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The use of membrane processes for the removal of phosphorus from wastewater

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ABSTRACT

Phosphorous (P) is one of the major nutrients (next to nitrogen) contributing in the increased eutrophication of natural waters, that is, rivers and lakes. One of the major inputs of anthropogenic P is caused by the discharge of municipal and industrial wastewater with high P concentration. Therefore, the control of P discharged from treatment plants plays a key role in preventing eutrophication of surface waters. Removal of P from wastewater can be made with the use of the conventional methods, such as chemical precipitation and biological treatment. However, in previous recent years an increase in the application of membrane processes for this purpose has been observed. The work focuses on the critical review of the possibilities in the removal of P from selected wastewater using membrane techniques such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), forward osmosis (FO), and their combination with other treatment methods. The removal rates of total phosphorus (TP) in the processes of MF UF, NF, and FO could reach 34%, 26%, 97%, and 99.7%, respectively. The usage of pretreatment causes an increase in the efficiency rate of TP up to 98.1% for coagulation-MF, 96.7% for biological treatment-UF, and 98% for MBR-NF. The removal rates of PO_4^{3-} may reach up to 11% for MF, 99% for UF, 100% for NF, 91% for MF-NF, 99.7% for MF-softening, and 98% for OMBR (FO). The removal of phosphorus from wastewater in membrane processes is highly effective and can contribute to the prevention of eutrophication. Moreover, the P retained at the purification stage could be further processed, for example, initially recycled, and then used, for example, for fertilizing purposes. It is in the line with the principles of the circular economy (CE) model, which promotes the recovery of raw materials from wastes.

Keywords: Wastewater treatment; Membrane processes; Filtration; Phosphorus (P); Circular economy

1. Introduction

The removal of phosphorus (P) from various types of wastewater has become an emerging worldwide concern because it causes eutrophication in natural water [1]. Eutrophication, which is the increase in the concentration of nutrients (phosphorus and nitrogen) in an ecosystem (mostly undesired), is a leading cause of impairment of many freshwaters and coastal marine ecosystems in the world [2]. The enrichment of water with nutrients could be of a natural origin (natural eutrophication), but it is often dramatically increased by human activities (cultural or anthropogenic eutrophication) [3]. As a consequence of intensive human activity, the anthropogenic eutrophication speeds up the process of natural eutrophication. Due to P being one of the major nutrients contributing in the increased eutrophication of natural waters and lakes, its presence causes many water quality problems, including increased purification costs, decreased recreational and conservation values of an impoundments, loss of livestock, and the possible lethal effect of algal toxins on drinking water. There are three main sources of anthropogenic P input: erosion and leaching from fertilized agricultural areas, and discharge of wastewater from cities and industrial plants [4]. One of the possibilities to reduce the impact of P on eutrophication is the use of highly

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effective methods of municipal and industrial wastewater treatment. This is especially important for the regions, where wastewater discharged to natural reservoirs has high concentration of compounds that contain phosphorus. In Europe, there is a desire to bring the P concentration of wastewater treatment plants' (WWTPs) final effluent to the levels found in natural waters [5].

The removal of P from wastewater could be made with the use of the conventional methods, such as chemical precipitation and biological nutrient removal (BNR) [6–8], which have already been implemented at laboratory, pilot- and full-scale. However, there are many disadvantages, which accompany these methods [9]. Chemical coagulation, which could be conducted with the use of aluminum (Alum, $AlCl_3$), iron ($FeCl_3$, $FeCl_2$, $Fe_2(SO_4)_3$), calcium ($CaCl_2$, CaO), and magnesium ($MgCl_2$, $MgCO_3{\cdot}3H_2O$) [10,11] faces the disadvantages connected with the use of chemical precipitation, the higher maintenance cost [6] associated with chemical dosing and problems with the final handling of the chemicals [12,13]. Moreover, the disposal of the large amount of P-rich sludge from wastewater treatment is a challenge for the treatment plants. It has started to be an important issue for the WWTPs located in the countries, where the recovery of phosphorus from sewage sludge and sewage sludge ash is obligatory, that is, Switzerland and Germany. It requires the further costs associated with the development of P recovery technologies, creation of technical infrastructure and human resources [14]. The traditional biological P removal processes also face many problems. In many cases, biological treatment is not capable of achieving consistently low P concentrations in effluent [5] and requires a highly efficient secondary clarifier and maintenance of a biochemical oxygen demand (BOD): total phosphorus (TP) ratio of at least 20:1. Moreover, it was found that the common and important limitation of presented two processes (chemical precipitation and biological treatment) is that neither of them can produce an effluent containing less than 0.5 mg/dm^3 P [6]. Therefore, there is a significant need to look for other, highly effective P removal methods preferably based on the physical processes, including electrocoagulation and membrane techniques. The work focuses on the critical review of the possibilities of the removal of P from wastewater using the selected membrane techniques, that is, microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), forward osmosis (FO), and their combination with other treatment methods.

2. Membrane technologies in the removal of phosphorus from wastewater

Nowadays, an increase in the application of membrane processes for wastewater treatment has been observed [15]. Development of membrane technologies is the largest among other advanced technologies for wastewater treatment. As they are becoming more and more profitable, meet increasingly stringent environmental legislation, reduce the pollutant load in wastewater, and, at the same time, allow for recycling of recovered water [16,17]. The current paper describes the possibilities of the removal of P from wastewater using MF, UF, NF, and FO [18]. Those techniques are used as a standalone unit processes, as well as in combination with other conventional methods of aqueous solutions treatment [19,20].

In a wastewater treatment system, the phosphorus removed from liquid phase can be classified as soluble phosphorus (SP) and particulate phosphorus (PP). The SP can be divided into soluble PO_4^{3-}, soluble polyphosphate and soluble organic phosphate (SOP) [10]. In the literature, the most popular analyzed forms of the P are TP (as in law restrictions regarding the quality of aqueous solutions) and PO_4^{3-}. The comparison of the effectiveness in the removal of those two P forms from selected wastewater during the membrane processes is presented in Table 1.

2.1. Microfiltration

MF is a separation process, driving force of which is the pressure difference on both sides of the membrane. There is static MF (the flow is perpendicular to the membrane surface) and dynamic (the flow is parallel to the membrane surface). In this solid-liquid separation process, colloids, macromolecules, microorganisms, and microparticles present in the suspension can be effectively removed [22]. MF is not commonly applied as a single stage process and in most cases, the pretreatment with the use of coagulant is proposed before membrane filtration. It contributes to the improvement of the effectiveness of the removal of filtrated substances and reduction of fouling, especially its irreversible form [41].

In the work of Ditrich et al. [42], MF was investigated in order to prove that it was a technically feasible and economically competitive process for disinfection and P removal from secondary effluent. Three different MF systems (systems with flat sheet, tubular, and hollow-fiber modules) with a pore size of 0.2 µm were tested in small-scale pilot plants to find out, whether they were suitable for municipal wastewater treatment. The samples were collected from the final effluent of the Berlin–Ruhleben WWTP. It was indicated, that with a low ferric dosage of 0.014 mol/m^3 prior to the MF, the average effluent TP concentrations of all three MF units were below the target concentration of 0.05 mg/dm^3. Due to unfavorable energy consumption (about 0.2 kWh/m^3) the tests with the cross-flow MF were discontinued. At the same time, authors underlined that when using MF systems in the final effluent of WWTPs, evidence must have been produced in a full-scale MF unit to demonstrate that MF was really suitable for practical application. This, as well as a reliable calculation of investment and operating costs, was the main objectives of further investigations of those authors [43]. In the subsequent research, the authors summarized a survey of dead-end MF systems using different membranes and modules. The competitive trial period, with the most suitable polymeric dead-end MF unit used in parallel with a ceramic dead-end MF unit showed no advantage over an inorganic membrane. MF was indicated as suitable for reaching low target concentrations of TP, <0.05 mg/dm^3 [43].

Alonso et al. [21] studied two membrane filtration techniques, MF and UF, in order to remove the TP and PO_4^{3-} from the secondary effluents from CENTA's in the Northern WWTP I of Seville. During the filtration of wastewater, one MF MEMCOR hollow fiber membrane produced from synthetic propylene, with the filtration area of 30 m^2 was used. MF was conducted using a dead-end configuration at transmembrane pressure fluctuated around 0.04 MPa. The retention coefficients during the MF process were equal to

Table 1
The effectiveness in the removal of P from selected wastewater

References	Membrane process	Medium	Removal rate (%) of phosphorus	
			Total phosphorus	PO_4^{3-}
Alonso et al. [21]	MF	Municipal wastewater – secondary effluents	14	9
Zhang et al. [22]	MF	Wastewater from an automobile plant	–	11
	MF-softening	Wastewater from an automobile plant	–	99.7
Guo et al. [23]	MF	Biologically treated wastewater	–	5
	Flocculation-MF	Biologically treated wastewater	–	97
Lu and Liu [24]	Precipitation–MF	Wastewater from TFT-LCD manufacturing company	–	84–96
Li et al. [25]	MF	Municipal wastewater	34.1	–
	Precipitation-MF	Municipal wastewater	98.1	–
Alonso et al. [21]	UF	Municipal wastewater – secondary effluents	26	–
Kim et al. [26]	UF	Industrial and municipal wastewater	16.7	–
	UF-RO	Industrial and municipal wastewater	83.3	–
Mohammadi and Esmaeelifar [27]	UF	Municipal wastewater	–	85
	Coagulation-UF	Municipal wastewater	–	99
Ravazzin et al. [28]	UF	Municipal wastewater – raw wastewater	18.5	2.4
	UF	Municipal wastewater – primary clarifier effluent	18	5.6
Camarillo et al. [29]	UF	Domestic wastewater	–	99
	Coagulation-UF	Domestic wastewater	–	95
Zheng et al. [30]	Coagulation-UF	Domestic wastewater	–	90
Jun et al. [15]	AnMBR-UF	Municipal food -wastewater	96.7	–
Visvanathan and Roy [31]	NF	Municipal wastewater	95	–
Leo et al. [32]	NF	Wastewater from a pulp and paper plant	70	–
Chon et al. [33]	MBR-NF	Municipal wastewater	–	79–91
Li et al. [34]	NF	Municipal wastewater	–	100
Arola et al. [35]	NF	Municipal wastewater	96	–
	MBR–NF	Municipal wastewater	98	–
Nir et al. [36]	NF	Municipal wastewater	97	–
Qiu and Ting [37]	OMBR (FO)	Municipal wastewater	–	98
Qiu et al. [38]	MF-FO	Municipal wastewater	–	97.9
Wang et al. [39]	FO	Municipal wastewater	99.7	–
Praveen and Loh [40]	Nicroalgal assimilation-FO	Synthetic wastewater	–	89

14% and 9% for TP and PO_4^{3-}, respectively. It was only possible to observe some slight increase in the rate of phosphorus removal (up to 26%) for ultrafiltrated water in relation to microfiltrated water.

In the work [22], the application of crossflow MF for the treatment of phosphorus-containing wastewater did no bring the high removal efficiency. The removal of PO_4^{3-} from wastewater taken from an automobile plant (Hefei, China) was conducted in the tubular MF module with the ceramic membrane at a suitable transmembrane pressure (0.15 MPa) and cross flow velocity (2.1 m/s). The retention coefficient for PO_4^{3-} in the MF process reached 11%. In order to improve the removal efficiency of phosphate and mitigate membrane fouling, lime softening as a pretreatment was investigated. At the lime dosage of 680 mg/dm^3 the removal of phosphate increased to 99.7%, and the permeate flux was about 60% greater than when lime was not used. Moreover, it was observed, that for direct MF the degree of irreversible fouling was higher than that of the softening-MF [22]. The pretreatment should be introduced in the integrated system of P removal from aqueous solutions in order to protect the surface of the used membranes, extend their lifetime and increase the level of removal of impurities.

The effect of pretreatments, namely floating medium flocculation (FMF) and powdered activated carbon (PAC) adsorption, on phosphorus removal in the hybrid membrane system was studied by Guo et al. [23]. Biologically treated wastewater effluent was subjected to the hybrid membrane systems with and without chemical coupling of flocculation and adsorption. In this study, flat-plate MF membrane was used, with total membrane area 3.24×10^{-3}m^2. The solution was circulated along the surface of the flat plate. The membranes used were modified polyvinylidene difluoride (PVDF) Minitan-S microporous sheets (pore size of 0.65 μm). The critical flux was equal to 150 L/m^2 h for the wastewater with no preflocculation, and 200 L/m^2 h with the flocculated wastewater. The results showed that the pretreatment of flocculation did not improve the critical flux significantly as it removed only the organic colloids from the biologically treated effluent. The effectiveness in the removal of P by single stage process – cross-flow MF (CFMF) membrane was very poor and reached 5% for PO_4^{3-}. The implementation of flocculation as a pretreatment process to CFMF improved the removal efficiency of phosphorus to more than 97%, whilst the pretreatment of PAC adsorption increased the organics removal to more than 98%.

An assessment of the capability of enhanced coagulation followed by MF to simultaneous removal of dissolved P (DP), including the dissolved reactive phosphorus (DRP) and dissolved nonreactive phosphorus (DNRP) was carried out by Arnaldos and Pagilla [44]. Samples of wastewater were collected from the Stickney WWTP located in Cicero (Chicago, USA). In the experiments, the analytical grade hydrated aluminum sulfate ($Al_2(SO_4)_3{\cdot}18H_2O$) from Fisher Scientific was used as aluminum source. MF was performed on the samples treated by coagulation using nitrocellulose bottle top 0.22 μm pore size filters (Millipore Corp.). It was observed that DP removal was feasible by enhanced coagulation and MF. The coagulant dose achieving maximum simultaneous removal was 3.8 Al(III)/initial P for DP and at this dose the removal efficiency of DP was equal to 72% with the residual DP concentrations 0.25 mg P/ dm^3. It should be mentioned that the presented research involved the determination of diverse dissolved P species at the level below 0.5 mg P/ dm^3.

The removal of phosphate from wastewater from TFT-LCD manufacturing company in Taiwan in the hybrid precipitation-MF process was studied by Lu and Liu [24]. Calcium salt was used to form precipitates, followed by crossflow MF for solid–liquid separation with the mixed cellulose ester hydrophilic (MCE) membranes with pore size of 0.22 and 0.45 μm. The filtration procedure was repeated with filtration pressure at 0.05 and 0.1 MPa bar, and crossflow velocities at 0.48 m/s for laminar flow and 0.96 m/s for turbulent flow, respectively. It was found, that the chemical precipitation of phosphate was significantly affected by molar ratio and pH. The removal efficiency of PO_4 in hybrid precipitation–MF process increased with increasing molar ratio. When Ca:PO_4 increased from 1.5:1 to 2.5:1, the residual PO_4 concentration decreased from 30.87 (84% removal) to 8.11 mg/dm^3 (96% removal) at pH 8.5. The highest removal efficiency for PO_4 was observed at pH 10.5 [24].

The P removal efficiency from municipal wastewater collected from Stanley Sewage Treatment Works (STW) in Hong Kong was investigated by the Li et al. [25]. An integrated system consisted of an iron-dosing MBR with the flat-plate ceramic membrane module (0.0384 m^2, Meidensha) with an average pore size of 100 nm, and side-stream fermentation for P removal and recovery. The removal efficiency of P by the aerobic MBR was equal to 34.1% with an average TP concentration of 4.2 ± 0.2 mg/dm^3 in the effluent. Authors have indicated that the consumption of particulate P by microbial growth and membrane filtration were the main causes of P removal, which could only retain a small amount of TP from wastewater into the activated sludge of the MBR. An addition of ferric iron ($FeCl_3$) greatly enhanced the P removal efficiency. It was observed that 98.1% of the TP in wastewater was removed by ferric iron-induced precipitation and membrane filtration in the aerobic MBR. In addition to the high P removal efficiency, nearly 53.4% of the TP could be recovered via anaerobic fermentation from the MBR sludge. The P extracted from the sludge reached 47.3 ± 3.4 mg/dm^3 as soluble orthophosphate (OP) in the supernatant. After adjustment of the solution pH to 8.0 by NaOH, over 99% of soluble P was reprecipitated with ferrous iron in the supernatant to form Fe(II)-P precipitates, mainly of vivianite, for collection and recovery [25].

2.2. Ultrafiltration

During the UF process, the separation of compounds with a molecular weight above 500 Da is conducted. The applied pressures do not usually exceed 1 MPa. The process uses membranes, in which the hydraulic resistance is determined by the thickness of the skin layer, up to 1 μm. UF membranes are characterized by high hydraulic efficiency, good separation properties, and resistance to mechanical, thermal, and chemical factors [45]. There is a lot of reported research regarding the evaluation of separation abilities and the possibility of using different UF membranes to remove selected compounds from aqueous solutions.

In the study [26], membrane filtration was used to treat a secondary effluent emanating from a sewage treatment works that treated a combined industrial and municipal wastewater. The UF process was studied as the pretreatment stage before the direction of wastewater to reverse osmosis membrane. The retention coefficient of TP during the single UF process was equal to 16.7%. It increased during the integrated membrane treatment (UF-RO) up to 83.8%. The benefits of UF pretreatment included increased RO flux and overall efficiency, prolonged operation time between cleaning and reduction in operating and chemicals costs.

The effectiveness in the removal of P with the use of UF membrane and samples of Behshahr Ind. Co. wastewater as feed was studied in [27]. One polymeric membrane made of polysulfone (UFPHT20-6338, DOW Company, Denmark) with a molecular weight cut-off (MWCO) 30 kDa was used in this study. In the single UF process, the retention coefficient was equal to 85% for PO_4^{3-}. In UF-PAC experiments, PAC was added to feed tank at different concentrations, and it consequently entered in feed circulation loop. An application of hybrid UF-activated carbon adsorption caused an increase of removal efficiency to 99% (with optimum concentration of PAC about 0.1%). The result showed that UF treatment was an advantageous method for treatment of the wastewater.

In the work [28], UF of municipal wastewater was evaluated in order to compare filtration of raw sewage and primary clarifier effluent from WWTP De Groote Lucht, in Vlaardingen (Netherlands). In the study, a crossflow filtration unit was built using hydrophilic, tubular PVDF membranes of 5.2 mm diameter (X-flow F 4385) with a total filtration area of 0.073 m^2. The retention coefficients for raw wastewater were equal to 2.4% and 18.5% for PO_4^{3-} and TP, respectively. During the treatment of primary clarifier effluent, the obtained retention coefficients reached 5.6% and 18% for PO_4^{3-} and TP, respectively. In the investigated range of transmembrane pressure (TMP: 0.03, 0.05 and 0.1 MPa) and crossflow velocity (1.0, 1.5, and 2.0 m/s), direct UF of raw sewage and primary effluent resulted in the same fouling mechanism.

An assessment of P removal efficiency in the integrated membrane system (coagulation-UF) was investigated by Citulski et al. [46]. The secondary effluent from municipal WWTP was used as the feed with an initial concentration of P approximately 5 mg/dm^3. In the first stage of research, conventional coagulation-flocculation-settling treatment was indicated as the pretreatment process. The alum and ferric chloride were used as the coagulants. Both, alum and ferric chloride effectively removed P to below the 0.3 mg/dm^3 threshold when applied as a pretreatment at optimized doses, both of which were below the WWTP's current coagulant dose (as ferrous chloride). Authors indicated that presented simplified pretreatment scheme provided consistent enhanced removal of phosphorus and organic compounds. The obtained findings suggested that simplified in-line coagulant addition in advance of immersed UF membranes enhanced the ability to produce treated effluent suitable for water-reuse applications.

The use of UF for the removal of phosphate ions from treated domestic wastewaters was described by Camarillo et al. [29]. A method of micellar enhanced ultrafiltration (MEUF) with hexadecyltrimethylammonium bromide (CTAB) was proposed to remove P from treated domestic wastewaters from the reclamation station of University Campus of Toledo (Spain). The obtained phosphate rejection coefficients were equal to 99% (a phosphate concentration in permeate of 1 mg/dm^3 if feed concentration was 95 mg/dm^3). The further stage of research focused on the comparison of the behaviors of different surfactants – hexadecylpyridinium chloride (CPC) and octadecylamine acetate (ODA), and their influence on the P removal efficiency. Best results in terms of both, phosphate rejection coefficient and permeate flux were 95% and 186.4 L/m^{2}h (LHM) at 1 mM phosphate concentration, 0.1 mM CTAB concentration, 25°C, tangential velocity 3 m/s, and transmembrane pressure 0.4 MPa.

Phosphorus removal by applying in-line coagulation prior to ultrafiltration of treated domestic wastewater was studied by Zheng et al. [30] in the lab and pilot scale. The following coagulants were used in the first stage of the research: $FeCl_3$, $AlCl_3$, and polyaluminium chloride (PACl) in order to P removal from treated domestic wastewater collected from the WWTP Ruhleben in Berlin (Germany). The lab-scale Amicon cell filtration test was used to quantify the filterability of different water samples. In this phase, the UF membrane from hydrophilized polyethersulfone (PES) with a filtration area of 0.00287 m^2 (NADIR®UP150) and MWCO 150 kDa was installed in the Amicon cell (Amicon 8200, Millipore, USA). In the pilot test, a UF pilot plant (W.E.T., Germany) with PES membrane (Dizzer 450, Inge AG, Germany) was used to filter different waters. The membrane module had a filtration area of 4.5 m^2 and the MWCO of the membrane was 100 kDa. In the work, it was operated in dead-end filtration mode at a constant flux of 60 L/m^{2}h. It was proven (in lab-scale experiment) that the single stage UF could remove some P-compounds as part of them was in colloidal form. Relationship of relative coagulant dosage (mol Me^{3+} to mol TP or OP in feed water) and P concentration in permeate was evaluated in lab-scale UF filtration tests. The TP concentration in the permeate after UF filtration was lower than 0.05 mg/ dm^3, when the relative dosage (using $FeCl_3$ and $AlCl_3$) was higher than four, while the removal efficiency was equal to approximately 90%. In the case of OP the removal efficiency reached almost 99%. At each dosage level, the most significant removal was achieved using $FeCl_3$, while the lowest using PACl. P removal effects obtained through lab-scale experiments could reflect to the performance of coagulation in pilot-scale tests. The calculation based on long-term monitoring showed that a relative dosage of 2.5 mol/mol (for $FeCl_3$ and $AlCl_3$ to TP in feed water) was sufficient to keep TP concentration lower than 0.05 mg/ dm^3 in permeate. The presented value was lower than that obtained from the lab tests. Authors also indicated that UF pilot plant was operated for at least several days and the accumulated fouling layer may also have contributed to the removal of P. Moreover, $FeCl_3$ was considered a suitable coagulant for fouling control and simultaneous phosphorus removal during in-line coagulation with UF.

The UF was tested in a pilot-scale anaerobic membrane bioreactor (AnMBR) installed and operated in a side-stream configuration at the Cheongna municipal food-waste treatment plant in Incheon (Korea). Leachate discharging from the food waste treatment facility was directed to methanogenic reactor, which was connected to a 100 kDa MWCO PVDF tubular UF membrane system (Jinshui Membrane, China). UF was conducted in a cross-flow mode with feed pressure from 0.09 to 0.3 MPa, and cross-flow velocity was maintained between 1.2 and 2.1 m/s. The membrane system was composed of two separate membrane skids operated in parallel. Each skid had six membrane vessels in series and each membrane vessel encased seven tubular membranes of 11 mm in diameter and 3 m long. Consequently, the membrane system offered a total membrane area of 13 m^2. The investigated 100 kDa UF membranes removed up to 96.7% of phosphorus. The high P removal suggested that P-rich compounds existed as suspended solids in the form of mineral precipitates. Authors also indicated that mineralization of phosphorus in the methanogenic digester could be a consequence of several favorable scaling conditions induced by anaerobic fermentation in AnMBR [15].

2.3. Nanofiltration

NF membranes are characterized by relatively low retention of monovalent ions and high rejection of bi- and polyvalent ions and organic compounds with molecular weights exceeding 200–300 Da. The pressures used for NF range from 0.5 to 2 MPa. The application of the NF method allows to obtain a high efficiency of removing pollutants, even in the case of co-occurrence of a significant amount of organic substances (humic acids) in water environment. The combination of NF into systems integrated with conventional processes allows to increase the efficiency of removing contaminants in comparison with unit processes [47,48].

An evaluation of possible use of NF for the removal of phosphate from wastewater was conducted by Visvanathan and Roy [31]. One flat sheet NF membrane (type Desal-5 thin film) in a plate and frame module was used. During the experiment, the influence of pressure, initial feed concentration, competing compounds, and on NF performance were determined, at the TMP 0.4–1.0 MPa. Results showed that P removal efficiency during the NF process could be higher than 95%. Moreover, it was observed that higher pressure and concentration showed positive response, while the presence of competing compounds showed negative response on P removal from liquid phase.

NF was tested for the removal of pollutants from the domestic wastewater by Choi et al. [49]. One hollow-fiber NF membrane (Toyobo Co.) produced from cellulose acetate was used with the effective surface area of a module equal to 11.7 m^2. During the experiment, TMP and relative productivity were 0.036 MPa and 1.0–1.2, respectively. It was primarily expected, that phosphorus would be removed by the effect of size exclusion to some extent. However, the PO_4^{3-} and TP were not removed except for the initial 20 d, when the rejection might have occurred due to the surface charge effect. Authors indicated that a tighter membrane could have been chosen to remove phosphorus by the size exclusion effect.

Leo et al. [32] tested the possible usage of NF membranes for the removal of P from wastewater samples from a pulp and paper plant. The following commercial membranes, DK5, MPF34, NF90, NF270, and NF200, were characterized and tested in permeability and P removal experiments. The highest rejection of P was achieved for NF90 membranes, retention coefficient more than 70% for a feed containing 2,500 mg/ dm^3 of P at a pH < 2. In addition, NF90, NF200, and NF270 membranes showed higher permeability than DK5 and MPF34 membranes. The separation performance of NF90 was a consequence of P concentration and pressure, which may have been due to concentration polarization and fouling. Authors underlined that by adjusting the pH to 2 or by adding sulfuric acid, the separation performance of NF90 could be improved. However, the presence of acetic acid significantly impairs the P rejection.

The membrane bioreactor and NF hybrid system were investigated to demonstrate the performance of removal of phosphorus from municipal wastewater from Gwangju WWTP (Republic of Korea). Authors investigated the MBR-NF hybrid system comprising of one submerged MBR with the U-shaped hollow-fiber polyvinylidene MF membrane (Cleanfil®-S30V, Kolon Membrane, Korea), produced from polyvinylidene fluoride with effective surface area 0.05024 m^2, and three different types of NF membranes (NF40, NF70, and NF90, produced from polyamide thin-film composite by Woongjin Chemical, Korea, with effective surface area 0.00582 m^2 and MWCO 1,000, 350, and 210 Da, respectively). The phosphate ions were found to be the major fractions of TP concentration (>98%) in the raw and treated municipal wastewater. PO_4^{3-} in the MBR permeate was considerably removed by the NE40 membrane (79%) and the NF70 membrane (91%), indicating that the numbers of negative charge had a great influence on the removal of negatively charged ions by negatively charged membranes. The MBR-NF hybrid system is believed to be a promising option for the removal of nutrients from municipal wastewater using membrane processes to replace the combination of a CBWT and an integrated membrane system (i.e., MF or UF and RO) and an MBR/RO system [33].

Li et al. [34] studied the process of NF in the removal of pollutants from municipal wastewater. The effluent of an oxidation ditch from the WWTP in Changping District of Beijing (China) was directed to a laboratory-scale NF system (Shanghai Shiyuan Bioengineering Equipment Co., China), which consisted of three spiral wound NF modules connected with a centrifugal pump (MG80B2-19FT100-D1, GRUNDFOS, Denmark). The NF performance was optimized, and its fouling characteristics after different operational durations (i.e., 48 and 169 h) were analyzed to investigate the applicability of NF for water reuse. The optimum performance was achieved when TMP 1.2 MPa, pH = 4, and flow rate = 8 dm^3/min using a GE membrane. At these conditions and at the initial concentration of PO_4^{3-} 0.151 ± 0.11 mg/dm^3, the removal rate of P was equal to 100%. The authors have indicated that the permeate water quality could satisfy the requirements of water reclamation for different uses and met the local standards for water reuse in Beijing.

In the work Arola et al. [35], the real wastewater was directed to the pilot scale membrane module in a small municipality in Finland. In the research, an MBR pilot unit (Alfa Laval) containing two separate process lines was evaluated: concept A – MBR without chemical precipitation of P and concept B – MBR with chemical precipitation of P. The presented lines were operated in parallel with a full-scale conventional activated sludge process (CAS). The MBR without P precipitation was combined with high permeability NF, regarded as the tertiary treatment. The removal rate of TP with the use of NF270 membrane was equal to 96% and the final removal rate reached 98%. This enabled the recovery of P from the NF concentrate, which meant that the recovered P could be in a more readily usable form, as it was not tightly bound with iron salts, such as the P recovered in the traditional MBR process (concept B). Authors underlined that the MBR without chemical precipitation of P combined with NF was a potential alternative for future sustainable wastewater treatment, especially when very efficient rejection of nutrients was desired.

Nir at al. [36] introduced a new approach for the removal and recovery of P from wastewater. It comprised of a low-pressure NF step applied to a tertiary effluent, followed by a Ca-P crystallization step applied on the retained solution. Cleaning with nitric-acid was applied for maintaining the performances of the acid-durable membrane and recover N-P-Ca liquid fertilizer. In the filtration experiments, a cross-flow NF setup, supporting a flow cell housing a 0.0070 m^2 flat sheet membrane, were used. The membrane used was "Duracid" by GE Osmonics, a commercial acid-durable membrane which was designed (and tested) to operate continuously at extremely acidic conditions (e.g., 37% HCl). The process of sodium-phosphate salts filtration was conducted at the applicable pH range (6–8). The high TP removal rates showed that the Duracid membrane could be used in the phosphate polishing step. At pH 7–8, removal rates exceeded 97% even at low permeate flux, while at pH 6 P rejections were in the range 92%–95%. Authors indicated that the results of a comprehensive thermochemical theoretical analysis and economic analysis indicated that low-pressure NF was a technoeconomically viable alternative for P removal and recovery from wastewater and the further study should be conducted.

2.4. Forward osmosis

FO is a membrane separation techniques with a semi-permeable membrane placed between a feed solution (FS) of a low osmotic pressure and a (DS) of high osmotic pressure, and is driven by the osmotic pressure difference across the membrane. The process of FO has a range of potential benefits (including low energy consumption, low fouling propensity, reduced or easy cleaning, low costs, high salt rejection, and high water flux), mainly due to the low hydraulic pressure required by this osmotically driven process [50]. Recently, this process has attracted growing attention in many potential applications such as wastewater treatment, power generation or desalination.

Direct phosphorus removal and recovery from municipal wastewater collected from WWTP in Kern, Balingen, (Germany) with the use of osmotic membrane bioreactor (OMBR) was conducted by Qiu and Ting [37]. The bioreactor had an effective volume of 4.85 L, housed a plate-and-frame module holding two pieces of OsMem™ CTA-ES flat-sheet membrane (Hydration Technologies Inc., Albany, OR) with the active layer of the membrane facing the mixed liquor, and an effective membrane area of 2 × 0.018 m^2. Due to the high rejection property of the FO membrane, the significant removal efficiency of PO_4^{3-}-P was achieved within the bioreactor – up to 98%. In the further stage of the research, more than 95% of PO_4^{3-}-P was recovered via ACP precipitation, with the phosphorus content in the recovered solids >11.0%. In principal, this process can recover almost all the P, apart from that assimilated by bacteria. Global evaluation showed an overall P recovery efficiency of 50% [37]. In the further research of above authors [38], a hybrid MF-FO membrane bioreactor (MF-FOMBR) for direct P recovery from municipal wastewater was used. In this process, a FO membrane and a MF membrane were operated in parallel in a bioreactor. The use of FO membrane allowed for PO_4^{3-} rejection and resulted in its enrichment in the bioreactor. The P compounds were subsequently extracted via the MF membrane. Authors underlined that P was then recovered from the nutrients enriched MF permeate via precipitation without addition of an external source of calcium or magnesium. The use of seawater brine as a draw solution was indicated as the novel aspect of the presented system. The removal efficiency for PO_4^{3-}-P was equal to 97.9%, and it was rejected by the FO membrane and enriched within the bioreactor. It should be also noted that more than 90% P recovery was observed at pH 9.0. The precipitates were predominantly amorphous calcium phosphate with a phosphorus content of 11.1%–13.3%. In principal, this process could recover almost all the P compounds, apart from that assimilated by bacteria for growth.

Wang et al. [39] investigated the use of FO membrane system for concentrating low-strength municipal wastewater. A pilot-scale FO membrane system using a spiral wound FO membrane module with an effective area of 0.3 m^2 for concentrating real municipal wastewater was used in this study. It was indicated that during long-term operation, 99.7% of TP rejection rate could be achieved at an average flux of 6 L/m^2 h. It should be noted that a long-term investigation of FO systems under continuous flow operation for concentrating low-strength domestic/municipal wastewater was in great need in order to push forward the applications of this method to WWTPs.

In the work of Praveen and Loh [40], osmotic membrane photobioreactor (OMPBR) was used for tertiary wastewater treatment. A plate-and-frame membrane module was prepared using commercial thin film composite (TFC) FO membranes (HTI, USA) and the module was immersed in the bioreactor tank for osmotic filtration. Two pieces of membranes were used in the module, resulting in an effective filtration area of 0.036 m^2. During the study, the synthetic wastewater was used. The obtained removal rate of PO_4^{3-}-P was equal to 89%. The result showed that that OMPBR, which combined microalgal assimilation and FO filtration may have been a promising application in nutrients removal from wastewater.

As phosphorus causes eutrophication and unsatisfactory effects of its removal are noted in conventional processes, there is a justified need to look for highly effective methods of removing P-compounds from wastewater. In recent years, efforts have been made to introduce new technological solutions or to force modernization of already existing processes at WWTPs. The result is the development of highly effective integrated methods of wastewater treatment, involving the combination of physicochemical or biological methods with membrane processes. [51]. Therefore, the attention should be paid to the possibility of introducing developed membrane modules to the technological sequence of existing and planned WWTPs. The area of application, as well as technologies for the preparation of new membranes have been developing very dynamically in recent years, which positively affects the increase in the use of membrane technologies [52].

3. The use of phosphorus from wastewater sector

The use of high effective technologies for the removal of P from wastewater could contribute not only to the prevention of eutrophication but also to the protection of this valuable raw material. Phosphorus retained at the purification stage should be further processed – initially recycled, and then used, for example, for fertilizing purposes. It is in the line with the principles of a circular economy (CE) model, which promotes the recovery of raw materials from waste streams [53,54]. By increasing the recovery of P-compounds in various branches of industry, it is possible to achieve both, economic (reduction of imports) [55] and environmental (reuse of waste) benefits [56,57]. It is extremely important as P is an essential element for growth in all organisms, which cannot be replaced by any other element. Moreover, P-sources are extremely important for European economy [14]. Phosphate rock and phosphorus are listed by the European Union (EU) as the critical raw materials (CRMs) due to the risks of their shortage of supply and the impacts of a shortage on the economy are greater than those of most other raw materials [58]. The European Commission (EU) underlines that phosphorus is wasted and lost at every stage of the cycle. Household waste containing high levels of phosphorus (mainly sewage sludge) after being recycled in accordance with the CE model could cover about 20%–30% of the demand for phosphate fertilizers in the EU. However, this investment potential is still largely unexploited in European countries [59]. Controlling phosphorous discharged from municipal and industrial WWTPs [60], conducting research on advanced P rejection and recovery technologies also play a key role in preventing eutrophication of surface waters. Moreover, it may affect the keeping of this raw material in value chain, which will contribute to its supply safety [58].

4. Conclusions

The negative effects of P-compounds on the condition of the aquatic environment cause the necessity of the removal of these compounds from aqueous solutions in conventional methods. For removal of various forms of phosphorus from water and wastewater biological, chemical and physical processes can be used. High rejection rates could be observed in the membrane processes as MF, UF, NF, FO, and their combination with other treatment methods.

Many scientists have carried out research on the assessment of the possibility of removing phosphorus from wastewater. Based on the detailed review of available literature the following retention rates of TP and PO_4^{3-} were reported:

- TP: 34.1% for MF, 26% for UF, 97% for NF, 99.7% for FO, 98.1% for coagulation-MF, 96.7% for biological treatment-UF, and 98 for MBR-NF;
- PO_4^{3-}: 11% for MF, 99% for UF, 100% for NF, 91% for MF-NF, 99.7% for MF-softening, and 98% for OMBR.

The removal of P from wastewater with the use of membrane processes is high effective (up to 100%) and could contribute to the prevention of eutrophication. Moreover, the P retained at the purification stage could be further processed – initially recycled, and then used, for example, for fertilizing purposes. It is in the line with the principles of the CE model, which promotes the recovery of raw materials from wastes.

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Abbreviations

AnMBR — Anaerobic membrane bioreactor
BNR — Biological nutrient removal
BOD — Biochemical oxygen demand
CE — Circular economy
CRMs — Critical raw materials
CAS — Conventional activated sludge process
CF — Cross-flow
DNRP — Dissolved nonreactive phosphorus
DRP — Dissolved reactive phosphorus
DS — Draw solution
EC — European Commission
EU — European Union
FMF — Floating medium flocculation
FO — Forward osmosis
CPC — Hexadecylpyridinium chloride
CTAB — Hexadecyltrimethylammonium bromide
MEUF — Micellar enhanced ultrafiltration
MF — Microfiltration
MCE — Mixed cellulose ester hydrophilic
MWCO — Molecular weight cut-off
NF — Nanofiltration
NOM — Natural organic matter
ODA — Octadecylamine acetate
OP — Orthophosphate
OMBR — Osmotic membrane bioreactor
OMPBR — Osmotic membrane photobioreactor
PP — Particulate phosphorus
P — Phosphorous
PACl — Polyaluminium chloride
PVDF — Polyvinylidene difluoride
PAC — Powdered activated carbon
SOP — Soluble organic phosphate
SP — Soluble phosphorus
TFC — Thin film composite
TP — Total phosphorus
TMP — Transmembrane pressure
UF — Ultrafiltration
WWTPs — Wastewater treatment plants

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