CHAPTER

6

Latest development in arsenic removal by membrane technology

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6.1 Introduction

The word arsenic (As) derives from a Persian term, which means “yellow ornament” and in the past was used even for murders, as well reported also in the famous thrillers. This element is found naturally in rocks, soil, and air; in addition, pollution resulting from production activities also increases its release into the environment. The presence of arsenic in soils and waters is a global problem considering that this highly toxic element is a natural component of the earth surface capable of contaminating food also through irrigation. As is a semimetal normally present in dissolved water in the form of arsenate As(V) or arsenite As(III). In some territories (Cambodia, Vietnam, the Amazon region, China, Pakistan, Bangladesh, Southeast Asia, Sahel, and other regions of Africa), its concentration is so high to cause serious problems for human health. Exposure to As occurs by ingestion, inhalation, or contact with the skin, in particular, through the water used to drink and in the food preparation or by the ingestion of foods such as cereals and fish that absorb the water of their natural habitat. Tobacco smoke is another cause of exposure to As as well as the exposures in workplaces such as foundries. As is also present in the atmosphere and comes from various sources such as volcanoes (3000 tons/year released) or combustion of fossil fuels (80,000 tons/year released) (n.d.). According to the WHO, the consumption of drinking water containing As in quantities between 0.05 and 0.1 mg/L increases the risk of developing skin/lung/bladder cancer. Consequently, the maximum As concentration in the water for human consumption is set at 10 μg/L by the WHO and Directive 98/83/EC (n.d.). Therefore, considering that nowadays, the water availability, both superficial and underground, is limited and the natural resource cannot be directly used due to the harmful specific contaminations, a series of treatments are required in order to be able to make it consumable. In addition, treated municipal and industrial wastewater can also become sources of clean water useful for the people (Figoli et al., 2016). In this respect, this chapter deals with the different treatment technologies that can be used to purify waters contaminated by As. It begins with a short summary of the

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conventional As removal methods (not based on the use of membranes) and then focuses on the last development in As removal by membrane technology, with the details of the research works reported in the literature and future perspectives.

6.2 As removal methods

In recent years, a lot of research work has been done in the field of As removal techniques from water in order to reduce the risks to human health as much as possible, especially in the areas where the problem of the lack of drinking water reaches important levels. The methodologies currently in use can be divided into two groups: the first concerns processes that do not use membranes to carry out the separation of harmful elements from aqueous streams and the second, on the other hand, that uses membrane technology to recover water usable by humans.

6.2.1 Conventional "non-membrane" techniques

Main treatments that do not concern membrane processes are adsorption, ion exchange, phytoremediation, nanophytoremediation, phytobial remediation, chemical precipitation, electrokinetic processes, and electrocoagulation. These processes allow to obtain As removal efficiency values higher than 90% (with rejections usually higher for As(V) than As(III)), but each of them has its significant drawbacks, particularly concerning costs and efficiency. Table 6.1 shows a summary of the main advantages and disadvantages of the abovementioned methodologies, whereas Fig. 6.1 reports the typical As removal efficiencies (Alka et al., 2021).

The reported literature data (Alka et al., 2021) suggest that the highest removal efficiency for water treatment can be reached by electrocoagulation, followed by ion exchange. Nanophytoremediation and phytobial remediation are effective for treating contaminated soils. Nevertheless, long-term investigations on the impact of nanoparticles and microbes on the soil and plants are mandatory for future implementations.

6.2.2 Membrane technology

In general, a membrane is made of polymeric or inorganic material, which act as a selective barrier allowing the passage of some elements and blocking that of others. In order to promote the passage through the membrane (separation process), a driving force (pressure difference, concentration difference, electric potential difference, or temperature difference that generates a difference in vapor pressure) is required between the two sides of the selective barrier. In the field of technologies used for the As removal from contaminated streams, membrane processes are the most expensive ones, but offer significant advantages such as high efficiency, low energy consumption, high filtration performance, no production of toxic solid waste, ability to remove other contaminants and microorganisms, easy disposal of used membranes, minimal maintenance and operating requirements, and no added chemicals (Zakhar et al., 2018). In the specific case of membrane processes employed for the treatment of streams contaminated by As(III) and As(V), the most applied membrane operations are reverse osmosis (RO), forward osmosis (FO), nanofiltration (NF), ultrafiltration (UF), microfiltration (MF), membrane distillation (MD), electromembrane processes, supported liquid membrane (SLM), polymer inclusion membranes (PIMs), and membrane bioreactors (MBRs). The separation by these techniques depends both on the pore size of the membrane and the driving force (pressure gradient in case of NF, UF, MF, RO; concentration

TABLE 6.1 Summary of the main advantages/disadvantages of the conventional techniques.

Techniques	Advantages	Disadvantages
Adsorption (the elimination of substances from gaseous or liquid solutions occurs through the use of solids as granular activated carbon; activated alumina; waste carbonaceous materials; biochar; polymeric adsorbents; magnetite nanoparticles; iron-coated sand)	Easy system configuration with simple operating mode; excellent capacity thanks to the high selectivity for As; flexibility; low cost; simple disposal without sludge treatment	The adsorbent must be replaced at regular intervals; loss of separation ability over time; usable for wastewater with low As concentrations
Ion exchange (solid phase ions shared with equal ion numbers from contaminated water)	Total removal and recovery of metals; low sludge production	Removal at regular intervals; costly; each exchanger is specific for an As species; resin more reactive toward natural anions
Phytoremediation (it uses plants to treat contaminated soil)	Ecological; high economic value; prevention of the spread of the contaminant; land restoration	Long process time; the process is affected by climatic conditions, especially tropical ones; the presence of microbes leads to the further production of toxic material; lacks of extensive applications; problems for plant growth and development
Nanophytoremediation (it uses nanoparticles to decrease heavy metals; it uses nanoencapsulated enzymes to increase the activity of microbes and plants)	It favors the efficiency of phytoremediation, remediation in situ and degradation of pollutants into less toxic forms; cost effectiveness	Impact of nanoparticles on soil and plants not investigated yet
Phytobial remediation (Employees' interactions between plants and microorganisms)	Ecological; low costs; favors speed of phytoremediation and plant defense; guarantees biological control of phytopathogens and increases soil fertility	Impact of microbes on soil and plants not investigated yet
Chemical precipitation (forms a separable solid substance from a solution)	Simple and effective; selective	Sediment formation, difficulty in controlling the dose of chemicals and water quality, higher costs
Electrokinetic processes (works on fine-grained contaminated soils)	Inexpensive	Only works on a small portion of the soil at one time
Electrocoagulation (metallic cations directly produced by application of a current between iron electrodes to liquify soluble anodes in the waste treated)	Efficient; easy maintenance; operates with locally available materials	Not indicated for As(III) removal; produces sludge; high energy consumption; addicted to: coagulant form and dose, pH and more competing anion availability

gradient in case of FO; temperature gradient in case of MD; electric potential gradient in case of electromembrane processes). In general, As (V) is removed more effectively than As (III) and, consequently, in certain cases, oxidation of As (III) to As (V) is required before the removal process. Furthermore, some techniques (such as MF and UF) require coagulants

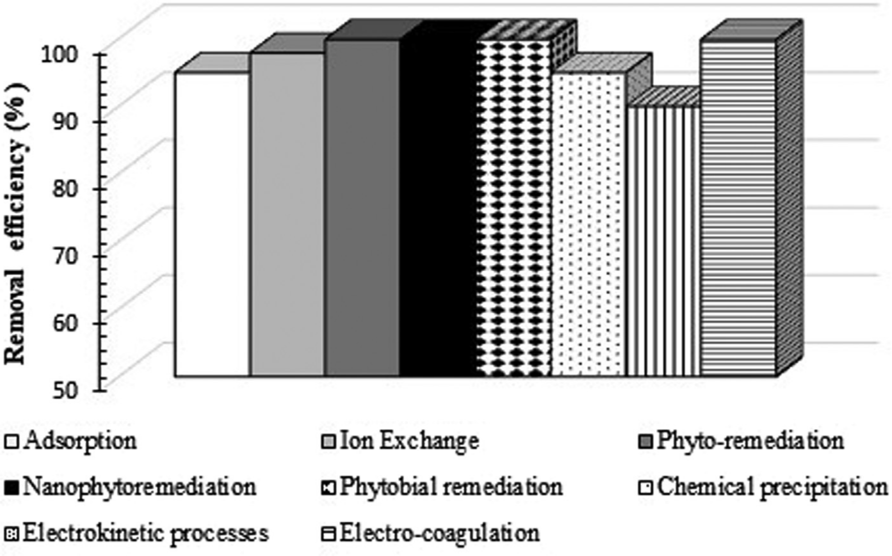

FIGURE 6.1 Removal efficiency of the conventional techniques.

or flocculants in order to increase the size of the particles that enclose As, thus increasing its rejection but producing a secondary solid waste. Others (such as RO) that do not produce solid waste (sludge) require energy input and system costs higher than other technologies (Figoli et al., 2016). In the following, main achievements reported in recent literature by applying membrane operations to the treatment of water contaminated by As are presented and discussed.

6.2.2.1 Microfiltration

Microfiltration is a pressure-driven membrane application very versatile used to remove micrometer-sized matter such as suspended particles, major pathogens, large bacteria, proteins, and yeast cells. The membranes employed in the process have pore diameters that range from 0.1 to 5 μm and, owing to their low hydrodynamic resistance, need low hydrostatic pressures to obtain a high rejection of contaminants and solvent flow. MF finds applications across many areas including wastewater treatment, pharmaceuticals, food, desalination, and biotechnology. Literature data show a wide variety of materials available for this application; early MF membranes were mostly based on cellulose nitrate, but today, the most popular material is poly(vinylidene fluoride) (PVDF). Other most common MF membrane materials include polyethersulfone (PES), cellulose acetate (CA), polypropylene (PP), polysulfone (PSf), and polyamide (PA). Besides polymers, several nonorganic materials (SiO_2, Al_2O_3) are employed, either as monolithic membranes or mixed matrix membranes, with ceramic nanoparticles embedded within a polymer matrix. Recent review papers focus on enhancing MF membrane performances by reviewing MF membrane fouling and addressing the effective challenges in applying MF technology as a pretreatment step or as a stand-alone process for efficient separation processes (Anis et al., 2019). In the specific case of As removal, the membranes generally used in MF are not able to remove As because their pore size is too large with respect to the dimensions of dissolved or colloidal species of As (As(III)/As(V)) (Amy et al., 2000). For this reason, MF is often used in an integrated process and a pretreatment (coagulation or flocculation) of contaminated solution is necessary to increase the particle size of As. About that, Mólgora et al. (2013) have presented a comparative study between electrocoagulation-MF (EC-MF) and chemical coagulation-MF processes (CC-MF) for As removal from drinking water. The best operating conditions were obtained using the CC-MF process. Effectively, after 30 hours of process, the residual As concentration in the permeate has reached a value above the limiting concentration of 10 μg/L recommended by the WHO, but below 25 μg/L, which is the limit set by the Health Ministry of Mexico. Finally, also an economic comparison suggested that CC-MF system performed better than EC-MF one: USD $ 0.066 per m^3 of treated water for CC-MF versus US$ 0.12 per m^3 for EC-MF. L. Pramod et al. (2020) have illustrated MF methodology coupled with heterogeneous Fenton process demonstrating good results in the treatment of water with higher As content. Fenton mode was mainly used to treat wastewater by radical oxidation and flocculation (Xu et al., 2020). The heterogeneous Fenton process was carried out using iron-loaded activated carbon (Fe-AC) catalyst resulting in 90.7% As removal. The study conducted with H_2O_2 alone and catalyst alone (adsorption) led to a lower removal of As than the combined process (adsorption + oxidation). After a subsequent MF step, the residual As content was reduced to 0.105 mg/L.

Bandyopadhyay and Majumder (2018) have presented a study that demonstrates the improvement of the arsenic removal efficiency of a low-pressure membrane system. In particular, they have considered point-of-use system equipped with a particular low-cost ceramic membrane in a hybrid process (adsorption under fluidized condition and crossflow microfiltration). The drinking water produced has respected the WHO recommendations and the flux obtained was 500 LPD (8 hour/day). Table 6.2 reports the specific data cited in the literature.

6.2.2.2 *Ultrafiltration*

Ultrafiltration is a low-pressure-driven membrane process generally used in various chemical and biochemical applications. The separation mechanism is a function of particle dimensions and used to separate dissolved macromolecules or small particles by the UF membranes with pore size ranging between 0.01 and 0.1 μm. UF membranes can be polymeric or inorganic and the polymeric UF membranes are generally made of PSf, PES, PA, CA, polyacrylonitrile, polytetrafluoroethylene (PTFE), PVDF, and PP. Ceramic materials such as alumina and zirconia are usually used as inorganic membrane materials (Figoli et al., 2016). To have significant removal efficiencies, increasing the dimensions of dissolved or colloidal species of As, several authors have proposed to use UF membranes negatively charged (Agarwal et al., 2013), colloid-enhanced UF (Figoli et al., 2016) configurations such as micellar-enhanced UF (if the colloidal species is a micelle-forming surfactant) (De and Mondal, 2012; Rahmanian et al., 2010), polyelectrolyte-enhanced UF (PEUF) (when the colloidal species is a polyelectrolyte), and electro-UF (Weng et al., 2005). Other methodologies and details are reported below. Rivas et al. (2010) have proposed a liquid-phase polymer-based retention (LPR) technique to remove As(III) from aqueous solutions. LPR process removes ionic species by functional groups of water-soluble polyelectrolytes and then using an UF membrane that does not let them pass through the membrane, thus separating them from the solution. In addition, the electrocatalytic oxidation process of As(III) into As(V) was contemplated. The proposed method was able to achieve the complete retention of the As and the results of

TABLE 6.2 Summary of principal results of microfiltration literature works.

Membrane type	Membrane/Module properties	Initial As concentration	Operating conditions	As removal (%)	References
Commercial Pall MF Membrane	Material = PVDF; Configuration = Hollow fiber membrane; Inner diameter = 0.7 mm; Outside diameter 1.3 mm; pore size = 0.1 μm; Filtration area = 0.02 m^2; Module length = 102 mm	As(V) 100 μg/L	(CC-MF) pH = 7.0; concentration of 4.0 mg/L of Fe^{3+}	(CC-MF) 97	Mólgora et al. (2013)

N/A	N/A	1.5 mg/L	pH = 5;
			catalyst = 2500 mg/L;
			$[H_2O_2]$ = 0.014 M; with aeration
				93	Pramod et al. (2020)

Clay-alumina-based ceramic membrane	Single channel 7 element ceramic MF module; Tubular Element (Inner Diameter) = 25 mm; Filtration area = 0.54 m^2; Module length = 1000 mm	Ground water with up to 1.5 ppm As	Filed trial using natural groundwater with up to12 ppm Fe; TMP <1 kg/cm^2	<10 ppb	Bandyopadhyay and Majumder (2018)

tests have shown that the retention capacity of the arsenate depends on pH, counter ion, quaternary ammonium group, and the polymer concentration. Arar et al. (2014) have demonstrated the efficient removal of As species by associating the polymer enhanced UF (PEUF) procedure with the electrocatalytic oxidation (EO) process of As(III) to As(V). The main interest of the research work was the dual function of polymer poly[glycidyl methacrylate N-methyl d-glucamine], P(GMA-NMG) on As removal by combining EO and PEUF processes without any other chemical reagent. The results have shown that it was possible to increase the removal of As up to a level of 80% starting from 60% for As(III) after complete electrooxidation of As(III) to As(V). Varol and Uzal (2015) in their work have used UF assisted with polyacrylamide as an ecological complexing polymer to remove As from aqueous solutions. In case of diafiltration with volume reduction, the authors achieved the complete removal of As. Gohari et al. (2015) have realized a laboratory-made nanocomposite UF membrane to carry out adsorptive studies of As. The tests have shown that the membrane performance was a function of titanate nanotube (TNT) concentration and the best results were achieved at the highest value of TNT corresponding to the highest As(V) adsorption capacity (~125 mg/g). The process was able to generate a permeate of high quality to meet the maximum As level set by the WHO (10 μg/L). Molinari and Argurio (2017) have illustrated that the combination between photocatalysis and complexation-UF (CP-UF) was able to remove As completely from contaminated water. For this purpose, in the CP-UF step, the performance of poly (dimethylamine-coepichlorohydrin-coethylene-diamine) (PDEHED) and poly(diallyl dimethyl ammonium chloride) (poly-DADMAC) have been evaluated and the best operating conditions in terms of pH and polymer/As weight ratio were established at 7.5 pH and polymer/

As ratio of 20 for As(V) removal with PDEHED polymer, 9 pH = 9 and polymer/As weight ratio of 30 in case of Poly-DADMAC polymer. However, if the process operated under the optimal chemical conditions, no As(III) removal was possible. A photocatalytic oxidation of As (III) to As(V) was performed under UV radiation using TiO_2 (0.05 mg/L) as the photocatalyst, O_2 as the oxidant at pH 9. The results have shown that in case of diafiltration with volume reduction, with Poly-DADMAC, the As concentration in the permeate was 15 μg/L. Hao et al. (2018) have presented a combined adsorption – UF technique using an aerated system where synthesized amino-functionalized coffee cellulose adsorbent (PEI-coffee) was employed for As removal from water. The membrane surface has been modified from hydrophobic to hydrophilic to obtain a low As affinity. Furthermore, the adsorption process was optimized by response surface methodology based on central composite design. The experimental results have demonstrated that the aeration not only increased the removal efficiency by oxidizing As(III) to As(V), but mitigated the membrane fouling process. Kumar et al. (Alka et al., 2021; Kumar et al., 2019) have presented a work using CA and cellulose acetate phthalate (CAP) as additives (1, 3, and 5 wt.%) to fabricate UF hollow fiber membranes for the removal of As from drinking water. The hollow fiber membrane prepared by the addition of 5 wt.% of CAP had the best performance: high hydrophilicity, less tendency toward fouling, high water uptake, and increased overall porosity as reported in the literature. Main findings on the performance of UF membranes are presented in Table 6.3.

6.2.2.3 *Nanofiltration*

Nanofiltration is a pressure-driven process that employs the membranes with pore dimensions in the nanometer range operating between UF and RO processes. The special features of NF membrane are high rejections of multivalent ions and organic compounds with a molecular weight of 150–1000 Da and a low rejection for monovalent ions. NF membranes are made of an active layer over a support and the top layer is generally in PA, polypiperazineamide, CA, etc. The excellent removal capacity of contaminants, the lower energy consumption, and enhanced membrane lifetime as compared to RO, the decreasing prices of the membranes, have led to an increased interest in NF applications. Effectively, today, NF is applied by many researchers in environmental field for the treatment of groundwater, surface water, and wastewater reclamation also for the removal of As, persistent organic pollutants, pharmaceutical active compounds, and hormones (Figoli et al., 2016; Mohammad et al., 2015). Padilla et al. (Bryjak et al., 2016; Padilla & Saitua, 2010) presented a work on the performance of simultaneous As, fluoride, and alkalinity (bicarbonate) rejection by a pilot-scale considering groundwater from Argentina. The separation behavior of the investigated membrane corresponded to a typical negatively charged membrane where the main mechanism driving salt rejection is Donnan exclusion. The results have shown that As(V) was rejected to a greater extent than the other ions (89% fluoride and 85% bicarbonate). When the pressure decreases from 7 to 2 bar, a decrease in ions removal was observed, to a less extent for arsenate, in accordance with the so-called dilution effect, which describes that at elevated pressures, the permeate water flux increases more than the salt flux. It is interesting to notice that, due to the lower operational pressure required for the process, a simple bicycle pump can be used for pressurizing the feed, which facilitates the NF application in areas where power supply is limited. In their work, Harisha et al. (Harisha et al., 2010; Obijole et al., 2022) have demonstrated the effective usage of an NF process for the removal of As from drinking water. The specific membrane used can decrease As ion concentration to the level set by the WHO for As-contaminated

TABLE 6.3 Summary of principal results of ultrafiltration membranes on arsenic removal.

Membrane type	Membrane/Module properties	Initial As concentration	Operating conditions	As removal (%)	Flux	References
Commercial PES 10 kDa	N/A	As(V) 30 mg/L	P = 3.5 bar; pH = 8; polymer/As molar ratio = 20	100	N/A	Rivas et al. (2010)
Commercial regenerated cellulose -RC10 kDa	N/A	As(III) 7.5. 10^{-4} M	P = 1 bar; pH = 10; P(GMA-NMG)/As molar ratio = 20	As(V)/after oxidation 80	N/A	Arar et al. (2014)
Commercial regenerated cellulose –RC 5 kDa	N/A	150 μg/L	P = 3 bar; pH = 10; Polyacrylamide/As molar ratio = 2	100	N/A	Varol and Uzal (2015)
Lab-made nanocomposite	Material = polyethersulfone (PES) and titanate nanotubes (TNTs), area = 12.56 cm^2, water permeability ~1250 L/m^2hbar, membrane thickness = 250 μm	As(V) 97.5 μg/L	TNT-PES ratio = 1.5; P = 0.1 bar; T = 25°C; initial feed pH = 6–8	As(V) content in the permeate < 10 μg/L	N/A	Gohari et al. (2015)
Commercial Iris 30 (Tech-Sep)	Flat-sheet configuration, material = PES, molecular weight cut-off = 30 kDa	As(V) 10 mg/L	(poly-DADMAC) P = 2 bar; pH = 9; Polymer/As molar ratio = 0.015; T = 25°C	99.9	N/A	Molinari and Argurio (2017)
Commercial (Tianjin Motian Membrane Eng. & Tech. Co., Ltd. -China)	Hollow fiber configuration; materials = polyethersulfone; pore size = 0.22 μm; dimensions (R × mm) = Ø50 × 386 MWCO = 50 kDa Area = 0.3 m^2I/O diameter = 0.8 mm/1.2 mm	As(III) + As(V) 1 mg/L	Adsorbent dosage = 3.5 g/L; pH = 5.5 and aerated stirring	~90	N/A	Hao et al. (2018)
Lab-made CA-5 PPSU = 14 g NMP = 85.5 g CA = 0.5 g) CAP-5 (PPSU = 14 g, NMP = 85.5 g, CAP = 0.5 g)	Configuration = hollow fiber; material = polyphenylsulfone (PPSU) (with cellulose acetate (CA) and cellulose acetate phthalate (CAP) as additives) CA-5: contact angle = 60.83 degrees; porosity = 26.97%; water uptake = 69.01%; permeability = 61.47 L/m^2hbar CAP-5: contact angle = 43.40 degrees; porosity = 27.96%; water uptake = 77.01%; permeability = 69.60 L/m^2hbar	1 ppm	pH ~6.8	CA-5: 34 CAP-5: 41	(CA/PPSU) 61.47 L/m^2hbar (CAP/PPSU) 69.60 L/m^2hbar	Kumar et al. (2019)

drinking water. The experimental results have shown high fluxes, significant rejection rates of total dissolved solid, and conductivity. In addition, the rejection rate increased when the initial As concentration decreased. A constant flux was achieved throughout an operation time of 180 minutes, which suggested that membrane was not affected by the fouling phenomenon during the process run. However, the efficiency of this process depended upon the pH of water and the presence of other ions. Finally, it is not to be underestimated that the system was very simple to operate, with low requirements for operator attention. As mentioned earlier, As can exist in two forms, As (III) and As (V), and research has shown that it is easier to remove arsenate. For this reason, some authors such as Mou et al. (Obijole et al., 2022; Sen et al., 2010) have carried out tests using membrane-integrated hybrid treatment systems in which a preoxidation step for conversion of trivalent As to pentavalent form was employed. In their work, three different types of NF membranes were compared and $KMnO_4$ was used as oxidizing agent in the preoxidation step. While transmembrane pressure (TMP) was found to have strong impact on both flux and retention of As, pH and preoxidation exhibited strong influence on percentage removal of As. The introduction of preoxidation step increased remarkably the As separation efficiency. When pH value varied from 3 to 10, As rejection increased by 23% for NF-1, 33% for NF-2, and 26% for NF-20 membranes. Figoli et al. (2010) have studied the influence of operating conditions (temperature, TMP, pH, and feed water concentration) on the pentavalent As removal for comparing the performance of two commercial modules. As concentration in the permeate generated by the module with higher flux and removal efficiency (NF90) was always lower than the specific limits imposed in Bangladesh. In addition, as already verified, the efficiency of As removal increased in correspondence with an increase of pH and a decrease of operating temperature. Finally, the permeate flux increased with temperature and pressure. Akbari et al. (Akbari et al., 2010; Figoli et al., 2016) have evaluated the influence of the variations of feed As concentration, TMP, and pH on the As(V) and As(III) removal. When the initial concentration of both As(V) and As(III) increased in feed water, the percentage of rejections decreased. With the increase in pressure, the percentage of rejection of As(V) increased, while it was found to decrease in case of As (III). Saitua et al. (Obijole et al., 2022; Saitua et al., 2011) have evaluated the efficiency and the As removal mechanism using an NF pilot plant from naturally contaminated groundwater rich in ions. The authors, first of all, have established the membrane rejection capacity of individual salt solutions present in the groundwater and subsequently the rejection of every groundwater component considering the influence of pressure. The results demonstrate the importance of ionic composition on ion rejection; ion rejection in multicomponent solutions was found to be significantly different to that of individual salt solutions. Whereas monovalent anion rejections remarkably decreased, divalent cation rejections were three times higher. Yu et al. (2013) have studied the effects of ion concentration and natural organic matter on As(V) removal under different TMPs and considering three typologies of membranes. When the TMP value increased, for all membranes, both As rejection and permeate flux increased; the permeate fluxes decreased after addition of humic acid (HA) to the solutions. The study of Al-Rashdi et al. (2013) highlights that the NF270 commercial membrane was not suitable to remove As(III) from a synthetic water with As_2O_3 because the membrane was a loose membrane and thus, it failed to reject the element. Chang et al. (2014), also, have obtained low As(III) rejection values. The research demonstrates that the feed As(III) concentration and ionic strength had less effect on the filtration flux but a great influence on the As(III) rejection performance of NF. Good results in terms of permeate flux and rejection of As(V) are shown by Pal et al. (2014). The authors have treated groundwater from West Bengal (India) using a membrane-integrated hybrid treatment system developed for continuous removal of As from contaminated groundwater with simultaneous stabilization of As rejects for safe disposal. The work has demonstrated that both trivalent and pentavalent As could be removed by crossflow NF following a chemical preoxidation step for the conversion of trivalent As into pentavalent form. In addition, the used flat-sheet crossflow membrane module showed high flux without the need for frequent replacement of the membranes. Fang et al. (Fang & Deng, 2014) have carried out a study on rejection and modeling of arsenate by NF considering the contributions of convection, diffusion, and electromigration to As transport. In particular, the rejection of arsenate, from synthetic water with $Na_2AsO_4 \cdot 7H_2O$, was analyzed in a crossflow system using two commercial NF membranes. The As(V) rejection of the DK membrane was slightly higher than the DL membrane for all the operating conditions investigated. Song et al. (2016) developed a novel laboratory-made membrane to treat synthetic water, which have registered good results. Gonzalez et al. (Alka et al., 2021; Gonzalez et al., 2019) have presented a work on As removal from geothermal influenced groundwater using a low-pressure NF pilot plant with the scope to produce drinking water in Nicaraguan rural communities. An NF pilot plant powered by solar panels was built and operated in rural community Telica, exposed to As-rich drinking water sources due to geothermal influences. The results of the research have shown that the permeate concentration (~5 μg/L) complied with the WHO guideline for drinking water and the concentrate (~55 μg/L) could be used by local villages for daily activities (e.g., laundry and bathing). Figoli et al. (2020) have treated three samples of natural As(V) contaminated groundwaters comparing the performances of two commercial membranes. NF experiments were conducted at different TMPs (3, 7, 11, and 15 bar) for each groundwater, and the best results, in terms of water flux and As rejection, were registered with HL membrane. Ahmed et al. (Ahmed et al., 2010; Figoli et al., 2016) have considered a particular vibratory shear-enhanced process (VSEP) equipped with two commercially available NF membranes with different pore sizes. The experiments have highlighted that the removal of As(V) by the UTC-70 membrane was only slightly influenced by the variation in operating conditions, whereas the removal of As(V) by the NTR-7450 membrane was strongly dependent on feed water composition, TMP, vibration amplitude, and pH. The removal of As(V) was higher than the removal of As(III) for all the conditions examined, in agreement with the other literature data. More details on the performance of commercial NF membranes are provided in Table 6.4.

Considering the literature data available in which the flux values are mentioned, it is possible to observe that the highest flux of 5000 L/min (120,000 L/m^2h) was obtained with a thin-film composite (TFC) NF-300 membrane made in PA when a stream of As(V) was fed (Harisha et al., 2010; Obijole et al., 2022). In addition, with a PA composite NF-1 membrane, in correspondence of initial As concentration of less than 1 mg/L for both As(III) and As(V), the flux values were around 145 L/m^2h (Pal et al., 2014). In both cases reported using synthetic feeds, As content in the permeate respected the WHO limit. Moreover, when groundwater samples, with a quantity of total As in the range 59.0–435.0 ppb, were processed, the highest

TABLE 6.4 Summary of principal results on arsenic removal using commercial nanofiltration membranes.

Commercial membrane and manufacturer	Membrane/module properties	Initial As concentration	Operating conditions	As removal (%)	Flux	References
NF-300 membrane (Osmonics Inc. USA)	Spiral configuration material: TFC polyamide, nominal MWCO = 180 Da, pure water permeability = 5.5 L/m^{2}hbar	180 μg (As(V))/L, 5 mg (F)/L, 84 mg(HCO$_{3)}$/L	pH = 8; $P = 7$ bar; P = 2 bar	93; 91.6	N/A	Bryjak et al. (2016), Padilla and Saitua (2010)
Thin-film composite (TFC) NF-300 (Permionics, Vadodara, India)	Membrane material = polyamide; area = 2.5 m^2; module length = 40 in; module diameter = 2.4 in	As(V) 0.0005 M, 0.0003 M, 0.0001 M	$P = 50$ bar	98.98;99.82; 99.99	Max value 5000 L/min (initial feed concentration of 5000 mg/L)	Obijole et al. (2022), Harisha et al. (2010)
Thin-film composite NF-1, NF-2 and NF-20 (Sepromembranes Inc.)	Flat-sheet crossflow configuration Membrane material = polyamide, membrane area = 100 cm^2, membrane thickness = 0.0165 cm	150 μg/L	$P = 2$ kgf/cm^2; $T = 35$°C; flow rate = 700 L/h; crossflow velocity = 1.16 m/s; pH = 7	Without preoxidation of As(III): 63 (NF-1); 57 (NF-2); 60 (NF-20); with preoxidation of As(III) ∼98 (NF-2 and NF-20); ∼99 (NF-1)	Without preoxidation of As(III) 40 L/m^{2}h (NF-1) 350 L/m^{2}h (NF-2) 96 L/m^{2}h (NF-3)	Obijole et al. (2022), Sen et al. (2010)
NF90–2540 (Dow-FilmTec) N30F-2440 (and Microdyn- Nadir GmbH)	Spiral-wound membrane modules NF90–2540 Membrane material = polyamide thin-film composite; area = 2.6 m^2; MWCO∼ 200 Da N30F-2440 Membrane material = hydrophilized polyethersulfone; area = 1.7 m^2; MWCO∼400 Da	As (V) 100 ppb	TMP = 12 bar; $T = 25$°C; pH = 8, TMP = 6 bar; $T = 40$°C; pH = 8	NF90–2540 >94, N30F-2440 >78, NF90–2540 95.4, N30F-2440 ∼70	NF90–2540∼83 L/m^{2}h N30F-2440∼59 L/m^{2}h, NF90–2540∼78 L/m^{2}h, N30F-2440∼53 L/m^{2}h	Figoli et al. (2010)
NF90–2540 (DOW-FilmTec)	Polyamide thin-film composite area = 2.6 m^2	As(V) 120 μg/L As(III) 118 μg/L	Feed water discharge = 15 L/min (900 L/h); $T = 27$°C; $P = 6$ bar; pH = 8	As(V) 98.35 As (III) 94.07	N/A	Figoli et al. (2016), Akbari et al. (2010)

(*Continued*)

TABLE 6.4 (Continued)

Commercial membrane and manufacturer	Membrane/module properties	Initial As concentration	Operating conditions	As removal (%)	Flux	References
NF-300 membrane (Osmonics Inc. USA)	TFC polyamide spiral-wound membrane nominal MWCO = 180 Da Area = 1.5 m^2	As(V) $HAsO_4$conc. = 0.428 ppm	$3 < pH < 10$; $T = 293$ K; feed flow = 417 L/h; $P = 10$ bar	>95	($P = 1.0$ MPa) 80 L/m^2h	Obijole et al. (2022), Saitua et al. (2011)
ESNA-1-K1 (Hydranautics)	Material = aromatic polyamide MWCO = 200–300, Water permeability = 2.4×10 $m^3/(s \cdot m^2 \cdot kPa)$, area = 139×10 m^2	As(V) 40 μg/mL	$P = 1$ MPa	68–85,	75 L/m^2h	Yu et al. (2013)
NF270 (DOW, FilmTec)	Material = polypiperazine-based MWCO = 200 Water permeability = 3.8×10 $m^3/(s \cdot m^2 \cdot kPa)$	As(V) 40 μg/mL	$P = 1$ MPa	80–95	100 L/m^2h	Yu et al. (2013)
ESNA-1-LF (Hydranautics)	Material = aromatic polyamide MWCO = 100–150 Water permeability = 1.9×10 $m^3/(s \cdot m^2 \cdot kPa)$	As(V) 40 μg/mL	$P = 1$ MPa	86–98	25 L/m^2h	Yu et al. (2013)
HODRA-CORE (Hydranautics)	Material = sulfonated polyethersulfone MWCO = 1000 Water permeability = 5.9×10 $m^3/(s \cdot m^2 \cdot kPa)$	As(V) 40 μg/mL	$P = 1$ MPa	37–47	130 L/m^2h	Yu et al. (2013)
NF270 (DOW, FilmTec)	Material = polyamide area = 7.6×10^{-4} m^2	As(III) 100 μg/L	$P = 0.30$ MPa $22°C < T < 25°C$; pH = 5	$45 < R < 60$	20 L/m^2h	Al-Rashdi et al. (2013)
DESAL HL (General Electr Co)	Material = polyamide Area = 139×10 m^2	AsIII 50 μg/L-400 μg/L	$P = 0.41$ MPa	$65 < R < 80$	54 L/m^2h	Chang et al. (2014)
NF-1 (Sepro Membranes Inc., USA)	Material = polyamide composite	AsIII ≅ 0.75 mg/L AsV ≅ 0.18 mg/L	TMP = 16 kgf/cm^2 pH = 7.2	$R > 98$	144 L/m^2h–145 L/m^2h	Pal et al. (2014)

DK and DL (GE Osmonics)	DK Two proprietary layers on polyester and polysulfone support; contact angle = 50.2 degrees of DL two proprietary layers on polyester and polysulfone support contact angle = 44.9 degrees; area $\sim 139 \times 10^{-4}$ cm^2	As(V) 100.0 mg/L	$P = 0.55$ MPa	(DL) $86 < R < 95.5$	(DL) 18 L/m^{2}h	Fang and Deng (2014)
Dow NF270–2540 (DOW FILMTEC)	Negatively charged NF membrane Material = polyamide; Area = 2.6 m^2	As (V) = 40.0 µg/L As (III) = 2.6 µg/L	$T = 43°C$; $P = 1.2$ bar	As total ∼90 (As in the permeate ∼5 µg/L)	16 L/m^2 h	Gonzalez et al. (2019)
HL (softening type) and DK (high rejection) (GE Osmonics)	Material = polyamide-TFC $150 < MWO < 300$ Da	Groundwater samples; As(V) (GW1) 59.0 ppb (GW2) 118 ppb (GW3) 435.0 ppb	$T = 16°C \pm 2°C$, flow rate = 9 L/min TMP = 11 bar No oxidation of As(III) made before NF tests	HL (GW1) 96 (GW2) 98 (GW3) 97	HL (GW1) 92.0 L/m^{2}h (GW2) 99.4 L/m^{2}h (GW3) 85 L/m^{2}h	Figoli et al. (2020)
NTR-7450 (Nitto Denko Co. Ltd., Japan) UTC-70 (Toray Industry Ltd., Japan.)	NTR-7450 sulfonated polyethersulfone UTC-70 polypiperazineamide membrane area = 0.045 m^2	As(III) and As(V) 50 µg/L	$0.2 \text{ MPa} < P < 1 \text{ MPa}$	As(V) UTC-70 $95 < R < 99.2$ NTR-7450 $74 < R < 82.5$ As (III) UTC-70 $75 < R < 80.5$ NTR-7450 $13.5 < R < 21.5$	N/A	Ahmed et al. (2010)
Lab-made SPEEK-coated composite membrane based on a commercial polyethersulfone UF hollow fiber membrane	PES UF hollow fiber membrane ID/OD = 0.8/1.3 mm; MWCO = 70,000 Da SPEEK-coated NF Pore size = 1.56 nm; MWCO = 6000 Da	As(V) 360 µg/L	$P = 0.30$ MPa; $T = 25°C$	∼97	32 L/m^{2}h	Song et al. (2016)

flux obtained was 99.4 L/m^{2}h with PA-TFC membrane (HL (Softening type)), with a corresponding As removal of 98% (Figoli et al., 2020).

6.2.2.4 *Reverse osmosis*

Reverse osmosis is a pressure-driven process where a semipermeable membrane is capable of rejecting constituents dissolved in a contaminated stream based on the principles of size exclusion (only particles less than 0.01-μm size are rejected), charge exclusion, and physical−chemical interactions between solute, solvent, and membrane (Malaeb & Ayoub, 2011). Among various materials used for the synthesis of RO membrane, the PA-TFC is the most common due to its excellent water permeability, high salt rejection, and stability. The commercially available modules in which the membranes are lodged are fabricated in spiral-wound and hollow fiber configuration (Hailemariam et al., 2020). Regarding the specific use of this methodology for the As removal from polluted streams, several authors have carried out research achieving excellent results. Akin et al. (2011) have investigated the potentialities of two commercial membranes, purchased from FilmTec, in the treatment of spiked water and natural groundwater sample evaluating As removal percentage for both As (V) and As(III). The study has demonstrated that As removal is function of the operating pressure and pH values; As(V) and As(III) can be effectively removed from water at pH above 4.1 and 9.1, respectively. Moreover, the rejection of As(V) and As(III) did not depend upon the As concentration in the feed water. In addition, a natural (ground) water sample containing 50 μg/L of As(V) and 12 μg/L of As(III) has been treated using spiral-wound element with PA-TFC membrane (SWHR) membrane; the results have shown that total As concentration could be efficiently reduced to 2.86 μg/L. Teychene et al. (2013) have presented a work in which a comparison among different membranes supplied by Hydranautics and the DOW Chemical Company has been realized in case of treatment of synthetic brackish water containing As(III). "Sea water" membranes are found to be more efficient than "brackish water" membranes and this research work also demonstrates that the rejection depends on the pH and TMP applied. In all cases, the rejection of As(III) was greater than or equal to 99%. In addition, the results suggest the investigation on the influence of new membrane parameters on the As rejection (i.e., solute permeabilities). Chang et al. (2014), using a low-pressure RO (LPRO) membrane with aromatic PA selective layer, have obtained rejections lower than the results reported previously (Akin et al., 2011; Teychene et al., 2013) as the specific value did not exceed the 90%. The flux increased with the operative pressure and it would reduce with the increase of the feed ionic strength, due to concentration polarization. The focus of the research of Abejón et al. (2015) was the minimization of costs and energy consumption and for this reason, they used four different types of membranes with the same membrane area to remove As(V) from synthetic water ($Na_2HAsO_4 \cdot 7H_2O$). The "BE membrane" provided the best results in case of arsenate removal from an aqueous solution, as well as the highest solvent permeability and rejection coefficient values. In addition, the study has demonstrated that the concentration of As (V) in the permeate water can be reduced to 0.5 μg/L at a feasible cost. Schmidt, Gukelberger et al. (2016) have presented a study on As removal system considering, as water source, groundwater from two Indian villages: Bind Toli and Ramnagar located in the Patna District in the State of Bihar, India. The process was based on RO technology with an energy recovery system. The experimental tests were conducted with aerated and nonaerated groundwater. Despite the RO process required a relatively higher pressure and, consequently, higher energy demand, the work has demonstrated that using an energy recovery system, this demand can be lowered, leading to an energy demand per liter permeate of 3–4 Wh/L only. The best results were obtained when aerated groundwater was considered: in this case, the As concentration in permeate respected the WHO limit. A double filtration for removal of arsenate and arsenite from drinking water was successfully proposed by Víctor-Ortega and Ratnaweera (2017). The results obtained have shown that after the first filtration, the total removal of As (V) was obtained, while a second filtration was necessary to have a removal of As (III) higher than 80%. In addition, to increase the removal efficiency of As(III), a membrane cleaning step was necessary after the 1st filtration. This suggested that a possible modification of the membrane structure after the 1st filtration, but nevertheless, the methodology led to treated water available to be reused. Table 6.5 summarizes the principal results, in terms of percentage of As removal and permeate flux of the recent RO literature as discussed here.

6.2.2.5 *Forward osmosis*

Forward osmosis is a process where a semipermeable membrane is placed between two solutions at different osmotic pressures. Unlike NF and RO methods, that usually use hydraulic pressure, FO employs as driving force across the membrane an osmotic pressure gradient that is generated between the aqueous feed and a concentrate solution (draw solution), to separate water from dissolved solutes (Bryjak et al., 2016). The membranes employed in this process are made in CA, cellulose triacetate (CTA), TFC-PA/PSf, polybenzimidazole, polyamide-imides, nanoporous PES, etc. (Lutchmiah et al., 2014). Despite the importance of As removal from contaminated water, only few studies have been performed to explore the efficiency of FO for this specific application (Bryjak et al., 2016). Jin et al. (2012) have investigated the influence of membrane orientation (feed solution facing the support layer or the active layer) and organic fouling on the performance of FO membrane in removing boron and As from contaminated waters. Experimental results demonstrated that membrane fouling with alginate caused a decrease in permeate water flux and rejection of inorganic contaminants was a function of the membrane orientation: the contaminants were rejected at a much lower rate when the membrane-active layer was facing draw solution (AL-DS) compared to the active layer-facing feed water (AL-FW) due to the greater influence of internal concentration polarization (ICP) effect in the latter orientation. The research of Cui et al. (2014) on novel FO process for the removal of heavy metal ions from wastewater has successfully demonstrated the potential of the method in the specific case of six heavy metal solutions, that is, $Na_2Cr_2O_7$, Na_2HAsO_4, $Pb(NO_3)_2$, $CdCl_2$, $CuSO_4$, and $Hg(NO_3)_2$. The results have shown that this FO performance outperformed most NF processes and suggested the great potential of the newly developed FO system for the treatment of heavy metal wastewater. Mondal, Hermans et al. (2014) have shown the effects of physicochemical factors on the separation of As from a contaminated stream when $MgSO_4$ and glucose solutions were used as two potential draw solutions. The rejection of As was higher when the membrane-active layer faced the feed solution (AL-FS) compared to the rejection when the membrane-active layer faced the draw solution (AL-DS) and different results were, also, obtained in case of use of $MgSO_4$ or glucose. The As (III) rejection was low at lower pH and oxidation of As(III) (pH = 7) increased the rejection to 95.7%. In another work, Mondal, Tran et al. (2014) with the same apparatus and Jin et al. (2012) have studied the rejection behavior of As(V) in the presence of several co-occurring solutes demonstrating that the increase of As(V) rejection follows this sequence: HA > bicarbonate > nitrate > fluoride > sulfate > phosphate. In addition, the fouling layer formed by HA and the increase in pH due to the presence of bicarbonate helped to enhance the removal efficiency. Yang et al. (2019) have designed and

TABLE 6.5 Summary of results on performance of selected reverse osmosis membranes cited in the literature.

Commercial membrane & manufacturer	Membrane properties	Initial As concentration	Operating conditions	As removal (%)	Flux (L/m^2 h)	References
SWHR BW-30 (FilmTec)	SWHR Flat-sheet membrane; area = 44 cm^2; Salt rejection = 99.6%; (BW-30) Flat-sheet; area = 44 cm^2; Salt rejection = 99.5%	As(V) 800 ppb As(III) 800 ppb	As(V) pH = 4.1 As(III) pH = 3.1 P = 20 mbar T = 20°C	SWHR As(V) ~95, As(III) ~80	11.60 As(V) at 35 bar; 7.07 As(III) at 35 bar	Akin et al. (2011)
(Sea water membranes) SW30HR and SCW5 (Brackish water membranes) BW30LE, ESPAB and ESPA2	Area = 140 × 10 m^2	Synthetic brackish water with $AsNaO_2$ 0.086 mg/L (AsIII)	pH = 7.6; P = 24 bar pH = 9.6; P = 40 bar	SW30HR/ SCW5 - >99, BW30LE/ ESPAB/ ESPA2−99	18.72 (SW30HR), 27.84 (SCW 5),—85.6 (BW30LE), 102.8 (ESPAB), 118.4 (ESPA2)	Teychene et al. (2013)
LPRO (DESAL AK /General Electr Co)	Area = 139 × 10 m^2	synthetic water with $NaAsO_2$ (AsIII, 50 μg/L -400 μg/L)	0.55, 0.82 MPa	$78 < R < 90$	42.48, 64.8	Chang et al. (2014)
AD (GE Osmonics) BE (Woongjin Chem) SW30HR (FilmTec) UTC 80 B (Toray)	Material = polyamide area = 140 × 10 m^2	As(V) 100 ppb	(AD) 1.0 MPa (BE /SW30HR/UTC 80 B) 4.0 MPa	(AD) $94 < R < 96$ (BE / SW30HR/ UTC 80 B) $90 < R < 98$	3.6(AD) 7.56 (BE /SW30HR/UTC 80 B)	Abejón et al. (2015)
SW30−2540 (DOW, FilmTec)	Spiral-wound element with polyamide thin-film composite membrane area = 2.8 m^2	Groundwater Bind Toli (AsIII ≅ 480 μg/L) Ramnagar (AsIII ≅ 67 μg/L)	1.0 MPa	(Bind Toli) 89−99 (Ramnagar) 70−97	25 (Bind Toli) 21.43 (Ramnagar)	Schmidt, Gukelberger et al. (2016)
TW30−4040 (DOW Co)	Polyamide thin-film composite area = 7.2 m^2	spiked tap water $NaAsO_2$: As(III) = 100 μg/L and $Na_2HAsO_4 \cdot 7H_2O$: As(V) = 100 μg/L	1.0 MPa	(after 2nd filtration) As(V): 100 As (III): 95	28	Víctor-Ortega and Ratnaweera (2017)

synthesized a series of novel imidazolium-based ionic liquids via one-step quaternization reactions and grafted these novel compounds on to conventional TFC FO membranes for the treatment of As-containing water. These membranes contained a functionalized selective PA layer grafted with either carboxylic acid/carboxylate or sulfonate groups that was able to enhance membrane hydrophilicity and consequently water permeation. The best results have obtained with $(CH_2)_{23}Na$-membrane. This particular membrane with charged nature can be employed to process other types of wastewater streams containing anionic components. Pham et al. (2020) and coworkers have presented a work which demonstrated that the pH of the solution affects the As removal, as well as the salt concentration in the draw solution influences the water flux. Furthermore, the concentration polarization with respect to surface orientation is very important for As retention; in case of FO process that used the membrane with the active layer oriented toward the feed solution (AL-FS), better results were achieved compared to those obtained using the membrane with the active layer oriented toward the draw solution (AL-DS). A summary of results on recent FO literature presented above is reported in Table 6.6.

6.2.2.6 Membrane distillation

Membrane contactors (MCs) are innovative membrane operation based on the use of a microporous membrane that keeps in contact two phases avoiding their mixing and allowing their contact at each pore interface. Several studies suggest the treatment of aqueous solutions containing As by MCs. MD is an example of MCs, which has been applied to purify water contaminated by As. In the treatment of contaminated aqueous solution using MD processes, the transport of water vapor and of other volatile compounds through a porous hydrophobic membrane is realized, thanks to a difference of partial pressure generated by

1. a cold aqueous stream at the distillate side, in case of direct contact MD (DCMD) (Fig. 6.2),
2. an air gap at the distillate side in case of air gap MD (AGMD) (Fig. 6.3),
3. with vacuum at the distillate side in case of vacuum MD (VMD) (Fig. 6.4)
4. a gas stream at the distillate side in case of sweep gas MD (SGMD) (Fig. 6.5).

Furthermore, in all configurations just cited, the membrane blocks the passage of nonvolatile compounds, such as salts or metal ions. For these reasons, MD processes can be employed to treat contaminated streams by As and other nonvolatile elements, by producing distilled water to reuse (Rahmanian et al., 2010). Membranes with pore sizes ranging from 0.01 to 1 μm can be generally used in MD. The polymers most used are PTFE, PP, PVDF, and polyethylene (PE) (Figoli et al., 2016). Among the different membrane configurations, most of the studies published in the literature are performed using the DCMD configuration. However, the research on AGMD and VMD configurations has significantly increased in recent years. In the following discussion, the results obtained with the different configurations, when applied to the treatment of water contaminated by As, are reported especially in terms of permeate flux and contaminant rejection.

6.2.2.6.1 Direct contact membrane distillation

Abass et al. (2016) have developed a novel system of DCMD integrated with acid-purged zero-valent iron (APZ) technology capable of simultaneous removal and immobilization of As. In this work, the authors have compared the performance of three composite hydrophobic flat-sheet membranes considering a groundwater situated in Datong Basin, (North China) with a total As concentration that varied from 0.025 to 1.8 mg/L. The configuration has shown a maximum As rejection efficiency greater than 90%, and among the membranes tested, PTFE0221 was the most efficient one. In addition, it was possible to

TABLE 6.6 Summary of results on arsenic separation using FO process.

Membrane type	Membrane properties	Initial As concentration	Operating conditions	As removal (%)	Flux	References
Commercial hydration technologies, Inc. (Albany, OR)	Asymmetric structure cellulose triacetate supported by embedded polyester mesh, area = 42 cm^2	1 mM $CaCl_2$, 7 mM NaCl solution; 10 mg(B)/L and 10 mg As (III)/L	Crossflow velocity = 23.2 cm/s, Tfeed = 24°C ± 0.5°C; pH = 6	$60 < R < 95$	–	Jin et al. (2012)
Commercial thin-film composite (TFC) FO	Flat-sheet interfacial polymerization on a macrovoid-free polyimide support and a novel bulky hydroacid complex $Na_4[Co(C_6H_4O_7)_2]0.2H_2O$ (Na–Co–CA) as the draw solute	As(V) 2000 ppm	*T* amb *T* = 60°C	>99.5, 99.7	11 L/m^2h, 16.5 L/m^2h	Cui et al. (2014)
Commercial hydration technology innovation (HTI, Scottsdale, AZ)	Asymmetric Material = cellulose triacetate-active layer with an embedded polyester mesh as the mechanical support contact angle (active layer) = 67.15 ± 0.9-degree contact angle (support layer) = 78.52 ± 2.1-degree plate and frame module 8 × 8 × 0.05 cm (length × width × depth)	As(V) 300 µg/L	*T* = 22.5°C pH draw and feed sol. = 7 $MgSO_4$ concentration = 0.25 M glucose concentration = 0.5 M	AL-FS mode > 98	AL-FS mode ~2.3 L/m^2h	Mondal, Tran et al. (2014)
Lab-made CH_2COOH-membrane CH_2COONa-membrane $(CH_2)_2COOH$-membrane $(CH_2)_2COONa$-membrane $(CH_2)_{23}Na$-membrane	$(CH_2)_{23}Na$-membrane Area = 4.5 cm^2 $\sigma p = 1.21 \pm 0.04$, $\mu p = 0.29 \pm 0.01$ nm, MWCO = 196.3 ± 7.14 Da	As(V) 1000 ppm	vfeed = vdraw solutions = 1.3 cm/s [NaCl] = 0.5 M (draw solution)	>99.5	11.0 LMH	Yang et al. (2019)

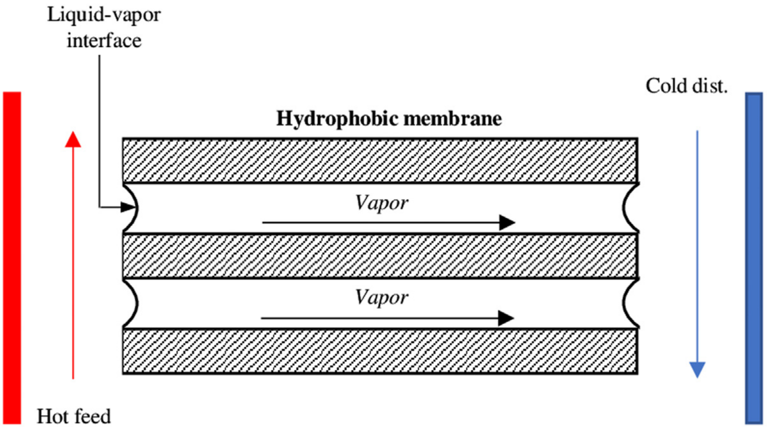

FIGURE 6.2 Scheme of direct contact membrane distillation configuration.

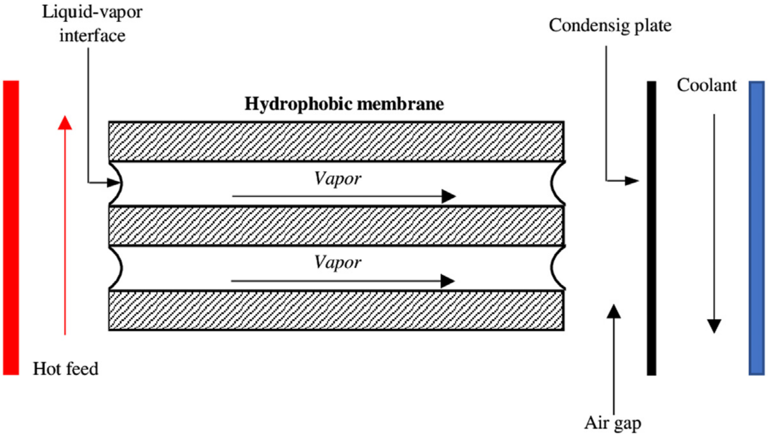

FIGURE 6.3 Scheme of air gap membrane distillation configuration.

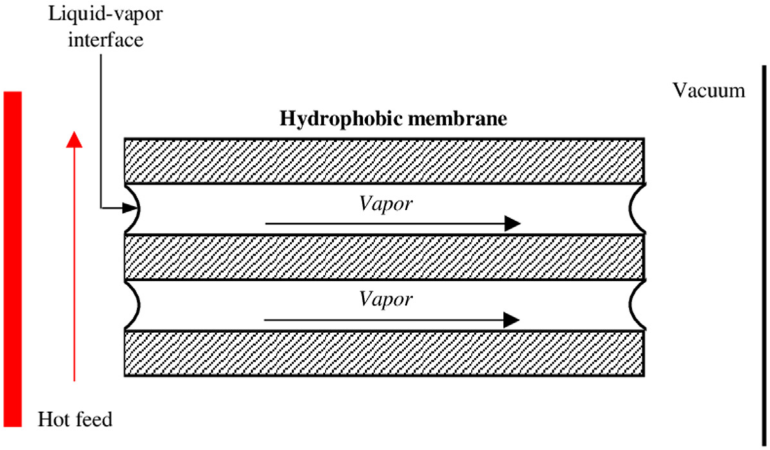

FIGURE 6.4 Scheme of vacuum membrane distillation configuration.

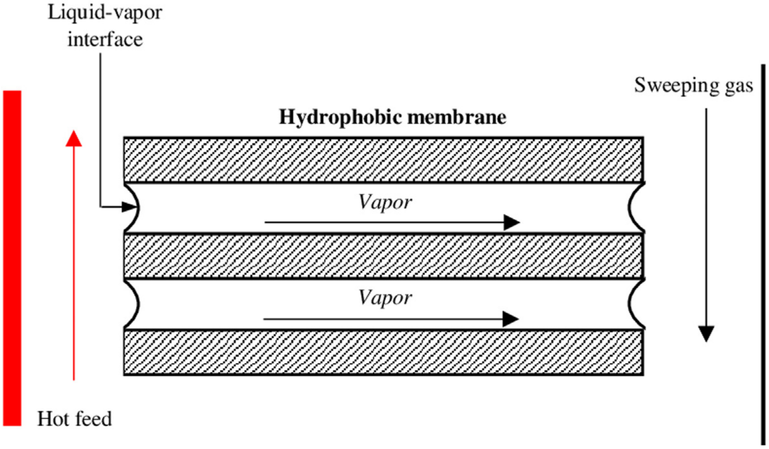

FIGURE 6.5 Scheme of sweep gas membrane distillation configuration.

observe that both species of inorganic As (As[V] and As[III]) can be effectively sequestered under anaerobic conditions without the possibility of secondary release.

Hubadillah et al. (2019) have used DCMD configuration to remove arsenite and arsenate from water using green, silica-based ceramic hollow fiber membranes. The effect of pH on the performance was negligible; in addition, the tested membranes named h-ASHFM and h-CSHFM (with different pore size) had high values of permeate flux (max value ~55.8 kg/m^{2}h) due to a large pore size. Concerning the As rejection, the h-CSHFM, with a pore size of 0.5 μm, showed higher rejection, up to 99.6%, while the h-ASHFM cannot reject As(III) and As(V) enough to respect the imposed limits, due to the larger membrane pore size (1.2 μm), which led to a partial wetting phenomenon. Moreover, the permeate flux decreased when the As concentration increased due to the concentration polarization. Finally, the h-CSHFM membrane was capable of performing at high temperatures of 80°C and for 4-hour operation.

6.2.2.6.2 Air gap membrane distillation

Khan and Martin (2014) have successfully presented a study in which they have tested an AGMD commercial prototype feeding three different feedstocks. The results showed that the tested AGMD prototype was capable of achieving excellent separation efficiency and the total rejection of the contaminant. Leaper et al. (2021) have successfully tested several POSS-functionalized graphene oxide/PVDF electrospun membranes in AGMD configuration demonstrating the possible applications of the methodology in many parts of the world suffering from As-contaminated groundwater. In particular, these membranes were realized by the addition of reduced graphene oxide functionalized with superhydrophobic polyhedral oligomeric silsesquioxane molecules (POSS-rGO) into the spinning solutions. The GP2 membrane (containing 2 wt.% POSS-rGO) has demonstrated the best performances: the flux was 21.5% higher than that of the pure PVDF membrane and almost double that of a commercial PTFE membrane when a prolonged test of 24 hours was carried out.

6.2.2.6.3 Vacuum membrane distillation

Criscuoli et al. (2013) have investigated the potentialities of VMD process for the treatment of water contaminated with As(III) and As(V) to recover clean water at the permeate side. In the work, a comparison among four commercial membranes (M1 and M2, made in PP and M3, M4 made in PVDF) was carried out to establish the best performance in terms of permeate flux and As rejection values. The results obtained have shown that the highest flux was reached with the M4 membrane. All tested membranes were able to reject As and to provide the same performance also after 1 month of continuous use and no wetting phenomena were observed. In addition, the work has highlighted that VMD process was able to efficiently treat, at low feed temperatures, water contaminated by both As(III) and As(V), avoiding the need of the preoxidation step to convert As(III) into As(V); this means reduction of the chemical consumption associated and plant complexity.

The work of Dao et al. (2016) started from the specific As contamination in Vietnam, where drinking water resources present both high As concentration and high salinity (5–15 g/L). About that, direct As(III) removal from brackish groundwater by VMD was proposed and synthetic brackish solutions containing NaCl (10 g/L) and As(III) (at different concentrations – between 300 and 2000 ppb) were fed to the MD module. High rejections were obtained and no effect of organic matter nor calcium on membrane scaling and fouling phenomena was observed at the concentration considered.

6.2.2.6.4 Solar-driven membrane distillation and flash vaporization membrane distillation

Pal and Manna (2010) have investigated a solar-driven MD (SDMD) method for As removal from contaminated groundwater comparing the performances of three flat commercial hydrophobic membranes. The results have shown that the Ms3220 membrane performed better than the others and the analysis of the effect of As feed concentration on flux highlighted that in respect of a variation from 200 to 1200 μg/L, an average of 12% flux decline was registered for all membranes. Furthermore, a total As separation was achieved without wetting membrane pore even after 120 hours of operation. In another work, Manna et al. (2010) have considered the same solar-driven membrane module (Yang et al., 2019) but testing a hydrophobic PVDF MF membrane. Also in this case, excellent results have been obtained with the complete rejection of As. The encouraging results showed that the design could be effectively exploited in the vast As-affected rural areas of Southeast Asian countries blessed with abundant sunlight particularly during the critical dry season.

The solar-driven flat-sheet flash vaporization MD (FVMD) module of crossflow mode proposed by Manna and Pal (2016) was able to remove almost completely the content of As from contaminated groundwater collected from Chakdah (West Bengal, India). In all tests made by modifying different parameters (feed and distillate temperatures, distillate velocity, As concentration in feed, and operating time), no presence of As was found in the permeate. The performance of the membrane in terms of flux and As rejection was the same also after a long period (40 hours) of investigation.

6.2.2.6.5 Forward osmosis-membrane distillation hybrid system

Another work in which it is highlighted that the pretreatment phase of conversion from As (III) to As (V) is not necessary when using MD is that of Ge et al. (2016) who have demonstrated the removal of As(III) from water by an oxalic acid complex $Na_3[Cr(C_2O_4)_3]$-(Na–Cr–OA) using an FO-MD hybrid system. Incorporating MD into FO not only made As(III) removal sustainable by reconcentrating the Na–Cr–OA solution simultaneously, but also reduced the As(III) concentration below 10 μg/L in the product water, meeting the WHO standard. Husnain et al. (2015) have presented an integrated FO and MD process for wastewater reuse. In particular, FO was used as a pretreatment barrier to remove most contaminants in the feed water, whereas MD recovered the draw solutes from FO effluent and simultaneously produced high-quality reusable water.

Table 6.7 summarizes the principal results of recent studies on MD process for arsenic removal.

6.2.2.7 *Supported liquid membrane technology (SLM)*

Another example of MC which was applied for the removal of As from water is SLM. The SLM technology is a feasible and promising separation technique that couples the advantages of liquid membrane with the mechanical resistance of membrane barrier (Güell et al., 2010; Güell, Fontàs et al., 2011; Lothongkum et al., 2011). SLM uses, in general, a microporous membrane with pores filled with solvents. Feed and strip solutions are passed through the membrane on either side. Commonly, used SLM has organic phase in the pores immiscible with the feed and strip solutions. The desired species is extracted by the organic phase from the feed and the strip solution strips the extracted species. Consequently, both extraction and stripping take place simultaneously (Kapoor et al., 2023). Despite these interesting considerations, some practical problems (e.g., system stability and flux) have to be appropriately solved to make the SLM-based separation processes technically and economically attractive in view of large-scale applications (Güell et al., 2010; Güell, Fontàs et al., 2011; Lothongkum et al., 2011). Lothongkum et al. (2011) have worked on the simultaneous removal of As and mercury from natural-gas-co-produced water from the Gulf of Thailand using synergistic extractant via hollow fiber SLM (HFSLM). The results have highlighted the superior performance of mercury removal compared to As by every single extractant. However, by 3-cycle separation, the percentage of As extraction was below the legislation limit. Güell, Fontàs et al. (2011) have carried out a comparison of a SLM containing Aliquat 336 and two different AEM (a common multivalent anion permeable membrane and a membrane with monovalent anion permselective properties) in case of As transport and separation from aqueous stream. The separation process is a function of pH values and at very acidic or very basic pH, worst results were obtained. Furthermore, a quantitative transport after 24 hours was possible employing SLM or multivalent AEM. Finally, $HAsO_4^{2-}$ was the main species responsible for As(V) transport and it was not possible to transport As(III) containing species in reasonably short time intervals, indicating the feasibility of using these membrane systems, especially the SLM system, for the speciation of As(III)/As(V) in water samples. Bey et al. (2010) have presented a work on the removal of As(V) by PVDF hollow fiber membrane contactors using Aliquat 336 as extractant testing membranes made in laboratory with the dry/wet spinning technique at different bore fluid composition and flow rate. In this case, the only extraction step was investigated. The average membrane thickness was in the range 0.19 to 0.40 mm, whereas the mean pore radius and the porosity vary from 0.10 to 0.14 μm and 77.52% to 81.65%, respectively. All the membranes investigated demonstrated good results for As(V) concentration range 20−100 ppm and the highest extraction value was reached after 6 hours by the membrane named 3 C realized with bore fluid composition of EtOH 30% and bore fluid flow rate of 24 mL/min. The results of the research suggested next investigation (i.e., the coupling of extraction and back-extraction) to increase the degree of extraction. Mafu et al. (2014) have developed a hollow fiber SLM (HFSLM) using Aliquat 336 and sodium hydroxide as the extractant and stripping phase, respectively, and optimizing operative conditions as sample pH, acceptor phase pH, type and concentration of acceptor phase solution, stirring rate, and extraction time. The specific application in the treatment of wastewater stream has demonstrated the capacity to preconcentrate and remove both As and selenium with a selectivity coefficient higher than one. More details are given in Table 6.8.

TABLE 6.7 Summarizes the principal results of recent studies on membrane distillation process for arsenic removal.

Membrane type	Membrane/module properties	Initial As concentration	Operating conditions	As removal (%)	Flux	References
Commercial PTFE0221; PTFE0221B; MSPP270045 (membrane solutions LLC -Shanghai, China)	Area = 538×10^{-4} m^2 Composite PTFE0221 material = PTFE; active layer = PTFE; substrate layer = PP; pore size = 0.22 μm; porosity = 82%; thickness = 170 μm; PTFE0221B material = PTFE; active layer = PTFE; substrate layer = PET; pore size = 0.22 μm; porosity = 80%; thickness = 160 μm MSPP270045 active layer = PP; substrate layer = PET; pore size = 0.45 μm; porosity = 75%; thickness = 160 μm	As total 0.5 mg/L	Tfeed = 80°C; feed flow rate = 0.78 L/min: distillate flow rate = 0.86 L/min	As(V) 95%–98% As(III) 90%–94%	55.5 L/m^2h	Abass et al. (2016)
Lab made	Green, silica-based ceramic hollow fiber modified via FAS grafting h-ASHFM: pore size = 1.2 μm h-CSHFM: pore size = 0.5 μm Area = 0.0354 m^2	As(III) 1 ppm As(V) 1 ppm	DCMD Configuration, h-CSHFM Tpermeate = 15°C; Tfeed = 80°C; pH = 7.45; t = 4 h	h-CSHFM As(III) and As(V) ~100	h-CSHFM As(III) 50.4 kg/m^2h As (V) 51.3 kg/m^2h	Hubadillah et al. (2019)
N/A	Material = PTFE pore size = 0.2 μm; area = 0.19 m^2	As-contaminated groundwater (medium concentration, 366 ppb) and As-spiked tap water (medium and high concentrations, 300–1800 ppb)	AGMD configuration T = 80°C	~100	14 L/m^2h	Khan and Martin (2014)
Lab-made GP2	Thickness = 70 μm; mean pore size = 9.80 μm; porosity = 91.9%	As(V) 600 ppb (sodium Arsenate)	AGMD air gap width = 3 mm; Feed flow rate = 750 mL/min; Tfeed = 80°C; coolant temperature = 20°C	>99.9	~28 L/m^2h (over 5 days of continuous testing)	Leaper et al. (2021)

(*Continued*)

TABLE 6.7 (Continued)

Membrane type	Membrane/module properties	Initial As concentration	Operating conditions	As removal (%)	Flux	References
Commercial M4	Support material = PVDF; pore size = 0.2 μm; thickness = 35 μm; porosity = 70%; LEP = 2.5 bar; area = 180 × 10 m^2	As(III) = 0.5 ppm + As(V) = 0.5 ppm	VMD configuration Temperature = 40°C; Reynolds number = 1700 Pvacumm = 10 mbar	100	12.5 L/m^{2}h	Criscuoli et al. (2013)
Commercial	Material = PTFE; pore size = 0.22 μm; area = 5.78×10^{-3} m^2	As(III) 300–2000 ppb	$T = 40°\text{C}$	>98.5%	8 L/m^{2}h	Dao et al. (2016)
Commercial Ms3220; Ms3020; Ms7020 (Membrane Solutions, Shanghai China)	Flat-sheet Ms3220 composite membrane having a thin polytetrafluoroethylene (PTFE)-active layer and polypropylene (PP) support sub layer pore size = 0.22 μm; Porosity = 80%; Thickness = 150 μm Ms3020 composite membrane of PTFE-active membrane layer with polyethylene terephthalate (PET) supporting sublayer pore size = 0.22 μm; Porosity = 80%; Thickness = 175 μm Ms7020 symmetric and isotropic membrane made of pure polypropylene pore size = 0.22 μm; Porosity = 35%; Thickness = 160 μm area = 120 cm^2	394 μg/L	DCMD configuration, $T_{\text{feed}} = 60°\text{C}$; $T_{\text{dist}} = 21°\text{C}$; $v_{\text{dist}} = 0.052$ m/s; $v_{\text{feed}} = 0.062$ m/s	Ms3220–100	Ms3220 50 kg/m^{2}h	Pal and Manna (2010)
Commercial (Sepromembranes)	Flat-sheet crossflow membrane module material = PVDF; nominal pore size = 0.13 μm; thickness = 150 μm; porosity = 70%–75%	300 μg/L < As < 500 μg/L	DCMD configuration $T = 60°\text{C}$; feed flow rate = 0.120 m^3/h	100	95 kg/m^{2}h	Manna et al. (2010)

Commercial (Commercial Membrane Solutions /Shanghai, China)	Composite flat-sheet material = polytetrafluoroethylene (PTFE); pore size = 0.22 μm; porosity = 80%; thickness of composite membrane = 150 μm; active layer (PTFE) thickness = 60 μm; support sublayer (polypropylene) thickness = 90 μm; tortuosity = 1.8; thermal conductivity = 0.076 W/mK	396 ppb	DCMD configuration *T* (Feed) = 70.5°C; T(dist) = 4.4°C; Feed flow rate = 100 L/h; Distillate flow rate = 200 L/h	99	52.94 kg/m^{2}h	Manna and Pal (2016)
Commercial	Material = PVDF; pore size = 0.28 μm	As(III) 1000 ppm	FO-DCMD *T* = 60°C	99	17 L/m^{2}h	Ge et al. (2016)
Commercial FO membrane (Hydration Technology Innovations-Albany, OR) MD membrane (GE Osmonics -Minnetonka, MN)	FO membrane asymmetric structure, Material = cellulose triacetate with an embedded polyester screen, support thickness ~ 50 μm, MD membrane, asymmetric material = polypropylene; pore size = 0.22 μm; thickness = 130–170 μm	As(III) = 80 ppm–202 ppm As (V) = 60–39 ppm	Integrated FO + DCMD *T* = 70°C	As(V), As(III) > 99.9	35 L/m^{2}h	Husnain et al. (2015)

TABLE 6.8 Summary of principal results of supported liquid membrane literature works.

Membrane type	Membrane/module properties	Initial As concentration	Operating conditions	As removal (%)	References
Commercial Celgard	HFSL material = polypropylene; number of fibers = 10,000; module length = 20.3 cm; module diameter = 6.3 cm; porosity = 30%; pore size = 0.05 mm; contact area = 1.4 m^2; area per unit volume = 29.3 cm^2/cm^3; fiber ID = 240 μm; fiber OD 300 μm	As(V) 0.279 ppm	Mixture of Aliquat 336 and Cyanex 471	98	Lothongkum et al. (2011)
Lab-made SLM (Donnan dialysis process) **1.** monovalent anion permselective membrane Neosepta ACS (Tokuyama Soda-Japan) **2.** multi-valent anion permselective membrane, PC-SA (PCA—Polymerchemie Altmeier GmbH—Germany)	PVDF support Thickness = 125 μm; porosity = 75%; pore size = 0.2 μm; impregnated with organic solution (0.5 M Aliquat 336 in dodecane and 4% 1-dodecanol) Neosepta ACS Thickness = 0.121 ± 0.001 mm PC-SA Thickness = 0.111 ± 0.001 mm	As(V) 10 mg/L	pH = 10 pH = 7 Aliquat 336 in dodecane and 4% dodecanol	(SLM) 100	Güell, Fontàs et al. (2011)
Lab-made 3 C	Material = PVDF Configuration = hollow fibers asymmetric structure, with sponge-like layer at the inner surface, and finger-like macro voids at the outer surface Outer diameter = 1.734 mm; Inner diameter = 1.350 mm; Average Membrane thickness = 0.19 mm; Mean pore radius = 0.11 μm; Porosity = 81.06%	As(V) 100 ppm	pH = 6,98; flow rate: 0.47 mL/s; Organic phase: [Aliquat 336] = 30% v/v + Kerosene + 4%v/v octanol, flow rate = 1.4 mL/s; $T = 25°C$; $\Delta P = 0.3$ bar (aqueous side)	Extraction value ~70	Bey et al. (2010)
Lab made	Hollow fiber SLM (HFSLM)	Metal concentrations = 0.1 g/L	Stripping reagent = 0.8 M NaOH Extractant = Aliquat 336 (undiluted); Extraction time = 8 min; Stirring rate = 200 r/min	78	Mafu et al. (2014)

6.2.2.8 *Polymer inclusion membranes*

PIMs are a relatively novel type of self-supporting liquid membranes for the extraction and separation of metallic and nonmetallic ionic species and small organic molecules from dilute aqueous solutions. PIMs entrap a solute-selective extraction reagent (carrier) in a base polymer matrix. When the PIM is in contact with the feed, the extractant reactively couples with the solute of interest and transfers it into or through the membrane. PIMs are simple and cheap to fabricate, which makes them ideal for use in developing economies where As contamination of streams is often a problem (Figoli et al., 2016). Güell, Anticó et al. (2011) have prepared a particular PIM capable of carrying, in specific operative conditions, As species (arsenite and arsenate) from their aqueous solutions. When the membranes did not contain plasticizer (2-Nitrophenyl octyl ether-NPOE and dibutyl sebacate-DBS) and the concentration of the extractant was high, As(V) could be quantitatively separated from As(III) even in the presence of other anions (chloride, phosphate, nitrate, sulfate, and carbonate). Furthermore, the experimental results suggested that the extracted As(V) species was $HAsO_4$, which was further transported across the membrane by a hopping mechanism and that for longer operating times (up to 25 hours), it was possible to transport a substantial fraction of As(III) in addition to the complete transfer of As(V). Based on these encouraging results, it can be concluded that PIM can be considered for the cleanup of contaminated As(III) and As(V) natural and wastewaters. The extraction of As(V) and As(III) at pH 13 by ionic liquid methyltrioctylammonium chloride (Aliquat 336) in dodecane modified with 4% dodecanol was further investigated in liquid−liquid theoretical studies by Güell et al. (2010). The As(V)−Aliquat 336 system was kinetically more favorable than the As(III) system. A numerical analysis of As distribution data suggested the formation of species with 1:2 and 1:3 stoichiometries in the organic phase in both cases. A SLM system (Güell et al., 2010) containing 0.1 M HCl as a stripping solution allowed the transport of As (V) at ppb levels, as well as the separation of As(V) and As(III). A PIM containing Aliquat 336 as carrier has been used by Vera et al. (2018) for the preconcentration of As(V) contained in groundwater samples. The tests were carried out with two typology of polymers used to make the PIM membranes: CTA or poly(vinyl chloride) (PVC). The experimental results have demonstrated that the transport of As(V) did not depend on the polymer used nor on the thickness of the membrane. Govindappa et al. (2022) have proposed, for the first time, the integration of recycled PVC material as a base polymer and benzalkonium chloride (BAC) as an extractant with the scope to study the performance of a new low-cost PIM for the extraction of As (V) from water. The new membrane showed high transportation efficiency values and good resistance to biofouling in case of a continuous run of 100 hours. Researches on PIMs carried out to date have demonstrated their potential for the development of membrane systems for separation in both industry and chemical analysis, but the relatively modest rate of membrane transport continues to be an obstacle to the implementation of this technique in the separation process. The specific data of the cited works are reported in Table 6.9.

6.2.2.9 *Electromembrane processes*

When a membrane method uses ion-exchange membranes and electric potential difference as driving force for ionic species transport, it is named electromembrane process and, in the specific regards: electrodialysis (ED) and its variations as electrodialysis reversal, electro-electrodialysis (EED) and bipolar membrane electrodialysis, electrodeionization

TABLE 6.9 Summary of principal results of polymer inclusion membranes literature works.

Membrane type	Membrane/module properties	Initial As concentration	Operating conditions	As removal (%)	References
Lab made	PIM (based on the polymer cellulose triacetate) containing 52.4% CTA and 47.6%	Spiked tap water As(V)10 mg/L + other ions	pH = 7; stripping solution = 0.1 M; NaCl Aliquat 336/CTA; *t* = 5 h	(transport efficiency) TE = 100%	Güell, Fontàs et al. (2011)
Lab made	PIM (based on the polymer CTA) 52% CTA–48% Aliquat 336	As(V) 100 mg/L	stripping phase = 2 M NaCl; *t* = 5 h	53 < transport efficiency-TE <81	Vera et al. (2018)
Lab made	Low-cost PIM with 10% recycled PVC	As(V) 100 mg/L	benzalkonium chloride (BAC) as an extractant	extraction efficiency 91%	Govindappa et al. (2022)

(EDI), and Donnan dialysis (DD). These processes can provide an efficient removal of toxic metal ions from polluted water with EDI or DD preferably for the treatment of water with low salinity (Bryjak et al., 2016; Figoli et al., 2016). In the preceding years, there has been a growing interest in the mechanisms of arsenate/arsenite transport across anion-exchange membranes (Bryjak et al., 2016; Güell et al., 2011; Velizarov, 2013; Zhao et al., 2010; Figoli et al., 2016) as well as to testing a DD-based pilot plant (Bryjak et al., 2016; Zhao et al., 2012; Figoli et al., 2016). Zhao et al. (2012) have proposed a household Donnan dialyzer using a table salt solution as a working solution. The membrane maintenance or replacement was not necessary because no significant reduction in As(V) removal due to membrane fouling occurred. Replacing the stripping solution every several months was the only required maintenance. The operating cost of this household Donnan dialyzer was less than 0.1 cents per liter of water. Furthermore, the structure was simple and easy to maintain. Finally, it was competitive in terms of sustained usability and could be easily employed in rural areas of developing countries. In another theoretical work, Zhao et al. (2010) have carried out batch DD modeling studies varying the values of pH and with two types of anion-exchange membranes, one homogeneous and another heterogeneous; in case of the homogenous membrane, the arsenate removal efficiency was higher. Velizarov (2013) have pointed out the potentialities of DD for arsenate removal from aqueous solutions, but at the same time, they have highlighted that it was necessary to pay attention to the anion-exchange membrane selection especially if the process is required to be performed under batch operating conditions and when a quantitative recovery of arsenate into the stripping solution was aimed. For this reason, the use of membranes with monoanion-permselective properties was not recommended. The most appropriate anion-exchange membranes allowing for efficient arsenate transport and high recovery are those possessing relatively open structure and low tortuosity of the polymeric matrix as PC-SA.

The ion-exchange membrane bioreactor (IEMB) concept has been extended to the simultaneous transport and chemical precipitation of arsenate. This specific process presented the advantages of leading to a well-balanced treated water (in terms of its ionic composition) and not forming a brine stream that requires further disposal/treatment. Furthermore, IEMB has been used successfully in the removal of bromate, nitrate, and perchlorate, but the extension of the application also to the case of arsenate, chromate, ionic mercury, etc. needed further investigation (Figoli et al., 2016). In general, the limited literature works have demonstrated that, using the DD method, As can be successfully removed from groundwater, but some considerations are necessary, such as the pH of the treated solutions influences the concentration of ionic forms of As, the concentration of stripping electrolyte (ion) should be appropriately chosen because too high concentration decreases the selectivity of the anion-exchange membrane and too low contents decrease the interdiffusion rate of ions. Finally, no suggestion has been given on how to utilize the exhausted stripping solution with a high As content (Bryjak et al., 2016). Table 6.10 shows a summary of principal results of abovementioned studies.

6.2.2.10 Membrane bioreactors

MBRs are a compact technology where an activated sludge process is coupled with a membrane filtration with the scope to process wastewater and recycling. MBRs could obtain high nutrient removal efficiency and complete biomass retention without a secondary clarifier. This advantage has increased the interest towards this methodology. Today, the MBRs in large scale are employed for the municipal wastewater treatment (Meng et al., 2017). Specific polymers are employed for membrane filtration in MBRs: PVDF, polyethylsulfone (PES), PE, and PP. The membrane modules used are the hollow fibers, the plate and frame, and the tubular one. MBRs have two potential configurations that depend on the module: the external, side-stream or pressurized MBR and the immersed or submerged MBR (Figoli et al., 2016). Literature data demonstrate that the removal of As by MBR is generally quite difficult since the As species remain in soluble form and few cases show that the fouling and clogging layers' phenomena negatively affect the performance of the process. Consequently, other processes should be integrated to improve As removal as elaborated B below with the details of some recent works. Bolzonella et al. (2010) and Figoli et al. (2016) have presented research on application of MBR technology for wastewater treatment and reuse in the Mediterranean region with the scope to improve the water availability in semiarid areas. The pilot-scale bioreactor used treated real wastewater using ZeeWeed-500C UF submerged hollow fiber membrane (GE-Zenon, Canada). The stream was processed considering two categories of runs in which the submerged membrane was coupled with the bioreactor (runs MBR1 and MBR2) and the experimental results indicated that the As removal efficiencies for the MBR systems ranged from 0% to 37%. This result was probably due to the particular chemistry of As in wastewater processes. The work of Choubert et al. (Choubert et al., 2011; Figoli et al., 2016) confirmed the difficulty of removing As from domestic wastewater. A secondary treatment stage (activated sludge, biodisk, and MBR) was able to remove most metals (removal rate >70%), with the exception of B, Li, Rb, Mo, Co, As, Sb, and V due to their low adsorption capacities. Yigit et al. (2020) have shown that, with a sulfidogenic anaerobic MBR used for the treatment of acid mine drainage, at high sulfide concentrations, As_2S_3 started to be dissolved, whereas removal efficiency of As was high at low sulfide concentrations. The high value of As removal efficiency was due to the formation of the yellow mineral orpiment and co-precipitation of As with amorphous iron. Salazar et al. (2022) have realized a membrane nanocomposite filters based on poly(vinylidene fluoride-hexafluoropropylene), PVDF-HFP, containing yttrium carbonate ($Y_2(CO_3)_3$), and

TABLE 6.10 Summary of the results of selected studies on electromembrane processes.

Membrane type	Membrane/module properties	Initial As concentration	Operating conditions	As removal (%)	Flux	References
Commercial JAM-I Huanyld (China)	Homogeneous strong base anion-exchange membrane; base membrane = glycidyl methacrylate and divinylbenzene copolymer; membrane thickness = 0.10–0.12 mm	groundwater spiked with 250 μg/L −500 μg/L As(V)	aeration at 4.7 L/min operation in batch mode	>80	Water produced per day = 35 L	Zhao et al. (2012)
Commercial PC-SA Standard homogenous membrane (PCA-Polymerchemie Altmeier GmbH/Germany)	flat-sheet area = 11.3 cm^2; thickness = 90–130 μm	As(V) 7.5 mg/L	pH = 4.8; $t = 24$ h	As recovery in the stripping solutions = 94	N/A	Velizarov (2013)

TABLE 6.11 Summary of principal results of membrane bioreactors literature works.

Membrane type	Membrane/module properties	Initial As concentration	Operating conditions	As removal (%)	Flux	References
Lab made	Double sided flat-sheet polyethersulfone (PES) UF membrane modules immersed in the AnMBR (Anaerobic Membrane Bioreactor); pore size = 0.2 μm	As(V) 2.5 mg/L	$T = 35°C \pm 2°C$ sulfide conc. <50 mg/L	~99	~10 L/m^2h	Yigit et al. (2020)
Lab made	Nanocomposite membranes based on 10 wt.% Y@Fe_3O_4/PVDF-HFP prepared with 5 wt.% of $Y_2(CO_3)_3$ and 5 wt.% of Fe_3O_4 NPs contact angle = 126 degrees Young modulus = 60 MPa	5 mg/L	contact time = 24 h pH = 4; flow rate = 10 L/h	adsorption efficiency As (III) ~ 33 As(V): 70	N/A	Salazar et al. (2022)

magnetite (Fe_3O_4) to adsorb neutral and anionic species of As(III) and As(V), in an upscaled membrane reactor. The experimental results have demonstrated that the optimal conditions, in terms of maximum adsorption capacities [101.9 and 212.8 mg/g for As(III) and As(V)], were obtained at lower rates and acidic pH. Furthermore, tests in the upscaled membrane reactor showed that the membrane was able to remove As in the presence of other contaminants in the stream with efficiencies of 21.9% and 51.8% for As(III) and As(V), respectively. After membrane regeneration and utilization cycles, a total removal of As(III) and As(V) was achieved after 7 and 3 days, respectively. Table 6.11 reports some results on arsenic removal using MBRs.

6.3 Scope for further work and concluding remarks

The removal of As from contaminated streams is necessary to minimize health hazard especially in areas where water scarcity is a very serious problem. For this purpose, various membrane processes can be considered; however, it is difficult to select the best technique, because each of them has both advantages and drawbacks, and often their efficiency is affected by the type of water (As concentration, the presence of interfering species, pH, etc.) to be treated, as pointed out in Criscuoli and Figoli (2019) who compared the performance of NF and RO to that of MD. Membrane operations, with respect to conventional technologies, are able to treat As polluted waters without using chemicals for regeneration nor producing toxic sludges/solid wastes to be disposed, but the high costs associated with the production of water for reuse continue to limit their large-scale implementation. Table 6.12 summarizes the main performance and improvement proposals in As removal reported in the literature as discussed in this chapter. All the investigated configurations allow to obtain high rejection values, and in some cases, high distillate fluxes demonstrate the potential of membrane processes as a very efficient technique for the purification of contaminated waters. Most membrane operations are more effective in treating feeds containing As(V) and need preoxidation to convert arsenite in arsenate, whereas MD is capable to reject both As(III) and As(V) avoiding chemical consumption, but with high thermal energy demand and lower fluxes than RO. Literature data have highlighted the potential of membrane technologies in the treatment of arsenic-contaminated streams, providing at the same time, useful perspectives for addressing the future research. For example, in UF, the combination between photocatalysis and complexation-UF (CP-UF) allowed to obtain an effective removal of As from polluted waters, thanks to the photocatalytic oxidation of As(III) to As(V) (Molinari & Argurio, 2017). In case of RO process, the research has to be mainly focused on solving fouling problems to avoid the decrease of permeability and selectivity of RO membranes. The accumulation of foulants on the membrane surface could be decreased by chlorine addition, but at the same time, chlorine can react with membrane material reducing its performance (Hailemariam et al., 2020). Therefore, appropriate cleaning agents and protocols must be identified. The application of a novel FO process for the removal of heavy metal ions from contaminated water led to better results than those obtained with NF, and further developments and optimizations of the technique are needed to assess its potential (Cui et al., 2014). Among MD configurations, VMD was able to reject both arsenic forms also at low feed temperatures (Khan & Martin, 2014). However, long-term tests to reduce the wetting risk, as well as the reduction of the specific thermal energy consumption, are needed for future implementations. Using solar energy to reduce the thermal energy demand and a new FVMD module operating in crossflow mode and in DCMD configuration, it was possible to remove almost completely the content of As from contaminated groundwater (Dao et al., 2016). Therefore, the coupling of MD with renewable energies is one of the tasks to be considered and optimized, together with the development of membranes specifically tailored for this thermally-driven membrane process. Therefore, considering the above observations and the details reported in the table, it can be concluded that, despite the great potential shown by membrane processes in the treatment of As-contaminated water, further research is needed aimed at improving the specific properties of membranes, optimizing the operating conditions of the processes, reducing energy consumption, and using integrated systems and hybrid techniques. A summary of the performance in As removal and scope of improvement is reported in Table 6.12.

TABLE 6.12 Summary on performance of membranes in As removal and scope of improvements.

Membrane process	Primary requirements and performance in As removal	Scope of improvement proposed
MF	– Need of pretreatments (coagulation/flocculation) to increase the As particle size – Need of preoxidation of As(III)	– Chemical coagulation coupled with MF (CC-MF) – Addition of Fe^{3+} – Use of either clay or bentonite as a coagulating aid or an organic flocculant to reinforce the solidity of the flocs – Use of iron-loaded activated carbon (Fe-AC) as a heterogeneous catalyst in the Fenton process – Adjustment of pH values
UF	– Need of pretreatments (colloidal species such as micelle-forming surfactants and polyelectrolytes) to increase the As particle size – Need of preoxidation of As(III)	– Use of UF membranes negatively charged, colloid-enhanced UF (CEUF) and electro-UF (EUF) – Use of polyacrylamide as an environmentally friendly complexing polymer – Increase of pH values – Use of nanocomposite UF (UF) membrane consisting of polyethersulfone (PES) and titanate nanotubes (TNTs) – Coupling photocatalysis and complexation-UF processes – Adsorption – UF process using synthesized amino-functionalized coffee cellulose adsorbent (PEI-coffee)
NF	– High rejection of As(V) – Low removal rate for As(III) – Need of preoxidation of As(III) – Most of the water is permeated through the membrane and most divalent solutes are retained instead of only the target trace contaminant(s) – High electric energy consumption – Need of membranes with resistance to compression under higher operating pressure	– Increase of pH values – Decrease of operating temperature – Use of negatively charged membrane – Integration with renewable energy
RO	– Need of pretreatments – High rejection of As(V) – Need of preoxidation of As(III) – Most of the water is permeated through the membrane and most of the solutes are retained instead of only the target trace contaminant(s) – High electric energy consumption – High operative pressure – Need of membranes with high resistance to compression	– Increase of pH values – Increase of operating pressure – Two RO steps (double filtration) – Use of energy recovery systems – Integration with renewable energy – Addressing the fouling problems
FO	– High rejection of As(V)	– Membrane orientation – Use of novel draw solutions

(*Continued*)

TABLE 6.12 (Continued)

Membrane process	Primary requirements and performance in As removal	Scope of improvement proposed
	– Freshwater extraction and draw solution regeneration can be an energy-intensive step	– Use of thin-film composite (TFC) FO membrane grafted with novel ionic liquids
MD	– High rejections of both As(III) and As(V) – Preoxidation step not necessary – Capability to treat water containing different types of pollutants by a single separation technique – High thermal energy demand and lower fluxes than RO	– Improvements in membrane characteristics to reduce the temperature polarization phenomena and enhance water fluxes (e.g., localized heating) – Use of hydrophobic green, silica-based ceramic hollow fiber membranes with big pores (derived from agricultural rice husk) – Use of electrospun polyvinylidene difluoride (PVDF) membranes enhanced by the addition of functionalized graphene oxide – Integration with acid-purged zero-valent iron (APZ) technology – Integration with RO/FO – Integration with renewable energy
SLM	– High extraction of As(V) – Separation efficiency depending on the type of extractant – Low system stability	– Optimization of pH conditions – Development of novel extractants
PIMs	– High extraction of As(V) – Separation efficiency depending on the type of extractant – Lower mass transfer rates than comparable solvent extraction (SX) systems	– Improvement in membrane transport characteristic by using other base polymers – Use of membranes that do not contain plasticizer – Use of a novel polymer inclusion membrane from recycled polyvinyl chloride
Electromembrane processes	– Good removal of As(V) – Not applicable in case of water of very low salinity and co-existing ions, removal of low molecular weight neutral noncharged compounds from the water is desired.	– Sustainable development and manufacturing of ion-exchange membranes with improved properties in terms of their exchange capacity, selectivity and operational stability – Development of integrated (hybrid) processes aiming at minimization of the volume and ecotoxicity of the brine streams – IEMB optimization and application also for arsenate removal
MBRs	– Usually low As removal	– Integration with other processes – Use of sulfidogenic anaerobic MBR – Use of specifically developed membranes, like the membrane nanocomposite filters based on poly (vinylidene fluoride-hexafluoropropylene), PVDF-HFP, containing yttrium carbonate ($Y_2(CO_3)_3$) and magnetite (Fe_3O_4)

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