Forward Osmosis in Wastewater Treatment

Subjects: Nanoscience & Nanotechnology | Water Resources
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Forward osmosis (FO), driven by the osmotic pressure difference between solutions divided by a semi-permeable membrane, has been recognised as a potential energy-efficient filtration process with a low tendency for fouling and a strong ability to filtrate highly polluted wastewater.

Keywords: forward osmosis ; wastewater treatment ; sewage concentration

1. Introduction

Freshwater is one of the scarcest resources in the world, and water shortage has become a serious problem with the growth of population and urbanisation [1][2]. In order to mitigate the negative impact of water shortage, water mining from wastewater was proposed by Lutchmiah et al. [3]. In this concept, wastewater is a resource for water, nutrients, and energy [3][4]. Various technologies such as advanced oxidation, activated carbon adsorption [5], anaerobic digestion (AD), and coagulation/flocculation (CF) have been applied to recover freshwater and nutrients from different types of wastewater, including sewage, landfill leachate, and textile wastewater [6]. However, one of the major challenges of nutrients and energy recovery from wastewater is the low-strength nature of wastewater, which not only impacts the recovery efficiency but also increases the construction cost for building reactors with large volumes [4]. Therefore, it is critical to find an effective approach to increase the concentration of matters such as chemical oxygen demand (COD) and nutrients (nitrogen and phosphorus) [4][7]. Membrane-based technology including microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) have been studied and applied for various sources of wastewater treatment, including landfilling leachate [8][9] and textile wastewater [10][11]. However, the UF membrane has a low rejection toward small-size particles [12]. NF and RO processes still face challenges, such as high energy requirements, low water recovery rates, and serious membrane fouling [13][14]. For example, a study reported an average energy requirement of 1.2–1.5 kW/m^3 to treat the pre-treated wastewater by a RO system [15].

Recently, forward osmosis (FO) membrane-based technology has shown a growing interest in desalination [16][17], power generation [18], food concertation [19], wastewater treatment, and resource recovery [20][21]. During the FO process, water molecules spontaneously transfer through a semipermeable membrane driven by an osmotic pressure gradient between feed solution and draw solution (Figure 1a). Due to no requirements of hydraulic pressure, the FO process shows advantages such as low energy cost (0.02 kWh/m^3 under certain operational conditions) [7], low fouling tendency, loose fouling layer, and reduced cleaning frequency [22]. Moreover, a FO membrane has a similar pore size to a RO membrane and good rejection of salts, heavy metals [23], and dyes [24]. Therefore, the FO process can be directly used to treat raw wastewater without complex pre-treatment processes [25][26].

So far, many cases about the application of the FO process as a single treatment process and hybrid systems integrating FO and other technologies have been investigated for treating various types of wastewater, such as sewage, landfilling leachate, and radioactive wastewater (Figure 1b). Besides the conventional inorganic draw solutes such as NaCl and MgCl2, several new types of draw solutions such as liquid fertilisers[27] [28] and wastewater from absorption column[29] have been reported. However, challenges including membrane fouling, internal/external concentration polarisation, salt accumulation in the feed solution, and system operational condition optimisation still exist in FO technology.

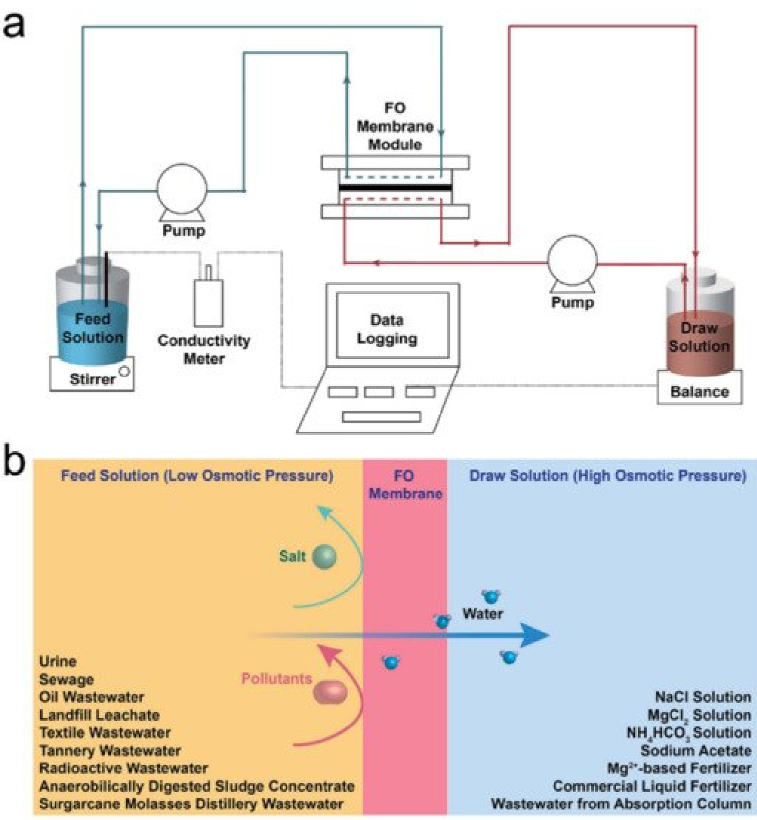

Figure 1. (**a**) Schematic of FO performance evaluation rig set-up, adapted from [30], and (**b**) major wastewater sources and draw solutions reported in FO-based wastewater treatment.

2. Integration of FO with Other Membrane Technologies

Current studies have revealed the feasibility of the FO filtration process in wastewater treatment. However, drawbacks such as the salt accumulation, requirement of draw solution recovery process, and further water mining from draw solution are still challenges that limit the wide application of stand-alone FO processes[31]. As a result, integrating the FO process with other membrane filtration processes such as ultrafiltration (UF), nanofiltration (NF), reverse osmosis (RO) and membrane distillation (MD) is proposed (Figure 2a). Some of the combinations are win-win strategies to apply the advantages of varied technologies and avoid their shortcomings.

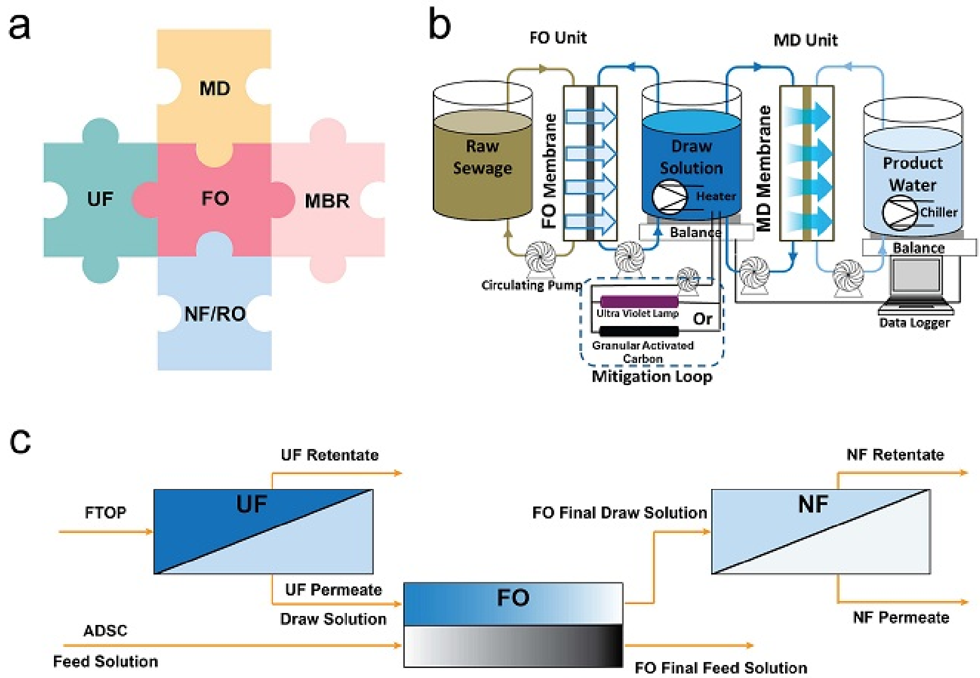

Figure 2. (a) Illustration of the integration of FO with other membrane-based technologies, (b) schematic diagram of the FO-MD hybrid system with the mitigation loop for the reduction of the concentration of feed contaminants in the draw solution, reprinted with permission from[32], and (c) schematic diagram of the UF-FO-NF integrated system, adapted from[33].

Zhang et al. [34] reported a study about the application of a forward osmosis membrane distillation (FO-MD) system for sustainable water recovery and acetic acid reuse from oily wastewater. Results indicated that the FO process showed a large water flux, a high oil removal ratio, and a moderate acetic acid permeation rate. After that, the MD process further rejected the NaCl and oil in diluted draw solution, while completed the regeneration of draw solution. The FO-MD system is also studied for treating wastewater from other sources. Industrial wastewater contains highly toxic heavy metals such as Hg, Cd, and Pb, and its discharge is strictly regulated[35]. Wu et al.[31] investigated the FO-MD system for the treatment of wastewater containing heavy metals. The result indicated that the FO system could effectively reject more than 97% of Hg, Cd, and Pb, and the MD system achieved around a 100% rejection rate of these heavy metals. Human urine consists of 95% water, 3.5% organics, and 1.5% inorganic salt[36], and is a potential water and nutrients source. Therefore, various technologies have been applied in water and nutrient recovery from urine, including electrodialysis, reverse osmosis, freeze/thaw concentration[37], microbial fuel cell, and ion exchange membrane[38][39][40]. Recently, Liu et al.[38] investigated the feasibility of a FO-MD hybrid system for treating urine. The FO membrane rejected most of the TOC, TN, and NH4+-N, but there were still some contaminants accumulated in the draw solution. Then, the MD process further rejected these contaminants, and the hybrid FO-MD system revealed nearly 100% rejection of TOC, TN, and NH4+-N.

3. Integration of FO with Other Wastewater Treatment Technologies

Besides integrating with other membrane-based technologies, the FO process can also be applied as pre-treatment or post-treatment for other wastewater treatment processes. In this section, the integration of the FO process with activated sludge (Figure 3a), AD, CF, Fenton’s oxidation, and ultrasonication (Figure 4a) will be discussed. Moreover, an advanced perspective about reverse salt flux is involved.

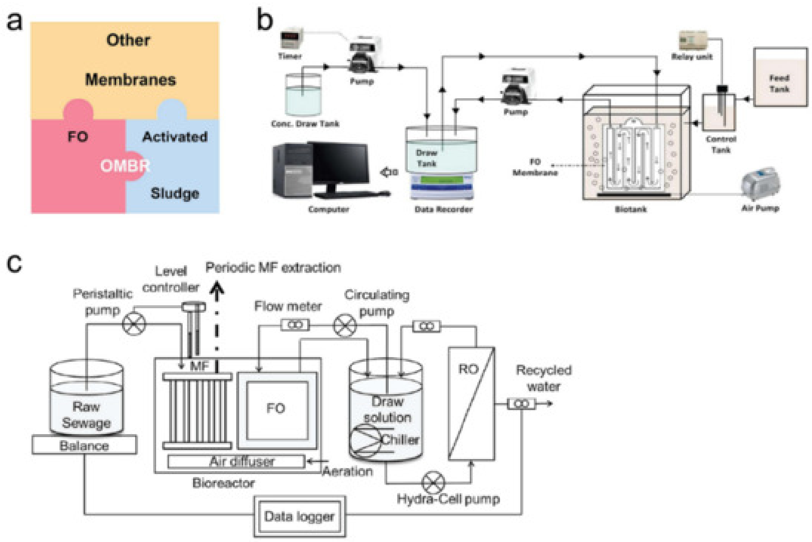

Figure 3. (**a**) Illustration of the integration of OMBR with other membrane technologies, (**b**) the submerged OMBR system, reprinted with permission from [41], and (**c**) the MF-OMBR–RO hybrid system, reprinted with permission from [42].

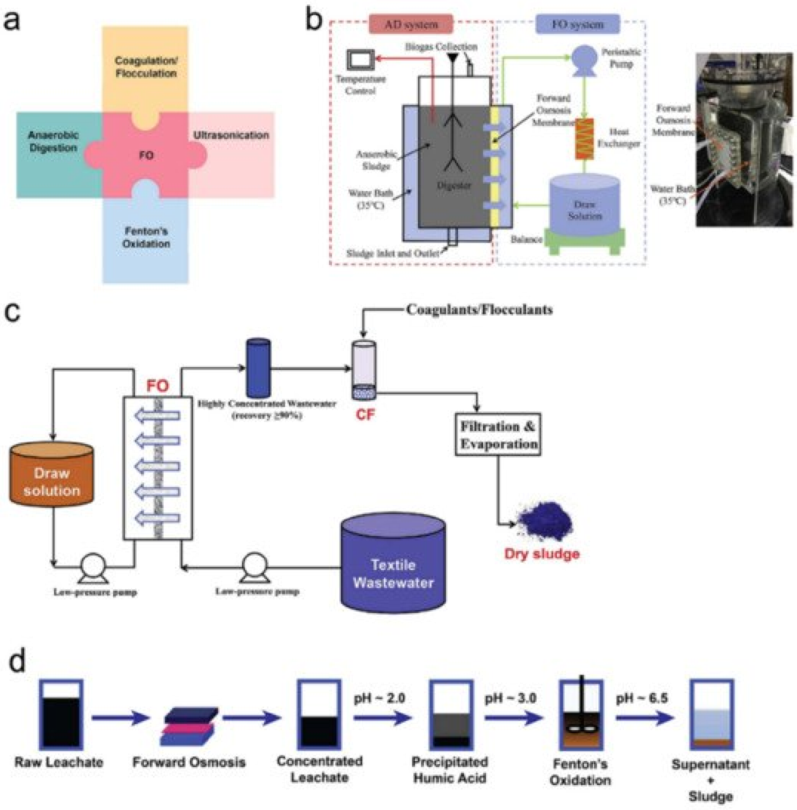

Figure 4. (**a**) Illustration of the integration of FO with other water treatment technologies, (**b**) the hybrid FO-AD system, reprinted with permission from [43], (**c**) the hybrid FO-CF system, reprinted with permission from [44], and (**d**) the integration of FO with Fenton's oxidation, adapted from [45].

3.1. Integration of FO with Biological Process

Apart from using FO as the pre-treatment process for MBR, another integration form between the FO and the biological process (i.e., activated sludge) is the osmotic membrane bioreactor (OMBR) (Figure 3a). In an OMBR system, the FO membrane is submerged inside the MBR and replaces conventional MF and UF membranes, as shown in Figure 6b [46][41]. The driving force in OMBR is osmotic pressure rather than external pressure or suction in MBR. Due to the application of the FO membrane, the OMBR system shows advantages including low fouling tendency and high-quality product water [47][48]. Moreover, the high selectivity property of the FO membrane can effectively concentrate salts, nutrients, and organic matter in the bioreactor, which can improve the efficiency of aerobic or anaerobic treatments [49][50][51][52][53]. Several studies have revealed the feasibility of OMBR systems for treating different wastewater. Qiu et al. [49] reported a study focusing on phosphorus recovery from municipal wastewater by the OMBR system. During the operation, the FO membrane effectively increased the concentrations of PO_4^{3-}, Ca^{2+}, and Mg^{2+}, while the organic matter and NH_4^+ were removed through the biological activities in the bioreactor. By controlling the pH at 9.0, the system recovered 95% of PO_4^{3-} through the precipitation between PO_4^{3-} and Ca^{2+}, Mg^{2+}, as well as NH_4^+. Additionally, during the 84-day continuing operation, the phosphorus recovery efficiency was around 50%. In another study, Yao et al. [46] investigated the removal and degradation mechanism of carbamazepine (CBZ) from wastewater through the application of OMBR. The result indicated that the OMBR system showed a high removal efficiency of COD (94.8%), NH_4^+-N (93.6%), and CBZ (88.2%). Chen et al. [54] reported that the AnOMBR process was more effective than the traditional AnMBR process and demonstrated good removal efficiency of organic carbon (96%) and TP (~100%). Gao et al. [55] also reported a study about the application of AnOMBR for water recovery from real municipal sewage, and results indicated that the AnOMBR system had great removal efficiency of COD (96%), NH_4^+-N (88%), TN (89%), and TP (~100%).

There are still several shortcomings of OMBR systems. Firstly, due to the reverse salt flux from NaCl draw solution and the concertation effect of wastewater by the FO process, salt is accumulated in the reactor [50][51][56]. The accumulated salt can reduce the osmotic pressure differences across the membrane as well as increase the density and viscosity of wastewater, consequently resulting in lower water flux of the FO process [57][58]. It also aggravates membrane fouling and damages the condition of activated sludge for microorganisms [59][55][58]. In order to mitigate the salt accumulation in OMBR systems, several strategies were proposed in current studies, including the selection of suitable draw solution with lower reserve salt flux [60], the fabrication of high-performance FO membranes [61], and the combination of MF/UF technologies with OMBR [62].

Luján-Facundo et al. [60] investigated the feasibility of the application of real wastewater solution from an absorption column consisting mainly of SO_4^{2-} and NH_4^+-N as the draw solution for the OMBR system. Before application, the draw solution was adjusted at 4.0 pH to avoid the chemical damage of FO membranes. Their result indicated that the OMBR system showed low membrane fouling tendency and high water flux with this draw solution. In another study by Luján-Facundo et al. [63], they used the same wastewater as the draw solution to treat tannery wastewater which contained high salinity and organic matter concentration in an OMBR system. Although the reverse salt flux was mitigated by using this new draw solution, the salt accumulation was also observed due to the dewatering process of the naturally high-salinity wastewater. As a result, the efficiency of OMBR and bacterial activity was still significantly affected.

Another effective strategy proposed to mitigate the salt accumulation in the OMBR process is the integration of OMBR with other membrane filtration technology, such as MF, UF, and RO processes. By submerging MF or UF membranes into the bioreactor of the OMBR system, the salt in the bioreactor can be removed by transferring through the MF and UF membranes. As a result, the salinity accumulation was mitigated [64]. Qiu et al. [65] exhibited the application of a MF/OMBR system in their study about phosphorus recovery from municipal wastewater. After the dewatering by the FO process, the wastewater was enriched in the bioreactor. Using the MF membrane, the phosphorous was recovered by extracting the supernatant in the bioreactor. Their results indicated that the hybrid system demonstrated high removal efficiency of TOC (90%) and NH_4^+-N (99%), as well as high phosphorus recovery (>90%). The long-term stability of the MF-OMBR system was evaluated by Luo et al. [66] in a 60-day continuing study, which further indicated that the MF membrane can remove the salts from the bioreactor of the OMBR system and prevent salinity accumulation.

Despite the advantages of the MF/UF-OMBR system, there are some challenges. For example, in a study about TOC and NH_4^+-N removals by a MF-OMBR system, Wang et al. [67] observed the membrane fouling, especially, the reversible fouling in the MF-OMBR was more serious than that in the OMBR system, which led to increased filtration resistance, aggravated ECP, and declined water flux. In addition, the fouling mitigation for the MF/UF-OMBR system may be complex due to the different fouling cleaning strategies between MF/UF (hydraulic backflushing) and FO membranes (osmotic backflushing) [64]. Another drawback of the MF/UF-OMBR system is a lack of technology to regenerate draw solutions. Therefore, Luo et al. [42] investigated a hybrid system integrating MF-OMBR and RO processes (Figure 6c). In this hybrid system, the MF membrane was applied to remove phosphorus from the enriched wastewater and to mitigate the salinity build-up in the bioreactor, while the RO process was for draw solute recovery and clean water production. The result indicated that the hybrid system can produce high-quality water. However, the accumulated organic matter and ammonia in the FO draw solution is a challenge requiring further investigation.

Similar to the stand-alone FO process, the efficiency of FO-integrated systems is impacted by factors such as the draw solution type, draw solution concentration, and cross-flow velocity. However, it is more complex when considering the optimal operating condition for FO-integrated systems since more factors in different treatment processes should be considered. Two of the major energy consumers of the FO process are draw solution reconcentration and recirculation pumps [68]. Cath et al. [69] estimated the specific energy consumption (SEC) of a FO-RO system. Their result indicated that 19 kWh/m^3 was required for solute reconcentration by the RO system, which accounted for 76% of the total SEC of the FO-RO system. Therefore, improving the solute reconcentration efficiency is critical to reduce energy consumption. McGinnis et al. [70] reported a study by using NH_3-CO_2 solution as the draw solution for the FO process. The diluted NH_3-CO_2 solution can be reconcentrated by waste heat, which reduced the energy consumption of the system to 0.84 kWh/m^3. Recirculation pumps also consumed a large proportion of the SEC, which was estimated at 25–30% [68]. He et al. [7][71] found that the energy consumption for recirculation pumps could be reduced by decreasing the recirculation flow rate. However, the reduced water flux of the FO membrane and increased fouling caused by a lower recirculation flow rate should be considered when determining an optimal recirculation flow rate. Park et al. [64] reported a model to find optimal design parameters for the OMBR-RO system. The result indicated that increasing flow rates and concentrations of draw solution could improve the water flux of FO membranes, and could consequently reduce the cost of purchasing FO membrane, but it would also increase energy consumption of the FO system. Vinardell et al. [72] evaluated the feasibility of retrofitting the RO plant (final water production of 45,000 m^3/day) to a FO-RO-MBR plant. The result indicated that the FO-RO-MBR system was economically competitive if the recovery rate of the FO system was maintained at 50%. When the recovery rate increased to 80% or higher, the cost of the hybrid system was larger than that of the standalone RO plant. Therefore, taking into account the factors of both FO membranes and FO systems is critical for a highly efficient FO process.

3.2. Integrations of FO with Other Water Treatment Processes

Anaerobic digestion (AD) is an effective method to treat the waste sludge from wastewater treatment plants. However, due to the long retention time and low-solid concentration in the reactor, AD has a relatively high construction cost for building a large reactor [43]. It has been revealed that using concentrated sludge as the feeding substrate and applying high-solid AD can improve the efficiency and reduce the cost [54][73][74]. Gao et al. [75] reported a system integrating FO with AD processes. Results indicated that the FO process can be an effective pre-treatment process to concentrate sewage which can then be used as the influent for the AD process. Zhao et al. [43] reported a study about integrating OMBR and AD processes. Distinguished from the research by Gao et al. [75], the feed solution dewatering happened at the same time as the AD process in Zhao's system (Figure 7b). The hybrid AD-OMBR system showed better performance than the conventional AD system, with high solid content, organic degradation, and methane content in biogas. Textile wastewater is one of the most polluting wastewaters [76]. The CF process is an effective approach for textile wastewater decolourisation [77][78]. Similar to the AD process, the efficiency of the CF process can be improved by using concentrated dye wastewater [79]. Han et al. [44] reported a FO-CF hybrid system to treat textile wastewater (Figure 7c). In this system, the FO membrane showed a 99.9% rejection rate of dye, and effectively reduced the volume of wastewater. Due to the successful enrichment of textile, the further CF process exhibited a high dye removal rate (>95%) by using 500−1000 ppm of coagulants and flocculants.

Fenton's oxidation is a process that can be used to remove COD and TOC from landfill leachate [80][81]. However, Fenton's oxidation process requires a strict pH condition between 2 and 4 [80][82], resulting in a large amount of reagent dosage such as H_2SO_4. In order to reduce the reagent dosage and enhance the efficiency of Fenton's oxidation, Iskander et al. [45] designed a system integrating FO with humic acid (HA) recovery and Fenton's oxidation technologies. In this integrated system (Figure 4d), the FO process was applied to reduce the volume and alkalinity of leachate by dewatering and the concentration gradient-driven movement of the alkalinity-causing species, respectively. As a result, the required amount of H_2SO_4 for maintaining pH was reduced. After the FO process, the HA recovery process was applied to remove the humid substances from the wastewater, which provided positive effects to reduce the reagent (Fe (II) and H_2O_2) for further Fenton's oxidation. Moreover, the recovered HA can be used to remove aqueous phosphorus, nitrogen, heavy materials by precipitation, and can be a type of fertiliser [45][83][84]. Compared to the single Fenton's oxidation process, the integrated system reduced the required amount of H_2SO_4 by 25.2%, NaOH by 34.6%, and H_2O_2 by 35%.

In another study, Nguyen et al. [85] proposed a hybrid system that integrated ultrasonication and OMBR technologies to further improve the efficiency for sludge disintegration and dewater of OMBR systems. The result showed that the application of ultrasonication could improve the sludge concentration performance of OMBR. Specifically, to increase the sludge concentration from 3000 to 20,400 mg/L, the conventional OMBR needed 26 h, while the ultrasonication OMBR system only required 22 h. Furthermore, the ultrasonication OMBR system could achieve NH_4^+-N removal efficiency of 96%, PO_4^{-3} of 98%, and dissolved organic carbon (DOC) of 99%. The major reason for this improvement was due to that the ultrasonic process affected the sludge solubilisation and reduced floc size, which facilitated the release of organic substances and bounded water into the liquid phase. Moreover, the application of ultrasound mitigated the membrane fouling by hindering the adhesion of foulants on the membrane surface.

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