical approach for sustainable water management in rural and peri-urban areas. It reduces the infrastructure and operational costs and facilitates water reuse for non-drinking items.^[35] However, decentralized systems confront different problems associated with the use of conventional membranes, such as logistical challenges, slow response to varying conditions, high investments for maintenance, and regulating the system, which necessitates the use of 3D-printed membranes. The possibility of rapid on-site fabrication and tunable treatment units using 3D-printed technology has many implications for water and wastewater treatment, including decentralization, reduction of infrastructure, customized design for the contaminant of interest, rapidly dealing with varying water feed qualities and acute problems, and minimal demand for chemicals and organic solvents for fabrication.

This thematic review paper investigates recent advances of 3D-printed membranes regarding their design, fabrication methods, and applications in water and wastewater treatment systems. First, general details about 3D-printed membrane fabrication and applications of 3D-printed membranes in liquid separations are presented. Then, we provide insightful perspectives about

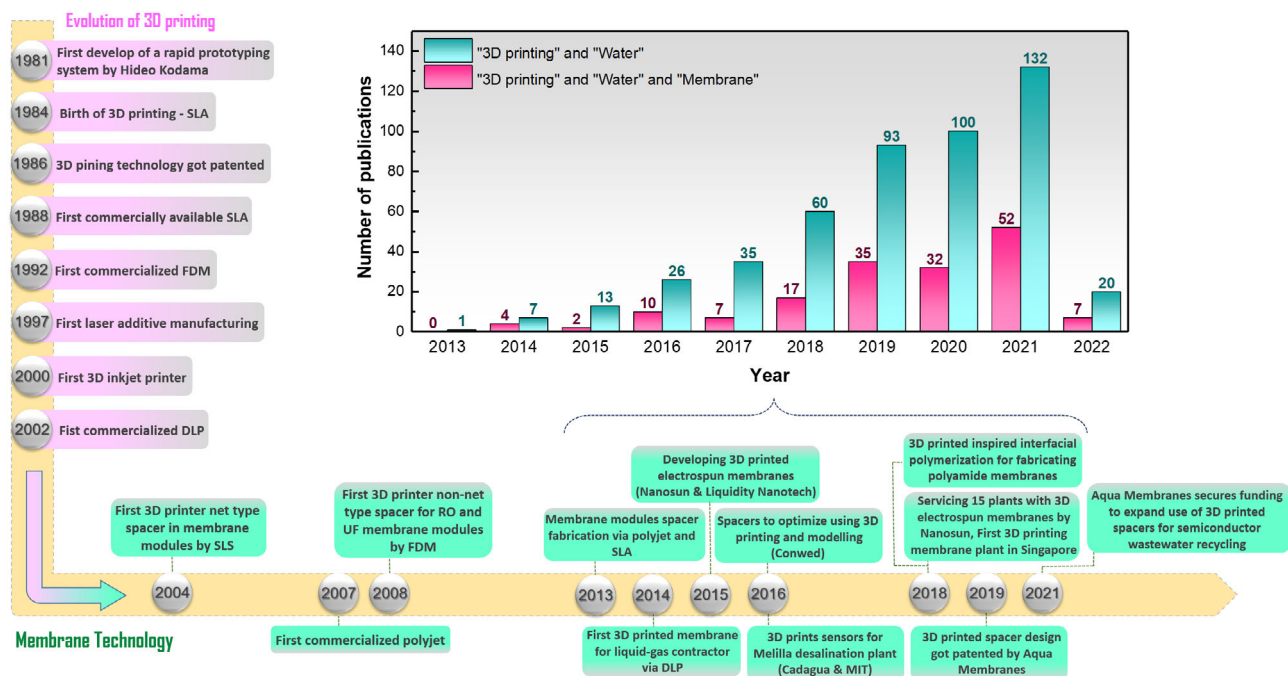


FIGURE 1 Schematic timeline of 3D printing and its application in membrane technology for water treatment. DLP, digital light processing; FDM, fused deposition modelling; RO, reverse osmosis; SLA, stereolithography; SLS, selective laser sintering; UF, ultrafiltration. (Adapted from Soo et al.^[9] and Lee et al.^[62]). The number of publications per year by searching keywords using Web of Knowledge (February 2022) is presented in a separate panel for the last 10 years. The green data bar represents the number of publications that contain '3D printing' and 'water', and the pink data bar shows the number of publications that contain '3D printing' and 'water' and 'membrane'

the implications of using 3D-printed membranes, which allow their use in decentralized systems. Finally, the trajectory of using 3D-printed membranes, as well as challenges and prospects of 3D-printed membranes at a commercial scale, is reviewed.

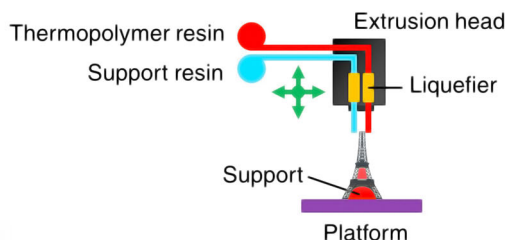
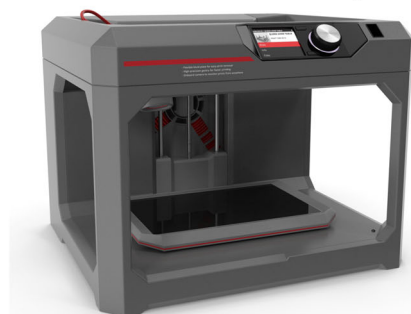
2 | OVERVIEW OF 3D-PRINTED MEMBRANES FABRICATION

3D printing is a software-based manufacturing process in which successive layers of materials are deposited on top of each other to fabricate a model.^[36] A 3D-printed structure is modelled based on a computer-aided design (CAD) and then transformed into readable formats. Subsequently, the file is sliced into countless two-dimensional (2D) layers before 3D printing modelling and fabrication.^[8] 3D printing technologies may be divided into seven categories, including material jetting,^[37] binder jetting,^[38] powder bed fusion,^[39] material extrusion,^[40] vat photopolymerization,^[41] sheet lamination,^[42] and direct energy deposition. Each of these technologies has a specific type of material input and generates parts with particular characteristics. Therefore, only a few of these technologies can be employed to make module parts and membranes.^[33] The most common methods for the

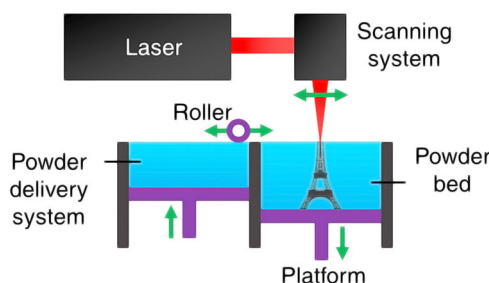
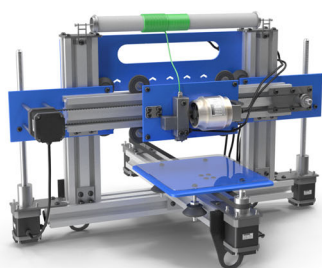
fabrication of 3D-printed membranes are fused deposition modelling (FDM), selective laser sintering (SLS), stereolithography (SLA), and inkjet printing (Figure 2).^[3]

FDM—resolution of 50–200 μm ^[43]—is a material extrusion-based method that is compatible with a wide range of polymers such as polycarbonate (PC), nylon, polylactic acid (PLA), and acrylonitrile butadiene styrene (ABS), among others.^[44] The thermoplastic polymers must be used as filaments with a low enough molten viscosity to be extruded. In FDM, thermoplastic filaments are heated above the glass transition temperature (T_g) and then extruded through a nozzle on the platform or on top of previously printed layers in the X-Y plane.^[45] Finally, the filaments solidify at room temperature to create the 3D-printed object.^[36] However, the FDM-fabricated objects have low mechanical strength due to the anisotropic properties caused by layer-by-layer deposition.^[46,47] In this method, composite materials must be used in a filament form. The resins could be used as the matrix in the reinforcement composition to enhance the mechanical properties.^[48] After printing, filling the voids with high-strength resins could also be effective in enhancing the mechanical strength.^[49] The morphological properties of the membranes fabricated by the FDM may be controlled by regulating the infill setup, the filament width, and the spacing between adjacent

Fused Deposition Modelling Printing (FDM)



Selective Laser Sintering Printing (SLS)



Stereolithography Printing (SLA)

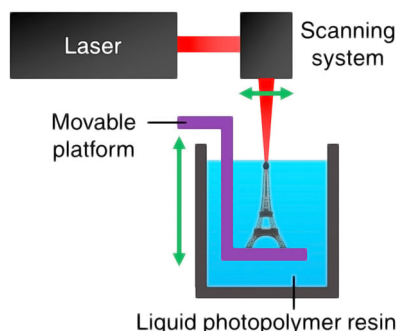


FIGURE 2 The schematic illustration of fused deposition modelling (FDM), selective laser sintering (SLS), and stereolithography (SLA) 3D printers (Adapted from Low et al.^[30]).

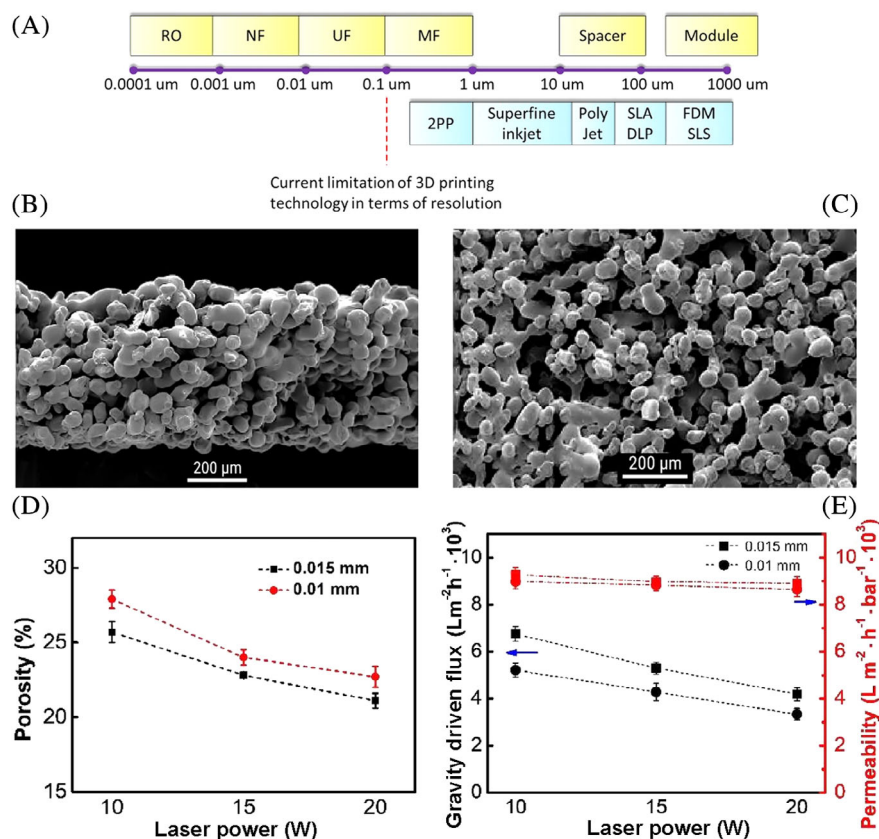
filaments.^[45] In addition, the presence of cavities or air pockets on the membrane surface due to heterogeneity between the layers and filament strands is a common issue when using FDM printing.^[50]

SLS—resolution of 80–250 μm ^[36]—is another technology available for fabricating 3D-printed membranes.^[51] SLS is a powder bed fusion-based technology in which a thin layer of powders is spread on a platform. A laser beam with a predefined direction scans the powders in each layer and sinters them by heating. Subsequently, the powders are fused together to form a pre-designed 2D shape. A roller is used to place the new layers of fresh powders on top of the previously sintered layers. The procedure is repeated for the new layers to complete the 3D model.^[36,52] The SLS technology is compatible with various materials, including polymers, ceramics, and alloys.^[53,54] The

morphology of the fabricated membranes using SLS mainly depends on the count of laser scanning, laser power, and hatch space. Denser membranes with smaller pore sizes could be fabricated by increasing the laser power or the number of scans and reducing the hatch space. These parameters affect the absorbed energy of the polymer powders and change the morphological properties.^[39] Exposing the layer to a higher energy value also leads to lower surface roughness.^[4] In addition, the state of the powder bed (solid- or liquid-based) could also determine the porosity.^[8] Compared to traditional membranes fabricated by phase inversion, the SLS membranes are denser since the space between two neighbouring pores is solid rather than porous.^[39,55]

SLA—resolution of $\sim 10 \mu\text{m}$ ^[43]—is another method for making 3D printed membranes. SLA is a vat

FIGURE 3 (A) The current limitation of 3D printing technology in membrane research,^[62] (B) cross-section and (C) surface of 3D-printed membranes by Yuan et al.,^[4] (D) change in the porosity with laser power, and (E) change in flux and permeability with laser power (adopted from Yuan et al.^[4] and Lee et al.^[62]). RO, reverse osmosis; NF, nanofiltration; MF, microfiltration; UF, ultrafiltration; SLA, stereolithography; FDM, fused deposition modelling; SLS, selective laser sintering; DLP, digital light processing



polymerization-based method that uses an ultraviolet (UV) laser beam in predetermined directions to start a chain reaction on a layer of photosensitive thermoset polymer resin or monomer solution. After the resin polymerization and forming of a 2D-patterned layer, the platform goes down to form a new layer. The procedure is repeated for the subsequent layers to create the desired 3D-printed object. Acrylic and epoxy resins are the polymers widely used in this technique.^[56] In the SLA method, the morphology of the membrane is significantly influenced by the laser power.^[57]

Inkjet printing—resolution of 5–200 μm^[58]—is a conventional 3D fabrication method that works by the deposition of liquid binder droplets through a nozzle on a platform, including spread powders. In this technique, particles stick together in a pattern and solidify to hold the subsequent layers. After forming a 2D-patterned layer, the platform lowers and the procedure is repeated for the new layers to reach the desired 3D model.^[59] In this method, the rheological behaviour of the ink, particularly viscosity and elasticity properties, mainly contributes to the morphological properties of membranes. The ink with liquid-like behaviour could rapidly spread on the substrate, leading to the formation of a non-porous structure. However, fabrication of a more porous membrane is available using inks with solid-like behaviour.^[60]

3 | APPLICATIONS OF 3D PRINTING MEMBRANES IN LIQUID SEPARATIONS

The 3D printing technology can precisely control the structural properties such as the pore size, roughness, and thickness of the membrane, enabling the fabrication of membranes with specific characteristics that are challenging to achieve with conventional techniques.^[61] In particular, due to the high flexibility of 3D printing in design and process, the fabrication of membranes with different porosities is feasible by regulating the 3D printing parameters related to each method. However, deviation from desirable porosity may appear due to the resolution restrictions in the X and Y axes.^[29] 3D printing technologies have a range of resolutions and specific material inputs, limiting their successful deployment for membrane fabrication (Figure 3A).^[62] Therefore, only a few studies have been assigned to fabricating 3D-printed membranes.^[63] Table 1 presents a comprehensive overview of 3D-printed membranes used in water and wastewater treatment. It should be mentioned that the applications of 3D printing technology to module parts and membrane patterning are not included here.

Most membranes fabricated via 3D printing technology have been used for oil/water separations. Li et al.^[60]

TABLE 1 Applications of 3D-printed membranes in water and wastewater treatment

3D printing type	Material	Application	Performance	Reference
Inkjet printing	Composite of cellulose acetate, polyvinyl alcohol, and silica nanoparticles	Oil/water separation	Rejection of 99%, high water flux (149 000 to 2 160 000 L/m ² · h ⁻¹), improved tensile strength and Young modulus, and high wettability	Li et al. ^[60]
SLS	Polyamide-12 membrane coated with sintering candle soot	Oil/water separation	Improved hydrophobicity and underwater oleophobicity, rejection of 99%	Yuan et al. ^[4]
SLS	Polysulfone membrane coated with sintering candle soot	Oil/water separation	Rejection of 99%, gravity-driven oil flux of 19 000 L/m ² · h ⁻¹	Yuan et al. ^[64]
SLS	Polyamide-12 and hierarchical micro/nanoscale ZIF-L on the surface	Oil/water separation	Numerous voids with a rougher surface, superhydrophobic and underwater superoleophobic, rejection of 99%, the gravity-driven flux of 24 000 L/m ² · h ⁻¹	Yuan et al. ^[65]
SLS	Plastic-based support with a zwitterionic hydrogel coating	Oil/water separation	Improvement of mechanical stability, hydrophilic/oleophobic surface, and the rejection of about 100%	Song et al. ^[6]
Inkjet printing	Graphene/nickel	Oil/water separation	Superhydrophobic/oleophilic surface, applicable in alkaline and neutral environments, high rejection	Li et al. ^[5]
SLA	ABS-like support with a thin film of PES	Pure water	Improvement of the pure water permeance	Al-Shimmery et al. ^[66]
SLS	Clay powders/maltodextrin binder	Water treatment	High rejection	Hwa et al. ^[67]
SLS	Polyamide-12	Water treatment	Tensile strength improvement	Yuan et al. ^[39]
FDM	Porolay Lay-Felt filament (composed of rubber-elastomeric polymer)	Water/atrazine monitoring	Efficient atrazine monitoring	Kaloom et al. ^[50]

Abbreviations: FDM, fused deposition modelling; PES, polyethersulfone; SLA, stereolithography; SLS, selective laser sintering.

fabricated a superhydrophilic and underwater superoleophobic 3D-printed membrane using direct inkjet printing to treat oil/water mixture. Printing inks, including cellulose acetate, polyvinyl alcohol, and silica nanoparticles, were employed in the membrane solution. The 3D-printed membrane showed a low water contact angle of $18.14 \pm 2.61^\circ$ and a high underwater oil contact angle of $159.14 \pm 0.59^\circ$, which confirmed its superhydrophilic and underwater superoleophobic properties. Polyvinyl alcohol and SiO₂ nanoparticles in the membrane structure could interact with water molecules and produce hydroxyl groups on the membrane surface. Interactions between polar hydroxyl groups and water molecules through van der Waals forces and hydrogen bonds created a layer that repelled the oil away from the membrane surface. In addition, a tensile strength of about 8 MP was achieved for this membrane. The 3D-printed membrane also had high water flux (149 000 to

2 160 000 L/m² h⁻¹) when the membrane pore size increased from 115 to 527 µm. However, the removal efficiency was substantially decreased when the pore size was larger than 230 µm. Results indicated that oil rejection of about 99% was achieved with the oily samples, including *n*-hexane, diesel oil, vegetable oil, and lubricating oil. The 3D-printed membrane retained its good performance even in acidic or alkaline environments. In addition, the 3D-printed membrane showed high antifouling and stability with 99% oil rejection after 50 cycles. It seems that such remarkable antifouling behaviour came from the water layer formation on the membrane surface, produced by the interactions between hydroxyl groups and water molecules.

In another work, Yuan et al.^[4] used the SLS technology to fabricate a polyamide membrane coated with sintering candle soot to treat oil/water mixtures. The SEM images of the fabricated membrane are illustrated in

Figure 3B,C. After coating the membrane with candle soot, the water contact angle increased from 101° to 120° , showing enhanced hydrophobicity. Increasing the laser power during the fabrication process decreased both the porosity and permeability features (Figure 3D,E). In addition, the membrane showed a high water rejection of 99.1% in the oil/water system. The high hydrophobic property of the membrane enabled *n*-hexane droplets to be absorbed and penetrated instantly to the membrane while the water was repelled. Soaking the 3D-printed membrane in ethanol and water switched the surface hydrophobicity to hydrophilicity. In this regard, water molecules interacted with candle soot particles and formed a water layer on the surface, which repelled the oil. The 3D-printed membrane maintained a high removal efficiency of 99% in both dry and wet states after 10 cycles.

In another study, Yuan et al.^[64] fabricated superhydrophobic 3D-printed polysulfone membranes coated with sintering candle soot with a switchable wettability. The membrane exhibited a water contact angle of 160° and a sliding angle of 5° . The rejection of different oil/water mixtures, including hexane, heptane, petroleum ether, and mineral oil, was more than 99% using this membrane with the gravity-driven oil flux of $19\,000\text{ (L/m}^{-2} \cdot \text{h}^{-1})$. The same research group fabricated a 3D-printed membrane made of polyamide-12 via SLS followed by the deposition of hierarchical micro/nanoscale zeolitic imidazolate frameworks-L (ZIF-L) on the membrane surface. Compared to traditional flat sheet membranes, the 3D-printed membrane had a higher porosity and a rougher surface. The existence of two layers of micro/nanoscale ZIF-Ls on the membrane surface increased roughness and endowed the 3D-printed membrane with superhydrophobic properties. Furthermore, the 3D-printed membrane showed $24\,000\text{ (L/m}^{-2} \cdot \text{h}^{-1})$ oil flux and 99% water rejection for the different oil/water mixtures.^[65]

SLS was utilized by Song et al.^[6] to fabricate a membrane including 3D-printed plastic support coated by a zwitterionic hydrogel. The rigid 3D-printed plastic support and the hydrogel coating were respectively used to improve the membrane's mechanical stability and hydrophilic/oleophobic surface characteristics. The 3D-printed membrane was used in a gravity-driven oil/water separation system. It could maintain its performance in a long-term operation, as the rejection fluctuated in the range of 85%–95% after 31 cycles of filtration. In the case of antifouling performance, the water flux was almost constant, about $350\text{ (L/m}^{-2} \cdot \text{h}^{-1})$, after 31 cycles.^[6]

In another study, a 3D-printed membrane with the superhydrophobic property was made by directly printing graphene on porous nickel foams to separate oil from

water. A remarkable amount of oil was permeated through the membrane in the first minute, and almost the whole oil was recycled after 20 min. In addition, the 3D-printed membrane maintained its performance after 10 cycles. Utilizing graphene/nickel 3D-printed membrane was feasible in alkaline and neutral environments; however, this membrane was incompatible with acidic environments.^[5]

Al-Shimmery et al.^[66] used the SLA method to deposit a thin PES film onto a 3D-printed ABS support. The performance of fabricated membranes on flat and wavy support surfaces was evaluated in terms of oil rejection, pure water permeation, and fouling behaviour. Both flat and wavy membranes showed a high oil rejection of $96\% \pm 3\%$ since the average oil droplet diameter ($9.9\text{ }\mu\text{m}$) was larger than the average membrane pore size ($54 \pm 10\text{ nm}$). The pure water permeance of the flat membrane was about $12\text{ (LMHbar}^{-1})$, which increased about 30% for the wavy one, resulting from a 13% higher surface area of the wavy membrane than the flat one. Regarding the fouling behaviour, the wavy structured 3D-printed membrane showed 52% more water permeability than the flat one after the first cycle. The wavy membrane also had some level of water permeation even after five cycles, while the flat one was almost fouled after the first cycle.^[66]

In contrast to the oil/water separation, much less attention has been paid to other water treatment applications using 3D-printed membranes. A recent study by Kalsoom et al.^[50] has investigated 3D printing technology in producing a passive sampler device with an integrated membrane (Figure 4). This device was used to monitor atrazine, one of the most contaminating herbicides, in water samples. Using FDM, the membrane was printed by Poro-Lay Lay-Felt filament (composed of rubber-elastomeric polymer). Results showed that atrazine could transfer across the membrane without interfering with other species, including unreacted polymer, plasticizers, or solvents. In this regard, the passive sampler, including the membrane with a thickness of 0.5 mm and an average pore diameter of 34 nm, showed a high atrazine depletion of 87% and a sampling rate of 0.19 Ld^{-1} .^[50]

The mechanical properties of the polyamide-12 membrane printed via SLS were studied by Yuan et al. The microfiltration (MF) membrane had a tensile strength ranging from 4.8 to 28 MPa. Furthermore, membranes made with higher laser power had higher tensile strength because fewer defects were detected in their structure.^[39] A 3D-printed ceramic membrane composed of clay powders was fabricated to treat river water. In this study, clay powders with different particle sizes were mixed with maltodextrin binder and sintered at 1300°C . The main privilege of the 3D-printed clay membrane was its

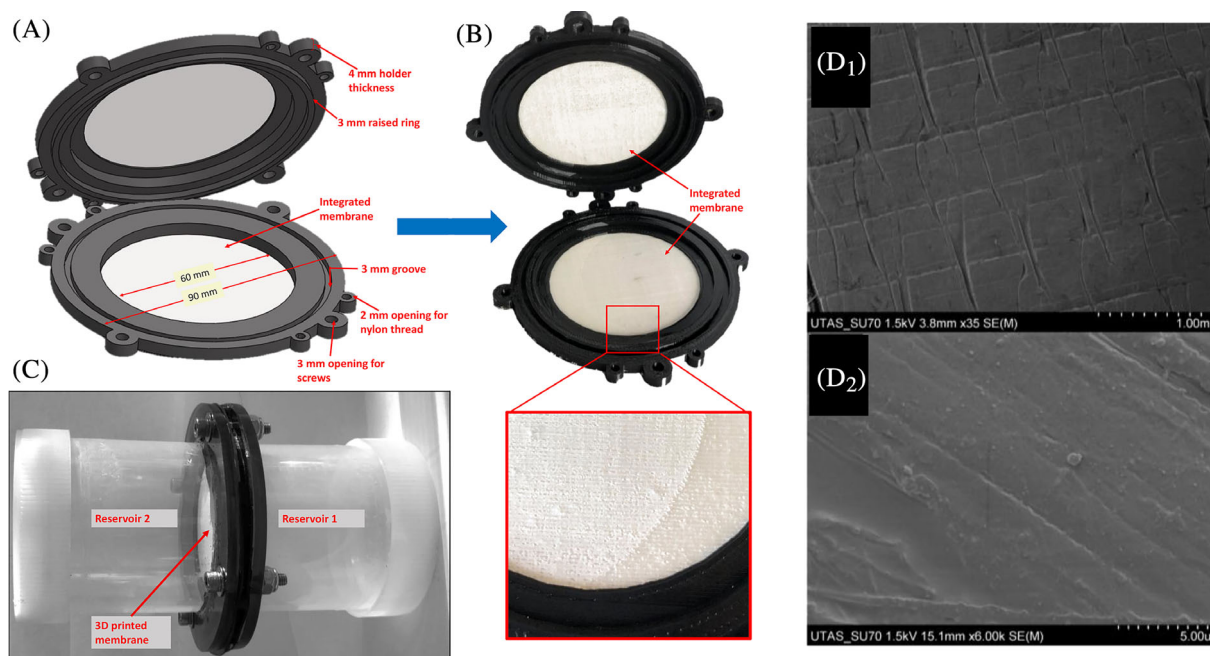


FIGURE 4 (A) Computer-aided design (CAD) and (B, C) images of passive sampling device, including 3D-printed membrane, (d₁) Low and (d₂) high-resolution scanning electron microscopy (SEM) image of the 3D-printed membrane (adapted from Kalsoom et al.^[50])

affordable cost to use in pilot scales. The membrane with a particle size of 75 μm showed a 97.78% and 53.85% reduction in chemical oxygen demand (COD) and total suspended solids (TSS), respectively.^[67]

4 | ENVIRONMENTAL IMPLICATIONS OF 3D-PRINTED MEMBRANES

4.1 | Decentralization

Global evaluations by the World Health Organization (WHO) show that access to enough or microbiologically safe water supplies is not available for a large part of the world.^[68] A decentralized system is considered a practical approach to treat small amounts of wastewater originating from single households, rural dwellings, and peri-urban clusters that have not been connected to the central sewer system.^[35] The use of conventional membranes in decentralized wastewater treatment confronts some challenges, which demands the use of more effective and easy-to-install systems. Conventional membranes cannot rapidly deal with varying water feed qualities, and any change in the decentralized system leads to huge investments. The excessive use of organic solvents and chemicals in the fabrication of conventional membranes is another problem associated with them. Fouling is still the main challenge of applying conventional membranes in decentralized

systems, which requires process control, regulation of the system, and membrane maintenance, leading to high investments and operating costs. Such operations may also be done by unskillful people, which increases the work safety risk. On the other hand, 3D printing technology allows the sustainable fabrication of customized 3D-printed membranes to remove particular contaminants, rapid response to acute problems, cost-effective changes in decentralized systems, and modification of separation performance.

4.2 | Targeted removal of contaminants

Rural areas may be exposed to different contaminants that require the design, evaluation, and installation of specific conventional membranes to remove pollutants related to that region. This procedure is too time-consuming and expensive to provide treated water for small communities. However, high precision in fabricating 3D-membranes with complicated designs enables researchers to predict and apply their intended structure for a specific purpose. This aspect of 3D-printed membranes even allows the targeted removal of contaminants such as endocrine disruptors, perfluoroalkyl, and poly-fluoroalkyl substances (PFAS) related compounds that are less efficiently separated by conventional membranes. In addition, 3D printing technology allows the fabrication of two or more materials across the surface and interface,

providing appropriate variations in physical and chemical properties. Membranes fabricated in this way could separate different pollutants by channelling the feed toward particular membrane parts.^[30] Such advantages allow small communities to easily supply the desired 3D-printed membranes for a specific purpose.

4.3 | Rapid response to acute contamination events

3D-printed membranes could be designed, installed, and configured quickly. Dealing with varying characteristics of wastewater is an essential criterion for membranes in decentralized systems.^[30] In addition, rapid response to acute contamination events like those created by chemical spills, toxic by-products from algal blooms, forest fires, extreme weather, etc., is required to avoid severe environmental problems. For instance, mitigating a new contaminant in an area using a decentralized system based on conventional membranes necessitates remarkable changes in the membrane design and the decentralized system configuration. Such changes are typically time-consuming and might expose the dwellers to severe health problems and multi-day shutdowns of piped water supply. In addition, reduction of infrastructure and operational costs, as main privileges of decentralized systems, would be a question by any change in the water condition since a high expenditure is required to alter the decentralized system. Nevertheless, 3D-printed membranes could be rapidly designed and fabricated in compliance with particular contamination and employed in the decentralized system without significant changes in the system. Therefore, such features show the high potential of employing 3D-printed membranes in small communities using decentralized systems such as rural areas, hospitality resorts, forward operating military bases, and oil and gas extraction bases, which require tunable membrane units.

4.4 | Sustainability and environmental aspects

The sustainability and recycling of the used materials in the membrane fabrication could have substantial economic and environmental effects on commercial scales. Fabrication of conventional MF and UF membranes typically needs the consumption of organic solvents such as *N*, *N*-dimethylformamide (DMF), *N*, *N*-dimethylacetamide (DMA), *N*-methyl-2-pyrrolidone (NMP), and tetrahydrofuran (THF).^[69,70] These solvents are toxic, volatile, flammable, and their regeneration is energy-consuming and inefficient. In addition, the interfacial polymerization approach to making NF and RO membranes could generate waste streams of monomers like *m*-phenylenediamine and

trimesoyl chloride solutions.^[45] Exposure of these solvents to the environment may cause severe problems for aquatic life and public health, and applying them in the fabrication of conventional membranes contradicts the environmental aspects of water and wastewater treatment.^[71,72] On the other hand, the fabrication of 3D-printed membranes is a solvent-free process, which could also donate special features to the membranes with the minimal use of modifying chemical additives. The possibility of designing membranes with minimal use of added chemicals and recycling the 3D-printed materials for the successive cycles shows the high potential of 3D-printed membranes in water and wastewater treatment plants. Fabrication 3D-printed membranes would be a huge step toward a circular economy by designing out waste and unnecessary use of chemicals.

4.5 | Economic considerations

Although the use of 3D-printed membranes is still in its infancy, economic considerations are considered a fundamental factor in their future perspective. The investments in decentralized systems typically include purchasing, transportation, and installation of equipment.^[73] Printing membranes on-site could remarkably decrease the costs associated with construction, supply chain, transportation, and installation. Furthermore, the operational expenses, including demand for chemicals and the maintenance and replacement costs, could be reduced by employing 3D-printed membranes. Fabrication and modification of 3D-printed membranes require fewer chemicals compared to conventional ones. 3D-printed membranes also have limited scrap as the printing materials could be entirely deposited. This point becomes crucial when rare and expensive (nano)materials can be fully used without material wastage or inefficient degradation to the support layer like integrated asymmetric membranes.^[45] In addition, the fabrication of 3D-printed membranes with the ability to design, regulate, and control the membrane structure and surface nanoarchitecture is hypothesized to be more effective in mitigating membrane fouling, resulting in fewer maintenance and membrane replacement costs.

5 | STATUS AND TRAJECTORY OF TECHNOLOGICAL DEVELOPMENTS TO ENABLE 3D-PRINTED MEMBRANES FOR WATER TREATMENT APPLICATIONS

Despite the fascinating advantages of using 3D-printed membranes for water and wastewater treatment, some challenges still necessitate further technical progress. Some of the major challenges are discussed below.

5.1 | Material selection for membrane printing

A wide range of available applications of 3D-printed membranes is restricted by printable materials.^[74] Low stability, incompatibility with water, nozzle clogging, and swelling are the associated problems of using an inappropriate material.^[3] Increasing the number of material choices, especially those used to fabricate conventional membranes, such as PVDF, polysulfone, polyacrylonitrile, polyvinyl chloride, PES, polytetrafluoroethylene, etc., is essential for further development of water and wastewater treatment using 3D-printed membranes.^[75] In addition, research about suitable compositions and efficient reinforcement could be considered in future studies.^[43]

5.2 | Printing resolution and speed

Although 3D-printing technology allows the direct fabrication of 3D-membranes with complex structures, the resolution restrictions could affect the precision.^[76] Membranes used in water and wastewater treatment often have pore sizes on the micro/submicrometer scale. However, the available resolution is restricted to at most 100 nm, which needs further improvement.^[77] In addition, although current 3D printers can provide satisfying resolution in the Z dimension, the same precision may not be obtained in X and Y dimensions, resulting in the deviation of the porosity and pore structure from the designed model.^[29] The printing time is directly affected by the required resolution for the fabrication of membranes, and it may take a long time. The capability of 3D printers to fabricate large sheet sizes with more than 1 m in width is essential for upscaling and mass production of membranes.^[3] Recently, continuous liquid interface production (CLIP) has significantly reduced the production time and could be addressed in future research works.^[78]

5.3 | Void formation and mechanical properties

Void formation, created by the reduction of interfacial bondings between layers, causes the delamination of layers and the reduction of integrity and mechanical strength of the membrane.^[79,80] Although different approaches, such as increasing the thickness of filaments,^[81] reducing the height of each layer,^[82] and optimizing the nozzle geometry,^[83] have been tried, more effective solutions are required to overcome this problem. 3D-printed membranes need to maintain their performance in

unfavourable conditions such as high pressures, extreme pH levels, etc. The void formation typically causes low mechanical strength during the printing process.^[43] In addition, each layer might have different mechanical properties than other layers due to the layer-by-layer fabrication.^[84] Using reinforcements and applying post-treatment processes could promote the mechanical properties; however, they increase the cost and time of the synthesis.

5.4 | Contaminants of interest

Currently, the pore size of 3D-printed membranes is relatively large and appropriate for MF treatments only. However, UF, NF, and RO 3D-printed membranes require subsequent modifications to remove the contaminants. Such changes could be costly and time-consuming. In addition, 3D-printed membranes also have shown good separation performance for oil/water mixtures. This could be extended to drinking and wastewater treatment, desalination, dye and heavy metal removal, and other emerging applications.

6 | COMMERCIALIZATION OF 3D-PRINTED MEMBRANES AND ITS CHALLENGES

3D printing is still an emerging technology, and it can be difficult to industrially upscale it because of many limitations related to the current status of this technology. A wider range of common membrane materials and bigger 3D printers, as well as quality control of products, can potentially tackle these drawbacks.^[3] Although the industry has a substantial interest in using machine printing, the overall production cost for larger industrial-scale applications of 3D printing should be considered, especially compared to many other conventional membrane fabrication techniques. Nevertheless, by using 3D printing, manufacturers are able to produce parts on-demand instead of producing-to-stock, which results in reduced storage and inventory costs. Also, 3D printing is beneficial for distributed manufacturing or digital manufacturing in remote locations by sending 3D CAD files to smaller sites.^[3] With the design optimizations and development of stronger, cheaper, more lightweight, and more environment-friendly materials, the next issue becomes apparent when the mass production of 3D components for water and wastewater treatment is demanded. In this way, 3D printing successfully implements the concept of continuous improvement in the water and wastewater industries, such as some industrial giants like Toyota, leading to notable growth and

worldwide expansion.^[85] The scalability of 3D printing would be vital, particularly for producing parts that are more expensive or even impossible if conventional manufacturing is used, which is a significant consideration to recover the cost in economies of scale.^[86] The high price of the existing 3D printers, particularly high-resolution ones, like poly-jet type printers, is another drawback of current technology.^[34]

One issue that serves as a limitation to the adoption of 3D printers at industrial scale is the lack of broad assessments to explain its economic feasibility in terms of the total production cost for water and wastewater treatment applications. For instance, Thomas et al.^[15] concluded that the commercial-scale application in membrane distillation is limited by the cost of fabricating 3D-printed components. Even though 3D printing has imparted design flexibility in the fabrication of components meeting the industrial demand, there is much room for optimizing the total production cost for a larger industrial scale. Based on the current situation, commercial 3D printing will become more economically feasible in the future as the 3D printing technology will continue to mature, particularly for water and wastewater applications. As mentioned by Huang et al., 3D printing has gained an extraordinary level of adoptions and investments and hence will become economically advantageous in the near future, particularly for 3D components used in membrane processes for wastewater treatment.^[87] Accordingly, Richard D'Aveni reported in Harvard Business Review that 3D printing technology is at the point of entering the mainstream market as it is gaining more adoption levels on an industrial scale.^[88]

7 | CONCLUSIONS AND FUTURE DIRECTIONS

The emergence of 3D printing in membrane technology could provide numerous applications and implications in water and waste treatment processes. As a practical approach to providing safe water in small communities and remote areas, a decentralized system is a realm that the use of 3D-printed membranes could revolutionize. The possibility for accurate design and rapid on-site fabrication of the membranes, tuning of the treatment units, targeted removal of a particular contaminant, easier maintenance, sustainability, and reduction of infrastructure and logistical costs are some of the main benefits of employing 3D-printed membranes in decentralized systems. However, technical problems related to 3D printing have restricted the extensive use of these membranes. In future directions, research about the use of novel printable materials and the mitigation of resolution and printing speed restrictions

is essential to developing these membranes' applications. So far, the implementation of decentralized systems using 3D-printed membranes has not been conducted, and its materialization will be crucial for future research to evaluate the process and costs. In addition, the use of 3D-printed membranes has been limited only to a few studies, while a wide range of areas may benefit from them.

AUTHOR CONTRIBUTIONS

Amir Aghaei: Conceptualization; investigation; methodology; writing – original draft; writing – review and editing. **Mostafa Dadashi Firouzjaei:** Conceptualization; formal analysis; investigation; methodology; writing – original draft; writing – review and editing. **Pooria Karami:** Investigation; methodology; writing – review and editing. **Sadegh Aghapour Aktij:** Investigation; methodology; writing – review and editing. **Mark Elliott:** Project administration; resources; supervision; writing – review and editing. **Yaghoub Mansourpanah:** Investigation; methodology; project administration; supervision; writing – review and editing. **Ahmad Rahimpour:** Investigation; methodology; supervision; writing – review and editing. **João B. P. Soares:** Investigation; methodology; writing – review and editing. **Mohtada Sadrzadeh:** Funding acquisition; project administration; resources; supervision; writing – review and editing. [Correction added on 2 August 2022, after first online publication: Middle initial of João B. P. Soares was missing in the original published article and has been added in this version.]

PEER REVIEW

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DATA AVAILABILITY STATEMENT

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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