

## Arsenic removal from water by nanofiltration membrane: potentials and limitations

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### Abstract

Arsenic, in addition to being a confirmed carcinogen, is one of the most toxic elements found in nature, and should therefore be removed if the concentration is greater than 10 µg/L. Nanofiltration (NF) membranes have succeeded in arsenate As (V) ions removal from water almost completely. It is reported in this review that, like reverse osmosis (RO) membranes, NF membranes have not yet performed alone arsenite As (III) ion rejection without being associated with another technology. Commercial NF membranes exhibited a rejection between 86 and 99% towards arsenate As (V) while As (V) removal reached 99.8% for synthesized NF membranes. Since commercial NF membranes have shown their limit, scientists have prepared novel NF membranes that demonstrated long-term efficiency, fouling reduction, cost reduction, an increase in separation of multivalent ions, rejection performance, or a high flux achievement, depending on the area of use. For small treatment plants, NF is a more cost-effective method. The review succinctly reported arsenic as a serious global contamination issue and focused on novel nanofiltration processes for arsenic rejection to safeguard water security. This article also developed a comparative study of nanofiltration and reverse osmosis techniques concerning arsenic removal. Finally, future trends and perspectives have been highlighted with particular emphasis on emerging synthesis techniques of NF membranes without hiding the unpleasant fouling issue that limits its competitiveness.

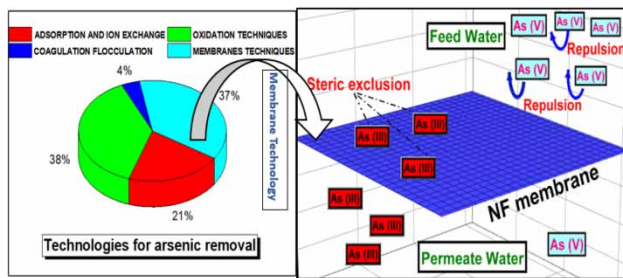
**Key words:** arsenate, arsenite removal, commercial nanofiltration membrane, drinking water, nanofiltration membrane preparation, nanofiltration process

### Highlights

- Commercial nanofiltration (NF) membranes for arsenic removal.
- Novel synthesized nanofiltration membranes.
- Potential and limitation of nanofiltration membranes in arsenic removal.
- Influence of operating parameters on arsenic removal.
- Performance comparison of reverse osmosis (RO) and nanofiltration (NF) membranes in the context of arsenic removal.

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## Graphical Abstract



## INTRODUCTION

Arsenic (As), in both surface and groundwater, has been found above the drinking water standards in different countries around the world (Alsayed & Ashraf 2020). People are living under the dire threat of arsenic toxicity (Chakraborti *et al.* 2013; Chowdhury *et al.* 2016), in Cambodia (Buschmann *et al.* 2008), in Inner Mongolia and China (Wade *et al.* 2009; Xia *et al.* 2009), in Bangladesh, West Bengal (Von Ehrenstein *et al.* 2006), and some other parts of northern Mali and Zambia in Africa (Amini *et al.* 2008), India (Krishna *et al.* 2009), Thailand (Ning 2002), Mexico (Armienta & Segovia 2008), Iranian Kurdistan (Mosaferi *et al.* 2008), El Salvador, Peru, Nicaragua (Bundschuh *et al.* 2008), northern Afghanistan, Eastern Croatia (Ćavar *et al.* 2005), and Vietnam (Buschmann *et al.* 2008). Arsenic (As) is a natural metalloid that shows great mobility in the environment. Mobility highly depends on the parent mineral form, the mechanisms of mobilization, and the oxidation state (Al-Abed *et al.* 2007). In terms of oxidation state, As could exist in four (4) different forms; which are the inorganic forms, arsenite As (III) and arsenate As (V), and the organic forms, arsenic As (0) and arsine As (-III). Among these arsenic species, the most common forms that are usually found in water are the inorganic As (III) and As (V) (Pous *et al.* 2015). Arsenic (As) is reputed to be very toxic to all forms of life (Singh *et al.* 2015). However, the most toxic of the four is arsenite and their toxicity decreases in order *Arsenite As (III) > Arsenate As (V) > Arsenic As (0) ≥ Arsine As (-III)*, as classified by the World Health Organization (WHO) in the first group of human carcinogenic substances (Van Halem *et al.* 2009). Several studies have been carried out on the fate and behavior of arsenic (As) in the environment due to numerous cases of arsenic pollution in the world and the hazards related with it. Diseases have arisen from chronic ingestion of inorganic arsenic present at concentrations up to 50 µg/L in drinking water, especially different types of skin lesions including hyperpigmentation and hyperkeratosis, and cancers (of skin, lung, kidney, and bladder). These diseases are referred to as arsenicosis (Nicomel *et al.* 2016). Hence the urgent need to develop appropriate technologies for treating arsenic-contaminated water.

Among the current arsenic (As) removal technologies, nanofiltration membranes (NFMs) have emerged as a promising technique for high-quality drinking water production and wastewater treatment. The perm-selectivity of the membranes was discovered in the early eighteenth century (Dumont & Cole 2014). However, the industrial development of membrane techniques dates back only to the 1960s for dialysis and 1970 for solvent transfer techniques. Designated for dialysis, the operation is to cross the membranes by a liquid, separating the constituents by diffusion. The solvent transfer operation consists, on the other hand, travel through semi-permeable membranes by a liquid, by forced convection, in order to purify the solvent (Pontié & Bedioui 2002). Membrane technology is a promising area for arsenic removal from water. Different types of membranes such as reverse osmosis (RO), nanofiltration (NF), ultrafiltration (UF), microfiltration (MF) are used for arsenic removal. Nanofiltration technology is between reverse osmosis and

ultrafiltration. The nanofiltration process allows the separation of components having a size in solution close to that of a nanometer (10 Å), hence its name (Figoli *et al.* 2010). Nanofiltration (NF) membranes are usually applied to separate multivalent ions from monovalent ones. However, it is also possible to achieve a certain separation of ions of the same valence by selecting the proper membrane and operating conditions (Lhassani *et al.* 2001). Often called 'Loose' RO membranes, because they provide higher water flux with lower trans-membrane pressure, NF membranes are usually asymmetric and negatively charged in neutral and alkaline media. A large number of studies have been carried out on the elimination of arsenic in water by NF membranes and almost all of them have shown promising results.

He, Tang *et al.* have prepared three water-stable metal-organic framework (MOF) UiO-66 nanoparticles with different diameters, which were synthesized and incorporated into a selective layer to form thin-film nanocomposite (TFN) membranes (He *et al.* 2017). These membranes were tailored to remove arsenic concurrently. The arsenic rejection was also studied by using three NF membranes HL-4040F1550, 4040-UHA-ESNA, and NF70 4040-B (Brandhuber & Amy 1998). It was found for all three membranes that As (III) rejection was 53% and the As (V) rejection was as high as 95%. Moreover, a commercial loose polyamide TFC membrane, NF-45, removed 10–20% of As (III). However, the removal of As (V) was 90% (Vrijenhoek & Waypa 2000). According to Urase *et al.* from the University of Tokyo (Urase *et al.* 1998), ES-10, a new low-pressure aromatic polyamide NF membrane, was able to reject 87–93% of As (V) and 50–89% of As (III) (Sato *et al.* 2002), used three commercial NF membranes (SE-10 (aromatic polyamide), NTR-7250 (polyvinyl alcohol), and NTR-729HF (polyvinyl alcohol)) to investigate arsenic removal efficiency from groundwater and synthetic water. The arsenite and arsenate released in both water types were almost the same, proving a negligible ionic composition effect of water source on arsenic rejection. Using a spiral wound TFC polyamide membrane (192-NF300), As (V) removal from the surface and synthetic water was studied by Saitua H. *et al.* (Saitúa *et al.* 2005). For retentate arsenic between 100 and 382 µg/L, 93–99% of As (V) was rejected by 192–NF-300. Waypa *et al.* used NF-70 to remove Arsenic from synthetic freshwater (Waypa *et al.* 1997). For both As (III) and As (V), 99% rejection could easily be performed with this commercial membrane. NFc and NF270 from Filmtec company were also used to investigate the arsenic removal from natural groundwater. The As (V) rejection was 96% for NFc and 99% for NF270. Recently, He *et al.*, have prepared a novel TFC-NFM consisting of a zwitterionic copolymer for arsenic removal. The results obtained for arsenate rejection were very interesting because they reached 99.8%.

At first sight, in this review, we succinctly present arsenic as a serious global contamination issue. Next, we discuss the current technologies worldwide used to remove arsenic from waste and drinking water. Membrane techniques are subsequently developed, with particular emphasis on novel nanofiltration membranes (NFMs) fabrication for efficient arsenic rejection. The operating parameters that come into play and influence the performance of nanofiltration membranes have been the subject of special development. Finally, a comparative study of nanofiltration and reverse osmosis techniques is made in relation to arsenic removal, and future perspectives are also developed.

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## ARSENIC, A SERIOUS GLOBAL CONTAMINATION

Arsenic is the twentieth most abundant element in the earth's crust (Matschullat 2000) and the forty-seventh most abundant element on earth among the eighty-eight existing elements (Losey & Vaughan 2006; Lee *et al.* 2017). It is a metalloid in the constitution of more than 245 minerals (Smedley & Kinniburgh 2002; Mandal *et al.* 2007). However, it is distributed over all the terrestrial compartments heterogeneously but is naturally concentrated in certain geological zones constituting the geochemical anomalies (Wedepohl 1995; Matschullat 2000; Smedley & Kinniburgh 2002). The first cases of

arsenic-induced skin injury were identified in Bangladesh in the 1980s and about two decades later, a methodological guide applied to the management of sites and soils contaminated with arsenic was drawn up by a group of experts (Laperche *et al.* 2003). Acute exposure to high concentrations of arsenic is known for its poisonous properties. Considering the high toxicity of arsenic, the World Health Organization (WHO) and USEPA set a maximum acceptable level of arsenic in drinking water at 10 µg/L (Nerenberg *et al.* 2002). Arsenic is especially found in the form of oxides, sulfides, or salts of iron, manganese, copper, and sodium, which are very toxic and represent a serious environmental and health issue worldwide (Singh *et al.* 2007). The toxicity of arsenic depends on the form in which it is presented (Navarathna *et al.* 2019; Siddique *et al.* 2020). Organo-arsenicals are less toxic than inorganic arsenic (As) compounds, and pentavalent arsenate As (V) is less toxic than trivalent arsenite As (III). It was reported that the oxyanion forms, As (III) and As (V) compounds, are especially lethal to the environment, and living creatures. Not only the toxicity, but also the mobility of arsenite As (III) are higher than those of arsenate As (V) due to the neutral charge, which decreases the probability of adsorption of As (V) in mineral surface (Sun *et al.* 2014; Wong *et al.* 2016; Navarathna *et al.* 2019). The main routes of arsenic contamination include respiration, oral, and dermal routes. And as a logical effect, contaminated soil, agricultural and fish products could also be the source of arsenic (As) exposure. The organoarsenic main route of contamination is through the ingestion of contaminated marine plants and animals. The health, especially of children, could be seriously affected by the deliberate ingestion of arsenic (As) (Saha *et al.* 1999). The main information on arsenic is summarized in Table 1, whereas Table 2 presents the stabilities of arsenic species (arsenite and arsenate) as a function of pH and oxidation-reduction.

**Table 1** | Main information on arsenic

Atomic number		33
Atomic mass (g.mol <sup>-1</sup> )		74.9216
Sublimation point		613 °C
Density		5.73
Atomic configuration		[Ar] 3d <sup>10</sup> 4s <sup>2</sup> 4p <sup>3</sup>
Valences		-3, 0, +3, +5
Ionic radius (Å)	As (III)/As (V)	0.58/0.46

**Table 2** | Stability of arsenic species (Zambrano 2012)

Reducing conditions		Oxidizing conditions	
pH	As (III)	pH	As (V)
0–9	$H_3AsO_3$	0–2	$H_3AsO_4$
10–12	$H_2AsO_3^-$	3–6	$H_2AsO_4^-$
13	$HA_2O_3^{2-}$	7–11	$HA_2O_4^{2-}$
14	$AsO_3^{3-}$	12–14	$AsO_4^{3-}$

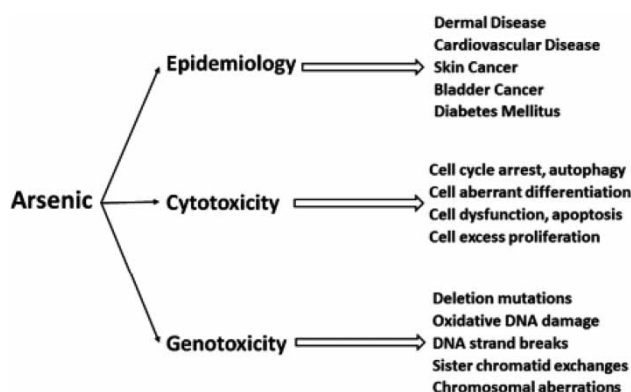
Groundwater and surface waters from several countries are contaminated with arsenic. The areas most affected by this sensitive phenomenon are listed in Table 3 (Petruševski *et al.* 2007).

It was demonstrated that As (III) can be transported into cells via aquaglyceroporins (proteins that are permeable to glycerol as well as water), while As (V) entry develops through phosphate transporters. It has been investigated that following entry into human cells, As (V) rapidly reduces to As (III). Subsequently, arsenite As (III) goes through a multistep pathway of arsenic metabolism in cells via

**Table 3** | Regions contaminated with arsenic around the world

<b>Asia</b> (BGS 2001; Petrusevski <i>et al.</i> 2007; Petrusevski <i>et al.</i> 2008)	Bangladesh, Cambodia, China (including provinces of Taiwan and Inner Mongolia), India, Iran, Japan, Myanmar, Nepal, Pakistan, Thailand, Vietnam
<b>America</b> (Hofstra <i>et al.</i> 1999; BGS 2001; Bhattacharya <i>et al.</i> 2006; Petrusevski <i>et al.</i> 2007)	Alaska, Argentina, Chile, Dominica, El Salvador, Honduras, Mexico, Nicaragua, Peru, USA
<b>Europe</b> (Hofstra <i>et al.</i> 1999; Petrusevski <i>et al.</i> 2007)	Austria, Croatia, Finland, France, Germany, Greece, Hungary, Italy, Romania, Russia, Serbia, United Kingdom
<b>Africa</b> (Petrusevski <i>et al.</i> 2007)	Ghana, South Africa, Zimbabwe
<b>Pacific</b> (Petrusevski <i>et al.</i> 2007)	Australia, New Zealand

arsenite methyltransferase (AS3MT) choosing S-adenosylmethionine (SAM) as the methyl donor, which results in the formation of methylated-As compounds including monomethyl-arsonic acid (MMAV), dimethyl-arsinic acid (DMAV), monomethyl-arsinous acid (MMAIII), and dimethyl-arsinous acid (DMAIII) (Khairul *et al.* 2017; Wang *et al.* 2020a, 2020b, 2020c). Biomethylation of inorganic arsenic, especially the processing of trivalent methylated metabolites, is the mechanism that sets As as a carcinogen and toxin (Singh *et al.* 2007; Pullella & Kotsopoulos 2020). The toxicity of various arsenic species increases in the order of As (V) < MMAV < DMAV < As (III) < MMA (III)  $\approx$  DMAIII. Schematic representation of arsenic toxicity in human health is depicted in Figure 1.

**Figure 1** | Arsenic toxicity in humans (Sun *et al.* 2014).

## NF MEMBRANES AND OTHER TECHNOLOGIES FOR ARSENIC REMOVAL

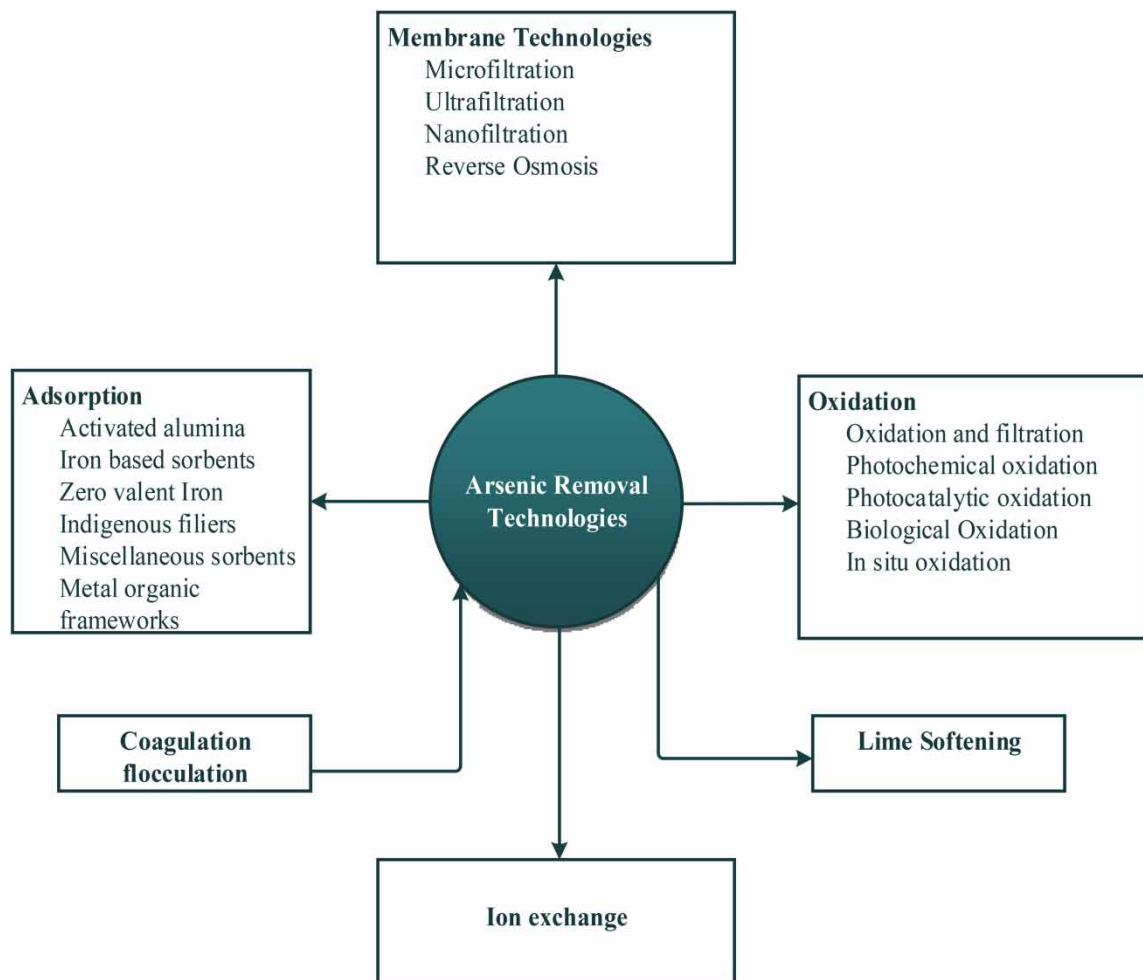
### Technologies for arsenic removal

The chemistry and initial composition of arsenic-contaminated water are the major factors that determine the arsenic (As) removal (Singh *et al.* 2015). Most techniques used to remove arsenic from water are efficient just for As (V) because As (III) is predominantly non-charged at pH below 9.2 (Brandhuber & Amy 1998; Uddin *et al.* 2007; Xia *et al.* 2007; Sen *et al.* 2010; Nicomel *et al.* 2016).

Arsenic (As) is not degraded but just separated from water or converted from an aqueous state to another state, appropriately to solid. Arsenic rejection from streams of water is a matter of serious research due to its severe harmful effect on the body. For more than a decade, arsenic has been the object of attraction in conferences, debates, and publications. Developing a technology that is

less expensive, efficient, and easily usable by a large number of people, were, among other things, the objectives that scientists set for themselves. Thus, five technologies have emerged in their large grouping: ion exchange (Lee *et al.* 2017), adsorption (Ungureanu *et al.* 2015), coagulation/flocculation (Pallier *et al.* 2010; Nidheesh & Singh 2017), oxidation (Bissen & Frimmel 2003), and membrane techniques (Yoon *et al.* 2009).

In Figure 2, current technologies used worldwide for arsenic removal are shown and in Table 4, the advantages and disadvantages of these techniques are detailed.



**Figure 2** | Current technologies for arsenic removal.

For drinking water production, membrane technology is suitable for arsenic (As) rejection and other contaminants contained in streams of water. Membranes are synthetic materials with billions of pores that act as selective barriers. Thus, some constituents of water are not able to pass through the membranes (Shih 2005; Luther *et al.* 2012). A transmembrane pressure (TMP) playing the role of the driving force is required between the feed side and the permeate side (Van der Bruggen *et al.* 2003; Bottino *et al.* 2009). The low-pressure membranes include microfiltration (MF) and ultrafiltration (UF) membranes, whereas high-pressure membranes are nanofiltration (NF) and reverse osmosis (RO). A study of the evolution of the number of publications on the different arsenic removal methods was made and the results are shown in Figure 3(a) and 3(b). Besides, it appears that nanofiltration membranes have aroused more attraction over the past ten years; indeed, the studies carried out on this type of membrane were far above the total studies carried out on all other membranes (MF, UF, and RO), Figure 3(c). In order to acquire data about the number of publications, *Science Direct*

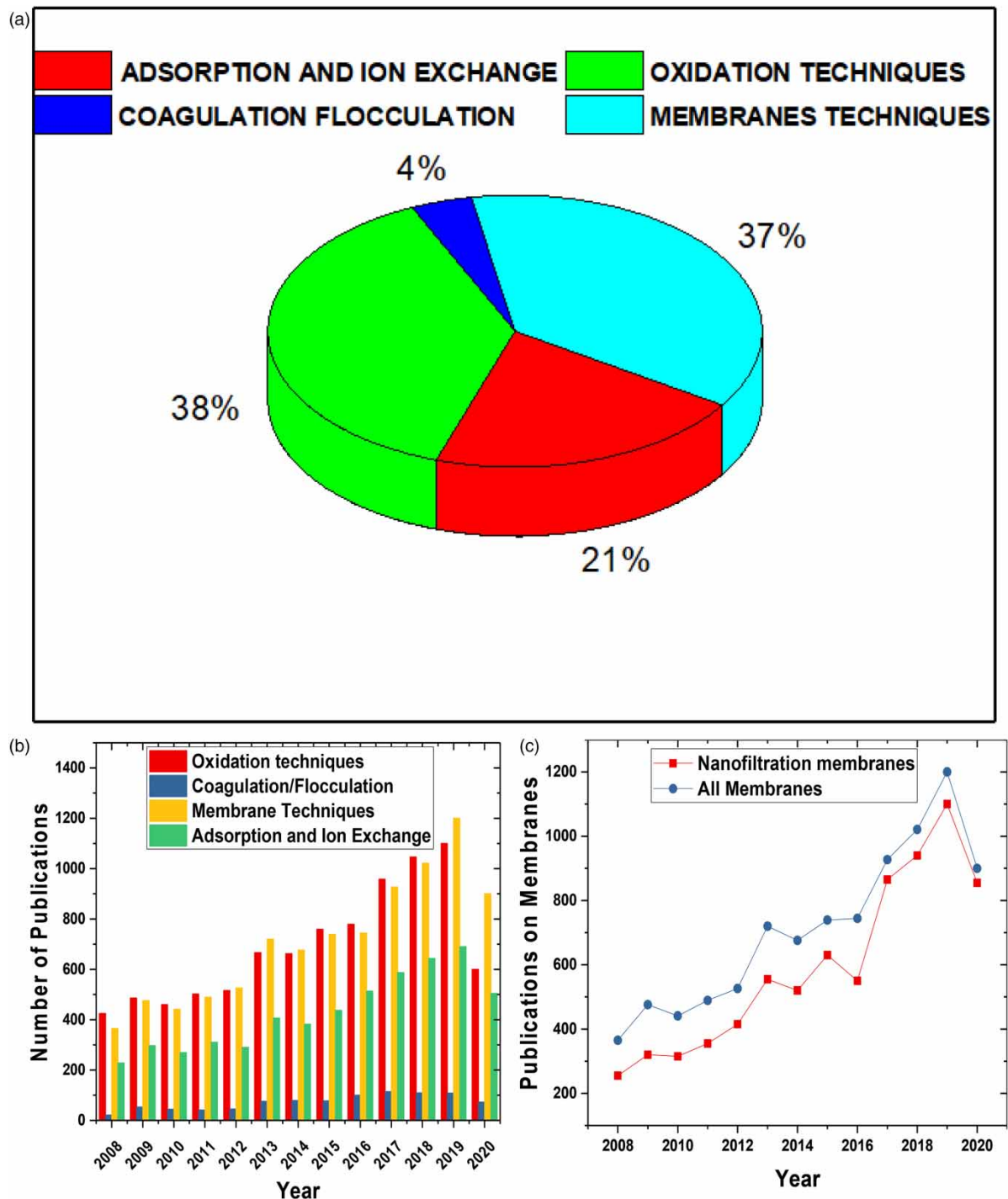
**Table 4** | Advantages and disadvantages of the current technologies for arsenic removal

Arsenic removal technology	Advantages	Disadvantages	Arsenic rejection	References
Coagulation	✓ Wide range of pH	<ul style="list-style-type: none"> <li>✓ Pre-oxidation of arsenite required.</li> <li>✓ High arsenic-contaminated sludge production.</li> <li>✓ Expensive process.</li> </ul> Additional filtration is required.	>90%	Choong <i>et al.</i> (2007); Mohan & Pittman (2007); Sahai <i>et al.</i> (2007); Malik <i>et al.</i> (2009); Hu <i>et al.</i> (2012); Asmel <i>et al.</i> (2017); Sun <i>et al.</i> (2018)
Ion exchange	<ul style="list-style-type: none"> <li>✓ pH-independent process</li> <li>✓ Only efficient for arsenite removal.</li> </ul>	<ul style="list-style-type: none"> <li>✓ Only efficient for arsenite removal</li> <li>✓ Expensive process</li> <li>✓ Low capacity</li> <li>✓ Sludge disposal problem.</li> <li>✓ Resin needs to be replaced again and again</li> </ul>	95	Mohan & Pittman (2007); Laatikainen <i>et al.</i> (2016); Sarkar & Paul (2016)
Oxidation	<ul style="list-style-type: none"> <li>✓ Low operation cost</li> <li>✓ Works on a large pH range</li> </ul>	<ul style="list-style-type: none"> <li>✓ Very slow process</li> <li>✓ Drinking water has a bad smell and color in addition to chlorine, permanganate ...</li> <li>✓ Sludge formation.</li> </ul>	>95%	Lee <i>et al.</i> (2003); Ghurye & Clifford (2004); Leupin & Hug (2005); Dodd <i>et al.</i> (2006); Nriagu <i>et al.</i> (2007); Sahai <i>et al.</i> (2007); Li <i>et al.</i> (2010); Neppolian <i>et al.</i> (2010)
Adsorption	<ul style="list-style-type: none"> <li>✓ Low cost</li> <li>✓ Ease of operation</li> </ul>	<ul style="list-style-type: none"> <li>✓ pH, effective surface area, and the nature of the adsorbent need to be maintained</li> <li>✓ Arsenite cannot be removed very well</li> <li>✓ Post-filtration required</li> <li>✓ Organic matter, other salts in water decrease the efficiency of the process</li> <li>✓ Removal of the generated heavy flocs are difficult</li> </ul>	>100	(Singh & Pant (2004); Mohan & Pittman (2007); Banerjee <i>et al.</i> (2008); Martinson & Reddy (2009); Khosa <i>et al.</i> (2013); Vadahanambi <i>et al.</i> (2013); Sarkar & Paul (2016); Chowdhury (2017); Hao <i>et al.</i> (2018); Siddique <i>et al.</i> (2020)
Membrane techniques	<ul style="list-style-type: none"> <li>✓ Easy operational</li> <li>✓ Technique</li> <li>✓ High arsenate removal efficiency</li> <li>✓ No sludge production</li> </ul>	<ul style="list-style-type: none"> <li>✓ Membrane fouling</li> <li>✓ A little expensive</li> </ul>	>99	Johnston <i>et al.</i> (2001); Chwirka <i>et al.</i> (2004); Ghurye <i>et al.</i> (2004); Shih (2005); Choong <i>et al.</i> (2007); Sarkar & Paul (2016)

*Journal* has been the major focal point using key words in the title and summary of the publications. Although this method is not exhaustive and accurate, it nevertheless has irrefutable effectiveness. On the other hand, [Figure 3\(b\)](#) shows in detail the evolution in number of publications from 2008 to 2020 in these different technologies.

### Rejection mechanism of arsenic through NF membranes

Nanofiltration (NF) membranes are commonly used to separate multi-valent ions from monovalent ions and show great potential in As (V) rejection from contaminated water. Commercial NF

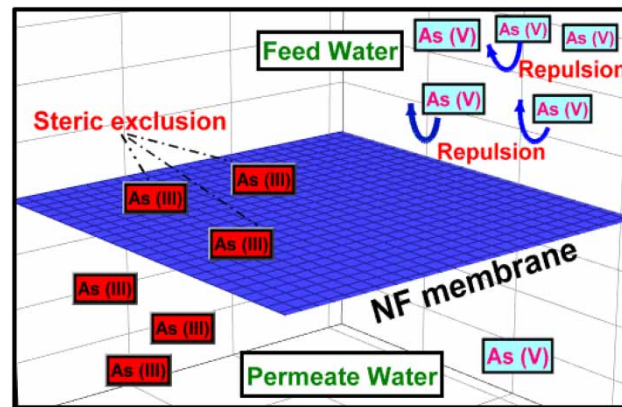


**Figure 3** | (a) Technologies for arsenic removal; (b) number of publications per year in these different technologies; (c) publications on nanofiltration membranes in comparison with other membranes (Data from *Science Direct*, October 31, 2020).

membranes have different levels of water purification and arsenic rejection. Most of the commercial NF membranes have a fixed surface charge, which is developed by dissociation of surface groups (e.g. sulfonated or carboxyl acids) and allow ion separation by a combination of the electrical effect, the pore size, and ion interaction mechanisms (Figure 4).

The NF rejection mechanism is reported to be extremely complex and dependent on both interfacial and micro-hydrodynamic events that occur at its surface, even within its nanopores (Worou *et al.* 2021a, 2021b). Donnan, steric, transport, and dielectric effects combined determine the NF





**Figure 4** | Separation mechanism of As through NF membrane as per pore size and Donnan exclusion.

membranes removal performance (Donnan 1995). The famous Donnan effect makes clear not only the equilibria but also the potential interactions between the membrane surface and the charged species inside the feed solution (Donnan 1995; Lv *et al.* 2016). Neutral solutes transport is performed via the steric mechanism, which is a size-based exclusion, and was already well settled through various studies on ultrafiltration (UF) membranes (Deen 1987). NF membranes' charge originates from their surface functional groups, which ionize, and also from the structure of their pores (Hagmeyer & Gimbel 1998; Ernst *et al.* 2000). These functional groups could be basic, acidic, or indeed both, depending on the nanoparticles (NPs) materials utilized during the preparation process. It was demonstrated that the pH of the solution strongly influenced the dissociation of such surface groups, since the membrane, at a specific pH value, may display an isoelectric point (Childress & Elimelech 1996). In addition, Afonso *et al.* have established that nanofiltration membranes show a weak ion-exchange ability, and even on some occasions, ions from feed solution can adsorb to the NFM surface, generating a modification of membrane charge (Afonso *et al.* 2001). The fixed membrane charge and ion valence in solution in contact with the membrane determine the electrostatic attraction or repulsion force that appears on the localized ionic surroundings as a direct consequence of the aforementioned dielectric phenomena. The dielectric exclusion phenomena are not well understood yet, since there are two competing hypotheses that try obscurely to explain the exact nature of the interaction that takes place. The first hypothesis is called the 'image forces' phenomenon (Yaroshchuk 1998; Nicomel *et al.* 2016), while the second is called the 'solvation energy barrier' mechanism (Bowen & Welfoot 2002). These two exclusion phenomena were investigated in detail (Oatley *et al.* 2012), and it was shown that they are a fruit of both nano-length scales and extreme spatial confinement that occur during the NFMs separations mechanism. It was also reported that solutes moving freely in the feed solution were disrupted by the dragging forces exerted by the solvent passing by the confined pores' structure. The transport of this solute across the nanopores structure is highly affected by the local environment and is credibly considered as hindered. Since it is understood that the dimensions of the NFMs layer are close to atomic length scale, the limitations in available measurement technologies, the knowledge of NFMs surface properties and structural characterization has been delayed and resulted in a significant debate (Schäfer 2005), and the dielectric exclusion phenomenon is especially contested (Wong *et al.* 2016).

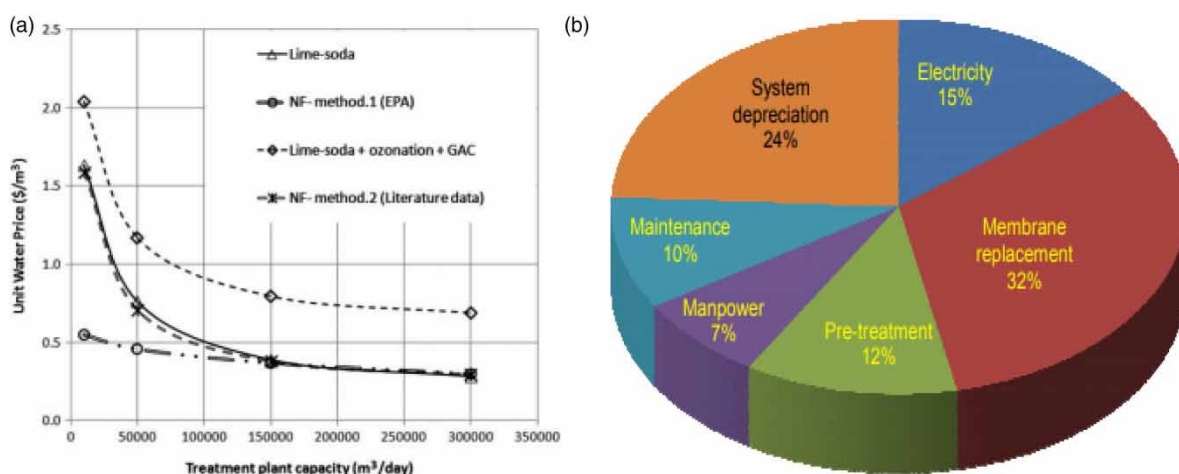
Currently, there are satisfactory results available to conclude the existence of a porous active film in most NFMs. To achieve the characterization of these pores, three main methodologies are used: (i) the gas adsorption/desorption method or Brunauer-Emmett-Teller (BET) technique (Wang *et al.* 2013; Fang *et al.* 2014), (ii) indirect measurement of pore size that consists of the use of neutral solute removal investigation and models (Oatley *et al.* 2013; García-Martín *et al.* 2014), (iii) reverse surface impregnation (RSI) combined with transmission electron microscopy (TEM) (Mohammad *et al.*

2015), and (iv) atomic force microscopy (AFM) for direct measurement of pore size, surface roughness, and topography (Misdan *et al.* 2015).

### Cost evaluation of NF membranes in drinking water

NF membranes are currently applied in water softening (Ghizellaoui *et al.* 2005), even in color removal and disinfection by-product (DBP) precursors (Lin *et al.* 2007; Chellam *et al.* 2008; Sobhani *et al.* 2012), mostly when the rejection of monovalent ions is inconsiderable and membranes operate at relatively low pressure in comparison with RO membranes. Nanofiltration is nowadays preferred over reverse osmosis due to a more dilute concentrate waste stream, and the permeate water generated requires less stabilization in order to minimize distribution system corrosion (Bellona 2014; Shahmansouri & Bellona 2015). It was reported that the NF process is the most appropriate technology for both hardness and organic rejection although it is not always the most economical technology (Ferndale 2013). It was also demonstrated in several studies that NF is an effective technology for drinking water treatment at large facilities (Cyna *et al.* 2002; Nidheesh & Singh 2017); nevertheless, it exhibits major seasonal fouling events due to changes in organic matter properties and microbial activity (Her *et al.* 2007; Xu *et al.* 2018). NF membranes have also been used for specific chemicals rejection including fluoride and pesticides (Ghaemi *et al.* 2012; Shen & Schäfer 2014; Zeng *et al.* 2016; Qin *et al.* 2020), various emerging organic contaminants (Zinadini *et al.* 2017; Bagheripour *et al.* 2018), inorganic carbon (Owusu-Agyeman *et al.* 2017; Gholami *et al.* 2020), DBP precursors (Lin 2013), heavy metals (Hedayatipour *et al.* 2017; Peydayesh *et al.* 2020), oxyanions (e.g. bromate, perchlorate, phosphate, sulfate) (Zhou *et al.* 2016), and arsenic (Xu *et al.* 2017; He *et al.* 2018a; Siddique *et al.* 2020; Zeeshan *et al.* 2020).

For cost comparison, different options for drinking water treatment have been investigated. To achieve this purpose, the costs of water units were calculated for three different processes: (i) lime and soda ash were applied; (ii) lime and soda ash + ozone injection + granular-activated carbon (GAC) for color removal); (iii) NF membranes were applied. For NF membranes' cost evaluation, two different methods were applied including the EPA technique, and the results are depicted in Figure 5(a). This investigation led to two main conclusions. For smaller treatment plants, NF is a more cost-effective method, as reported in the literature (Ferndale 2013). If highly colored water is being treated, NF membrane systems produce water at a cheaper price compared to technologies using ozonation, lime soda, and granular-activated carbon (GAC).



**Figure 5** | (a) Water cost for drinking water production. Life time: 25 years, costs: 2013 USD; design flow rate to average flow rate ratio = 1.7 for capacity > 50,000 m<sup>3</sup>/day and 2 for capacities < 50,000 m<sup>3</sup>/day. (b) Cost break-down of the NF membranes in arsenic removal from drinking water. The total cost for producing drinking water is 0.15 \$/m<sup>3</sup> (Song *et al.* 2015).

In a recent study performed by Song *et al.*, using a novel NF membrane constituted of a sulfonated poly (ether ether ketone) coated hollow fiber, a cost break-down of the membrane in arsenic removal from drinking water has been established (Figure 5(b)). A preliminary cost-estimation for arsenic rejection from water source was set assuming a working time of 300 days/year and a treatment capacity of 1,000 m<sup>3</sup> /hour. The total cost of producing drinking water was 0.15 \$/m<sup>3</sup>.

## COMMERCIAL AND NOVEL SYNTHESIZED NANOFILTRATION MEMBRANES FOR ARSENIC REMOVAL

Commercial and novel synthesized nanofiltration membranes exhibited high performance towards As (V) removal. Due to the imperious and specific needs of chemical removal from water streams, new membranes are more and more manufactured. Therefore, besides the availability of commercial nanofiltration membranes (Table 5), membranes are modified by a sol-gel deposition approach, *in situ* formation method, or other techniques for high flux permeate or excellent pollutant rejection (Table 6). Among the feats achieved in the fabrication of new membranes, there are (i) long-term efficiency of membranes, (ii) membrane fouling reduction, (iii) cost reduction, (iv) increase in separation, (v) rejection performance, and (vi) high flux achievement.

**Table 5** | Arsenic rejection (%) by commercial NF membranes

Model	Supplier	Water origin	Rejection (%)		References
			As (III)	As (V)	
NF70 4040-B	FILM Tec (Dow Chemical)	Surface water	53	99	Katsoyiannis <i>et al.</i> (2002)
NF90		Ground/surface water	50	90	Yoon <i>et al.</i> (2011)
HL-4040F1550	DESAL	Surface water	21	99	Katsoyiannis <i>et al.</i> (2002)
4040-UHA-ESNA	Hydranautics	Surface water	30	97	Katsoyiannis <i>et al.</i> (2002)
NF45	FILM Tec	Synthetic water	10	90	Levenstein <i>et al.</i> (1996)
ES-10	Nitto-Denko Co.Ltd	Groundwater	50–89	87–93	Vrijenhoek & Waypa (2000)
ES-10	Nitto Electric/ Industrial Co.Japan	Synthetic water	80	97	Urase <i>et al.</i> (1998)
NTR-729HF	Nitto Electric/ Industrial Co.Japan	Synthetic water	21	94	Urase <i>et al.</i> (1998)
NTR-7250	Nitto Electric/ Industrial Co.Japan	Groundwater	10	86	Urase <i>et al.</i> (1998)
192-NF300	Osmonics Inc.	Pure water Volcan river	– –	93–99 95	Sato <i>et al.</i> (2002); Ergican <i>et al.</i> (2005)
NF1 + pre-oxidation	Sepro membranes, Inc.	Ground/surface water	63	98	Sen <i>et al.</i> (2010), Pal <i>et al.</i> (2014)
NF2	Sepro membranes, Inc.	Ground/surface water	57	96	Pal <i>et al.</i> (2014)
NF20	Sepro membranes, Inc.	Ground/surface water	60	96	Pal <i>et al.</i> (2014)
NF1	Sepro membranes, Inc.	Ground/surface water	–	>98	Pal <i>et al.</i> (2014)
NF70	FILM Tec	Freshwater	99	99	Sato <i>et al.</i> (2002)
NF40		Ground/surface water	15	>90	Hering & Elimelech (1996); Oh <i>et al.</i> (2000)
NF270	FILM Tec (Dow Chemical)	Groundwater	–	99	Saitúa <i>et al.</i> (2005)
NFc	FILM Tec (Dow Chemical)	Groundwater	–	96	Spiegler & Kedem (1966), Saitúa <i>et al.</i> (2005)

**Table 6** | Novel nanofiltration membranes synthesized or modified for arsenic rejection

Membrane	Background	Arsenic Removal/Remarks	Year	References
Fabrication of thin-film nanocomposite nanofiltration membranes incorporated with aromatic amine-functionalized multiwalled carbon nanotubes. Rejection performance of inorganic pollutants from groundwater with improved acid and chlorine resistance.		Arsenate removal > 98%	2020	Gholami <i>et al.</i> (2020)
Polyamide intercalated nanofiltration membrane modified with biofunctionalized core-shell composite for efficient removal of arsenic and selenium from wastewater		Arsenate removal > 98.2%	2020	Zeeshan <i>et al.</i> (2020)
Integration of two-stage nanofiltration with arsenic intermediate chemical precipitation		Efficient removal of arsenate As (V)	2019	Andrade <i>et al.</i> (2019)
Removal of Arsenic (III) from water using magnetite precipitated onto Douglas fir biochar		Efficient removal of Arsenite As (III)	2019	Navarathna <i>et al.</i> (2019)
The P[MPC-co-AEMA] co-polymer was incorporated into the polyamide selective layer of thin-film composite (TFC) membranes	Pioneering work that may deliver valuable insights to design next-generation NF membranes for heavy metal removal in water treatment.	The novel copolymer modified TFC membranes show improved pure water permeability (PWP) and rejections for both pollutants due to their reduced pore sizes, higher hydrophilicity. It exhibits the highest PWP of 8.5 LMH/bar and impressive rejections of 99.8% toward $\text{HAsO}_4^{2-}$ . It finally proves robust durability under 180-h testing.	2018	He <i>et al.</i> (2018a)
Sodium-ion modified carbon quantum dot (Na-CQD) incorporated thin-film nanocomposite (TFN) membranes	This work may provide useful insights into the preparation of nanoparticle modified polymeric membranes for water purification.	The novel TFN membrane containing 0.05 wt% of Na-CQDs exhibits enhanced pure water permeability (PWP). The new Na-CQD modified Thin Film Nanofiltration membranes display the highest PWP of 10.4 LMH/bar and impressive rejections of 99.5% toward $\text{HAsO}_4^{2-}$ .	2018	He <i>et al.</i> (2018b)
Hydrophobic kaolin hollow fiber membranes (h-KHFM via grafting with fluoroalkyl silane (FAS) molecules)	Low-cost kaolin was utilized in the fabrication of the ceramic hollow fiber membrane.	A high permeate flux of 28 kg/m <sup>2</sup> h for As(III) and 25 kg/m <sup>2</sup> h for As(V) with 100% As removal was reached under feed temperature of 60 °C and it has met the standards-setting the maximum limit of the contaminant at 10 ppb.	2018	He <i>et al.</i> (2018b), Hubadillah <i>et al.</i> (2018)
UiO-66 incorporated thin film (TF) nanocomposite membranes	The effect of particle loading was also studied for the 30 nm UiO-66 TFN membranes. (The best performance)	The TFN membrane containing 0.15 wt% UiO-66 has the highest PWP of 11.5 LMH/bar and remarkable rejections of 96.5%, 97.4% and 98.6% toward $\text{SeO}_3^{2-}$ , $\text{SeO}_4^{2-}$ and $\text{HAsO}_4^{2-}$ , respectively. Higher rejections are also achieved in the concurrent removal of Se and As. Also, the new membrane shows strong long-term stability.	2017	He <i>et al.</i> (2017)

Performance of nanofiltration and reverse osmosis membranes for arsenic removal from drinking water	Four types of thin-film polyamide nanofiltration (NF) (NF270, commercial; NFTG20, synthesized) and reverse osmosis (RO) (XLE, BW30) membranes were used in a flat-sheet module.	It was found that the percentage of As (III) removal of RO membranes were in the range of 97–99% for all transmembrane pressure applied. In the range of operating conditions, As (III) and As (V) rejection were found almost equally good by RO membranes.	2016 Elcik <i>et al.</i> (2016)
Novel nanofiltration membrane with ultrathin zirconia film as a selective layer	Hydrolyzed poly-acrylonitrile (HPAN) ultrafiltration membrane is facilely co-deposited with polydopamine (PDA) and polyethyleneimine (PEI), and the PDA-PEI coating further induces the growth of zirconia (ZrO <sub>2</sub> ) nanoparticles on the membrane surface.	The formed zirconia selective layer is defect-free, uniform, and ultrathin (10–20 nm). The organic-inorganic TFC NFMs show high retention (>90%) for divalent cations with a salts-rejection sequence of MgCl <sub>2</sub> > CaCl <sub>2</sub> > MgSO <sub>4</sub> > NaCl > Na <sub>2</sub> SO <sub>4</sub> at pH 6.0 ascribed to the positively charged membrane surface. The water-flux reaches as high as 60 L/m <sup>2</sup> h under 0.6 MPa due to the ultrathin and hydrophilic properties of the inorganic selective layer.	2016 Lv <i>et al.</i> (2016)

Thin-film composite (TFC) nanofiltration membranes (NFMs) are the most currently synthesized membranes, which consist of an inorganic nanoparticles (NPs) layer coated the polymeric MF or UF substrate (Song *et al.* 2016; Cheng *et al.* 2017; Xu *et al.* 2017). For example, Lv *et al.* used as a polymeric substrate in their research polyacrylonitrile (HPAN) UF, which was hydrolyzed and then transferred into polydopamine/polyethyleneimine solutions before the growth of zirconium nanoparticles in the membrane surface. The top selective layer was uniform, ultrathin (<20 nm), and defect-free. The novel TFC NFMs showed high rejection, as high as 90%, for multivalent cations with a salts-rejection order of  $\text{MgCl}_2 < \text{CaCl}_2 < \text{MgSO}_4 < \text{NaCl} < \text{Na}_2\text{SO}_4$  at pH about 6. The water-flux reached  $60 \text{ L/m}^2 \text{ h}$  under 0.6 MPa due to the ultrathin and hydrophilic properties of the inorganic selective layer. Moreover, the novel membrane exhibited excellent structural stability for long term NF operation (Lau *et al.* 2012; Lv *et al.* 2016). Grafting polymerization (GP) and interfacial polymerization (IP) were also recognized as useful and practical membrane technologies (Dvir *et al.* 2006; Zhu *et al.* 2012; Lin *et al.* 2016; Wu *et al.* 2016; Matsumoto *et al.* 2018).

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## INFLUENCE OF OPERATING PARAMETERS

The operating parameters identified in the various studies included (a) transmembrane pressure (TMP), (b) pH, (c) feed concentration, (d) temperature, and (e) organic matter and background salt concentration, which had little effect on arsenic removal. Below are the results of the effects of the operating parameters on the release of arsenic.

Dompé and Ahoulé, in their thesis research on *Comparative performance of nanofiltration and reverse osmosis techniques for the treatment of drinking water contaminated with arsenic in Burkina Faso*, concluded that operating pressure and pH are important parameters that need to be worked upon to improve the efficiency of the membrane system for arsenic removal (Dompé & Ahoulé 2016). Sen and Pal studied the effects of TMP, pH, iron concentration, and pre-oxidation on the performance of three membranes in terms of arsenic rejection and flux (Sen & Pal 2009). The TMP showed a strong impact on flux and As removal, whereas pH and pre-oxidation were found to exhibit a high influence on As removal percentage. For all three membranes, As removal increased from 50–63% to 97–100% for TMP in the range of 5–12 kgf/cm<sup>2</sup>. Arsenic removal increased around 23% for NF-1, 33% for NF-2, and 26% for NF-20 while the pH increased from 3 to 10 (Sen & Pal 2009). It was also observed by a group of researchers that an increase of pH and a decrease of operating temperature and arsenic feed concentration determined a higher efficiency of arsenic rejection, while the TMP slightly affected the arsenic removal of the NF-30 membrane. In all cases, the permeate flux increased with temperature and pressure, and the maximum value was reached at pH 8 (Figoli *et al.* 2010). As a common trend, while As concentration in feed water increases, both diffusion and convection of the uncharged arsenite species increase, which leads to a decrease in the rejection of As (III) (Seidel *et al.* 2001; Xia *et al.* 2007). Otherwise, the concentration of humic acid and the formation of As (III) complexes vary in the same order. So, for an efficient arsenite removal, the size of the As (III) complexes may be larger than the nanofiltration membrane orifice (Livens 1991; Xia *et al.* 2007).

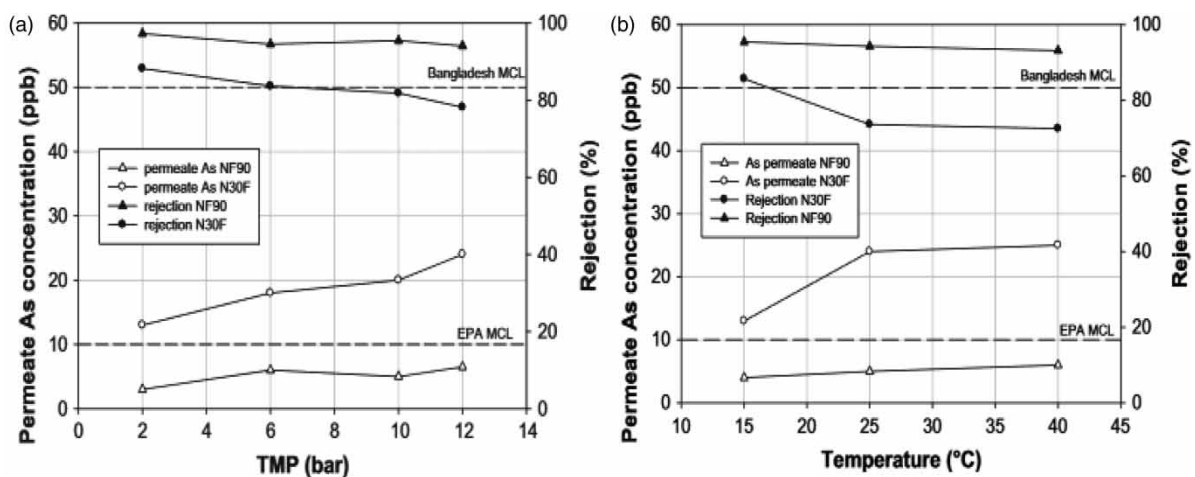
Additional salts present in the solution were shown to affect the arsenate removal. Research works have shown that by increasing the concentration of anions or cations the passage of arsenate is increased. The reduction in As (V) removal in the presence of divalent  $\text{Ca}^{2+}$  was greater than for monovalent  $\text{Na}^+$  by about 1%. On the other hand, nothing has been observed as an influence on the rejection of As (III) by the various additions of salts (Xia *et al.* 2007), while the pH increased As removal by NF membrane also increased (Xia *et al.* 2007). In the well-defined operating conditions of their research, Saitúa *et al.* found that the As (V) removal was independent of TMP, temperature, and crossflow velocity (Saitúa *et al.* 2005). The possibility of using NF membranes for the treatment of drinking water by varying the pH, pressure, and the initial concentration of arsenic was tested on the

separation performance of the TFC-NF membranes. The effectiveness of this technology depends only on two parameters, the pH and the presence of other ions (Van der Bruggen *et al.* 2008; Harisha *et al.* 2010).

Additionally, the capacity for genetic programming (GP) as a prediction tool to predict the performance of the NF system was studied. The input data in this model were TMP (transmembrane pressure) and feed concentration. (a) Permeate flux increased by TMP increasing; (b) solvent flux increased more than solute flux; (c) therefore the As removal increased (Garba *et al.* 2003; Okhovat & Mousavi 2012). According to Cuong *et al.*, the initial concentration of arsenic had almost no influence on the retention of arsenite and arsenate. However, the increase in the initial arsenate concentration marked a much more noticeable increase in arsenate release (Nguyen *et al.* 2009). On the contrary, the transmembrane pressure (TMP) acts only weakly on the rejection of arsenic according to Sato *et al.* (Sato *et al.* 2002). The results reported by Ahmed *et al.* indicated that As (V) rejection by the UTC-70 membrane was influenced slightly by the variation in operating conditions, whereas As (V) rejection by the NTR-7450 membrane was strongly dependent on retentate arsenate, feed water composition, TMP, vibration amplitude and pH (Ahmed *et al.* 2010).

### Effect of transmembrane pressure (TMP) on arsenic removal

The arsenic removal is shown in Figure 6(a) as a function of transmembrane pressure (TMP) for both NF 90 and N-30F membranes under operating conditions, a feed As concentration of 100 ppb, a pH of 8, and a temperature of 25 °C. The rejection of arsenite (As) by NF 90 membrane is more important than the rejection observed with the N-30F membrane over the TMP range investigated. The NF 90 membrane exhibited an excellent rejection of about 94% towards arsenate rejection while the rejection of the N-30F membrane is up to 78%. This difference is due to a lower molecular weight cut-off (MWCO) of NF 90 (200 Da) in comparison with N-30F (400 Da). In the range of TMP investigated by Figoli *et al.* (2010), the increase in operating pressure didn't improve the arsenic rejection a lot.



**Figure 6** | (a) Effect of TMP on the removal of arsenic (feed concentration = 100 ppb, pH = 8, temperature = 25 °C), (b) effect of temperature on the removal of arsenic (feed concentration = 100 ppb, pH = 8, TMP = 6 bar) (Figoli *et al.* 2010; Chen *et al.* 2018).

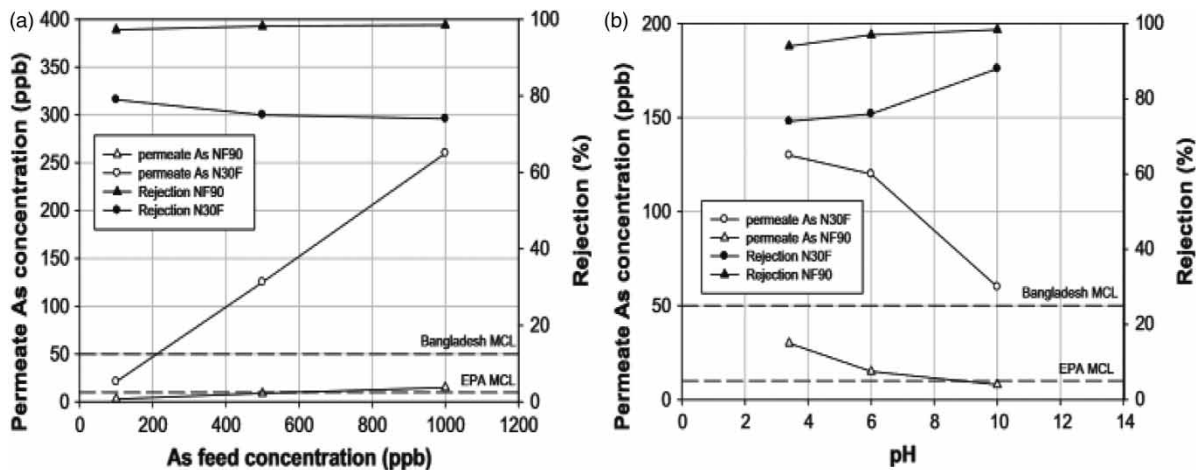
Saitúa *et al.* (2005) found similar results while investigating the nanofiltration process of synthetic solutions with spiral wound polyamide membranes (192NF-300, Osmonics, Inc.) characterized by a MWCO of 180 Da. Similarly, Sato *et al.* (2002) obtained that the arsenate As (V) removal was almost unrelated to the applied pressure using different membranes, ES 10, NTR 729HF, and NTR 7250 (from Nitto Electric Industrial Co., Japan).

### Effect of operating temperature on arsenic removal

Figure 6(b) depicts the arsenic removal for both NF 90 and N-30F membranes as a function of temperature. The NF 90 membrane exhibited a higher arsenic removal in comparison with the N-30F membrane under the range of temperatures studied. Indeed, for NF 90 membrane, the arsenic removal was 95.5% and 93.2% at 15 °C and 40 °C, respectively. For these membranes, there was a slight decrease in arsenic rejection while temperature increased. Yang *et al.* (2018) demonstrated similar behavior in drinking water treatment by charged UF membranes.

### Effect of arsenic concentration on arsenic removal

Figure 7(a) shows the influence on the arsenic rejection of the feed concentration for both NF 90 and N-30F membranes. The NF 90 membrane exhibited better arsenic rejection than the N-30F membrane under different arsenic feed concentrations. The arsenic rejection by the NF 90 membrane was as high as 97% whatever the arsenic (As) feed concentration. The rejection of arsenic by the N-30F membrane was between 74 and 79%.



**Figure 7** | (a) Effect of As feed concentration on the removal of arsenic (TMP = 6 bars, pH = 8, temperature = 25 °C), (b) effect of pH on arsenic removal (feed concentration = 500 ppb, temperature = 25 °C, TMP = 6 bar) (Figoli *et al.* 2010; Chen *et al.* 2018).

### Effect of pH on arsenic removal

Figure 7(b) depicts the effect of pH on arsenic rejection by NF 90 and N-30F membranes. N-30F membrane showed that arsenate As (V) rejection increased significantly while the pH increased. Arsenic removal increased from 74% to 88% while the pH increased; indeed, the monovalent ion  $H_2AsO_4^-$  is dominant in the solution for pH between 4 and 6, while the divalent ion  $HAsO_4^{2-}$  is dominant at pH above 7. Multivalent ions were better rejected by the N-30F membrane in comparison with monovalent ions due to large hydrated radii of multivalent ions (Vrijenhoek & Waypa 2000; Brandhuber & Amy 2001; Xu *et al.* 2018; Gholami *et al.* 2020).

Polyethersulfone (PES) membranes (such as N-30F) are negatively charged at high pH and their effective charge-density decreases with lower pH. The isoelectric point, which is defined as the pH required to get the net-charge of the membrane equal to zero, is located around 3 (Weis *et al.* 2003; Navarathna *et al.* 2019).

NF 90 membrane exhibited a rejection of arsenate As (V) between 94.3% and 98.3% for pH range 3.4–10. NF 90 membrane had a negative charge in the neutral pH range and like the N-30F



membrane, its charge got more negative as the pH value increased. [Urase \*et al.\* \(1998\)](#) demonstrated a similar trend with the ES-10 NF membrane (Nitto-Denko Co., Ltd) made of aromatic polyamide. Recently, [Chen \*et al.\* \(2018\)](#) confirmed the same trend with his novel synthesized TFC-NF membrane.

## PERFORMANCE COMPARISON OF NANOFILTRATION AND REVERSE OSMOSIS TECHNIQUES FOR ARSENIC REMOVAL

Drinking water production in compliance with the regulation of 10 µg/L for streams of water rich in arsenic depends on the efficiency of the process applied; an efficient process also requires optimum operating conditions. In the membrane filtration process, reverse osmosis (RO) is increasingly abandoned in favor of nanofiltration (NF), not for RO inefficiency but its cost. The objective of this section is therefore to investigate the effectiveness of NF and RO membranes with respect to the retention of arsenic under different operating conditions. The performance evaluation parameters include the quality of the permeate water relative to membrane rejection (concentration of As < 10 µg/L) and the productivity of the system (water flux performance).

### Elimination of As (III) and As (V) as a function of pressure and conversion rate

The performance of NF270 and RO (TW30) membranes in relation to the elimination of As (III) and As (V) as a function of operating pressure and conversion rate are summarized in [Table 7](#) below.

The retention rates of As (III) and As (V) obtained with the NF270 membrane ranged from 1.5 to 9%, and from 70.4 to 85.7%, respectively. These results are similar to those obtained by [Košutić \*et al.\* \(2006; Al-Rashdi \*et al.\* 2013\)](#). They obtained retention rates of As (III) by NF270 between 0 and 11% for a pH 5 solution with pressure varying from 3 to 5 bars. [Košutić \*et al.\* \(2006\)](#) found an As (V) retention rate of 81.8% for a concentration supply solution 229 µg/L at pH 6.89 with a pressure of 6.8 bar. With the TW30 membrane, the retention rates of As (III) and As (V) vary from 40.2 to 75.4%, and from 93.6 to 99.8%, respectively. [Chang \*et al.\* \(2014\)](#) indicated a retention rate of 89% for As (III) at 10 bar for a pH of 7.2 ([Chang \*et al.\* 2014](#)).

**Table 7** | Retention rate of As (III) and As (V) by NF and RO membranes ([Dompé & Ahoulé 2016](#))

		Retention rate (%)					
		P (bar)	Y = 10%	Y = 30%	Y = 50%	Y = 70%	Y = 90%
As (III)	NF	1	6,7	6,5	2,9	ND	ND
		2	ND	9	7,8	4,1	1,5
		3	ND	3,1	3,4	2,2	2,9
	RO	2	60,8	48,3	40,2	ND	ND
		4	ND	66,8	62,4	55,4	44,2
		6	ND	75,4	71,1	65,4	55,5
As (V)	NF	1	79,1	78,4	70,4	ND	ND
		2	ND	85,7	84,7	80,9	72,7
		3	ND	83,7	80,3	78	73,4
	RO	2	95,8	95,5	95,1	ND	ND
		4	ND	97,9	96,4	95,5	93,6
		6	ND	99,8	98,2	96,5	94,9

P: Pressure; Y: Conversion rate; ND: Not Defined; [As (III)] = 100 µg/L; [As (V)] = 200 µg/L.

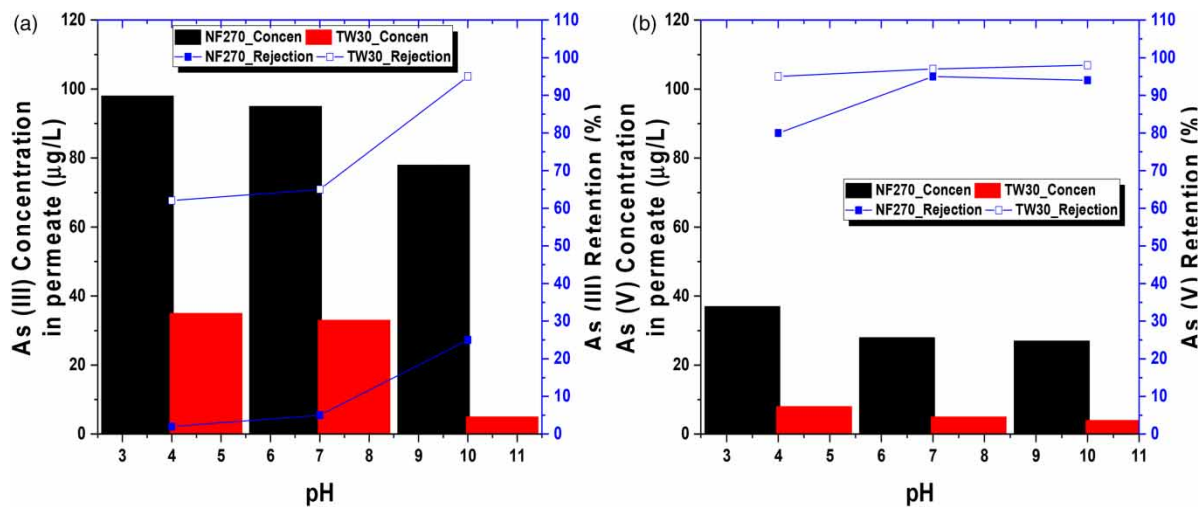
[Elcik \*et al.\* \(2013\)](#) investigated arsenic removal by two types of thin-film polyamide nanofiltration (TFP-NF) membranes, [NF270, NF90], and two reverse osmosis membranes (XLE, BW30) ([Elcik \*et al.\* 2013](#)). As (III) removal of these RO membranes was between 97 and 99% for all transmembrane pressures applied. As (III) retention with NF90 was higher than 62% while As (III) removal with

NF270 is higher than 40%. Similarly, Teychene *et al.* achieved >99% removal of As (III) by BW30 at pH 7.6 (Teychene *et al.* 2013).

### Elimination of As(III) and As(V) as a function of pH

The influence of pH on the retention of As (III) and As (V) by NF270 and RO (TW30) membranes was obtained through tests at a conversion rate of 70%, and at a pressure of 2 bars for the NF membrane and 6 bars for the membrane RO, corresponding to the optimum conditions of the system.

Figure 8(a) shows the retention of As (III) by the NF and RO membranes at different pH. It shows that the concentration of As (III) in the permeate remains substantially equal when the pH varies from 4 to 7 for both two membrane types and decreases quickly when the pH changes to 10. This results in a retention rate almost identical to pH 4 and 7 and an increase in the rate at pH 10. This slight variation in As (III) retention was highlighted by Seidel *et al.* (2001) who worked on nanofiltration membrane BQ01 at a pH range of 4.5 to 8.5. They observed that the retention of As (III) was almost unaffected by a change in pH from 4.5 to 8.5. Chang *et al.* (2014) also demonstrated a very slight As (III) rejection when pH ranged from 4 to 9 for both NF and RO membranes. Also, the low retention of  $H_3AsO_3$  compared to  $H_2AsO_3^-$  can be explained, according to Akin *et al.* (2011), by the fact that at lower pH 9.24, arsenous acid ( $H_3AsO_3$ ) can form hydrogen bonds with the active groups of membrane and can spread more easily through them such as boric acid and water. Arsenite is dissociable with the first pKa at 9.2; above this pH, it is found that As (III) removal increases with the increase in pH (Uruse *et al.* 1998; Elcik *et al.* 2013; Chang *et al.* 2014).



**Figure 8** | (a) Concentration of As (III) in permeate and rejection rates for different pH, (b) concentration of As (V) in permeate and rejection rates for different pH (Dompé & Ahoulé 2016).

As (V) removal by NF and RO membranes at different pH is highlighted in Figure 8(b). (Dompé & Ahoulé 2016). The retention rate of As (V) ranged from 81.8 to 86.2% for NF270 and 96.3 to 97.4% for RO. However, concentrations of As (V) at pH 7 and 10 appear to be quite equal. Elsewhere, it was observed a gradual increase in the rejection of As (V) with the increase in pH for the nanofiltration membranes NF30 and NF90 (Figoli *et al.* 2010).

### Elimination of As (III) and As (V) as a function of the concentration of As

Several scientists have investigated the influence of arsenic concentration in the feed solution on the performance of NF and RO processes for As (III) and As (V) release. The concentration of As (III) in the

permeate ranged from 47.29 to 144.06  $\mu\text{g/L}$  (a retention rate of 5.4 to 4%) for the NF270 membrane and 18.33 to 64.47  $\mu\text{g/L}$  (a retention rate of 63.3 to 57%) for the RO (TW30) membrane when the concentration of As (III) in the feed solution ranges from 50 to 150  $\mu\text{g/L}$  (Dompé & Ahoulé 2016). An increase in the concentration of As (III) in the permeate is observed with an increase in the concentration of As (III) in the feed solution. The same trend was observed by (Xia *et al.* 2007; Geucke *et al.* 2009; Nguyen *et al.* 2009; Al-Rashdi *et al.* 2013). For example, Al-Rashdi *et al.* (2013) observed a decrease in the retention of As (III) by the NF membrane when the initial concentration of As (III) ranged from 100 to 500  $\text{mg/L}$  (Al-Rashdi *et al.* 2013). Geucke *et al.* (2009) observed an increase in the concentration of As (III) in the permeate with an increase in the initial concentration of As (III) with the RO membrane (Geucke *et al.* 2009). According to Seidel *et al.* (2001), when the concentration of the feeding solution increases, the diffusion and convection of the unladen As (III) species also increase together (Seidel *et al.* 2001). This results in an additional decrease in its retention, therefore an increase in its concentration in the permeate. Besides, the concentration of As (III) in the permeate remains greater than 10  $\mu\text{g/L}$  for the two membranes (NF270 and TW30) studied at pH 7. Therefore, they cannot be used for the production of drinking water from a water source containing more than 50  $\mu\text{g/L}$  of As (III) without prior oxidation of As (III).

The concentration of As (V) in the permeate ranged from 17.41 to 125.94  $\mu\text{g/L}$  (a retention rate of 82.6 to 87.4%) for NF270 and 1.86 to 9.2  $\mu\text{g/L}$  (a retention rate of 98.1 to 99.1%) for TW30 when the concentration of As (V) in the feed solution ranges from 100 to 1,000  $\mu\text{g/L}$ . The concentration of As (V) in the permeate, therefore, increases with its concentration in the feed solution for both membranes (Geucke *et al.* 2009; Figoli *et al.* 2010; Dompé & Ahoulé 2016). Geucke *et al.* and Figoli *et al.* also made the same observation. However, As (V) concentration in the permeate with RO remains below the 10  $\mu\text{g/L}$  standards for all the initial concentrations studied, in contrast to the NF membrane. The RO membrane can therefore be used for the production of drinking water from a water source containing 1,000  $\mu\text{g/L}$  of arsenic provided that all arsenic is in the form As (V) or that the As (III) is previously oxidized to As (V). Seidel *et al.* (2001) also observed that the retention rate of As (III) decreased from 28 to 5% while that of As (V) increased from 50 to 85% when the initial concentration of arsenic increased from 10 to 315  $\mu\text{g/L}$  at pH 8 (Seidel *et al.* 2001). According to these authors, this is due to the primary role of electrostatic exclusion in the elimination of  $\text{HAsO}_4^{2-}$ .

### Effect of the ionic force on the retention of As (III) and As (V)

An increase in  $\text{Cl}^-$  and  $\text{Na}^+$  concentration in the feed solution leads to a decrease in arsenic retention (Geucke *et al.* 2009; Ahoranta *et al.* 2016; Song *et al.* 2016). However, this influence depends on the type of membrane used. It remains weak with the TW30 membrane (reverse osmosis membrane) and is more important with the nanofiltration membrane (NF270). Arsenite As (III) was not influenced by the salt composition of the feed solution for pH values below 9.24 for all types of membranes used in their work by (Xia *et al.* 2007; Dompé & Ahoulé 2016). Dompé and Ahoulé and Xia *et al.* also found that the addition of NaCl or  $\text{CaCl}_2$  had no apparent influence on the removal of As (III). This could be explained by the fact that As (III) at pH 7 is a neutral solute; its retention is governed by steric exclusion (Nguyen *et al.* 2009), so the presence or absence of other solutes in the feed solution cannot, therefore, influence its rejection.

### Effect of pre-oxidation step on arsenic removal by nanofiltration membrane

#### Pre-oxidation step by hypochlorite

The step of pre-oxidation is used to transform As (III) into As (V) since As(V) is easy to remove from water.

In a work performed by Elcik *et al.* (2013) and another recent work by Navarathna *et al.* (2019), using two types of membranes, oxidation of As (III) to As (V) had a negligible effect on the performance of RO membranes but had a significant effect on the performance of NF membranes. With the addition of hypochlorite of just 1 mg/L, the arsenic rejection reached 95.8% for NF90 and almost 88% for NF270.

#### Pre-oxidation step by potassium permanganate ( $\text{KMnO}_4$ )

Figure 9 depicts the effect of the pre-oxidation step before the application of nanofiltration membrane on arsenic removal. Sen *et al.* studied three different types of nanofiltration membranes, NF-1, NF-2, and NF-20 (Sen *et al.* 2010). These membranes were used to treat water that was previously dosed with  $\text{KMnO}_4$ . Pre-oxidation of arsenite As (III) by potassium permanganate ( $\text{KMnO}_4$ ) involved substantial improvement in the rejection efficiency of NF membranes. Arsenic rejection increased from 50–63% to 97–100% for all the three NF membranes tested over a TMP range of 5–12 kgf/cm<sup>2</sup> on pre-oxidation of trivalent arsenic. While pH increased from 3 to 10, arsenic removal increased by 26% for NF20, 33% for NF2, and 23% for NF-1 membranes.

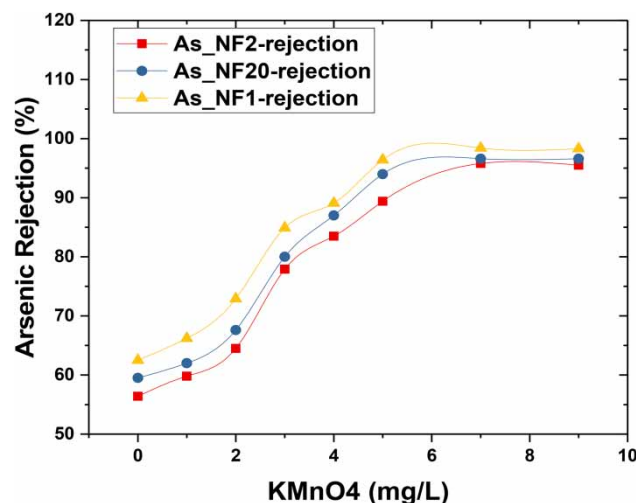
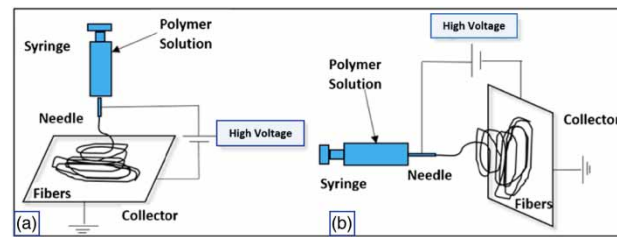


Figure 9 | The effect of the pre-oxidation step prior to the nanofiltration membrane on arsenic removal (Sen *et al.* 2010).

#### PERSPECTIVES AND LIMITATIONS

Electrospinning, an emerging synthesis technique of NF membranes, is reported to be a flexible method for nanofibrous porous membrane fabrication for drinking water production and wastewater treatment (Prince *et al.* 2012; Lalia *et al.* 2013). Electrospinning exhibits various advantages such as 3D interconnected pore structure, low start-up cost, high strength, high surface area to volume ratio, more contacting surface (fiber surface), and it favors a well-dispersed mixed matrix membrane. All these advantages make it a viable technology for membrane preparation (Ray *et al.* 2016). It has also been shown that the membrane porosity depends on the operational parameters like operating time, voltage, the distance between collector and spinneret, solution property, and feed rate. The desired membrane morphology can be reached just by tailoring the solution, property, and operational parameters. There is the possibility to obtain any NF membrane using electrospinning technology. A schematic diagram of electrospinning process is shown in Figure 10.

Another novel technology for arsenic removal investigated recently is graphene-based materials. Graphene oxide (GO) displays functional groups (carboxyl, C = C, hydroxyl, and epoxide) on the surface, which enables it to be used as a filler material in different types of membranes for their



**Figure 10** | Schematic diagram of the electrospinning process (vertical and horizontal) (Xue *et al.* 2019).

modification and improvement of the properties. B. Mi suggested the incorporation of GO in order to generate interconnected nanochannels in membranes, which led to water flux enhancement (Mi 2014). GO exhibited great antibacterial properties, antifouling, and anti-biofouling, it is also reported to be hydrophilic in nature because of the hydrophilic group that it possesses (Lee *et al.* 2013; Chae *et al.* 2015). Furthermore, the graphene-based materials used as nanocomposites consist of inorganic/organic compounds with GO, reduced graphene oxide (rGO), metal oxides. Copper oxides, iron oxide, etc. were combined with GO or rGO in the nanocomposites (Dubey *et al.* 2015; Khatamian *et al.* 2017; La *et al.* 2017; Su *et al.* 2017; Yoon *et al.* 2017). Inorganic compounds including silica, LD-hydroxide (Wen *et al.* 2013; Wang *et al.* 2015) and organic compounds epoxy, EDTA-chitosan, aminothiobiuret, amino pyrazole (Awad *et al.* 2017; Shahzad *et al.* 2017; Alimohammady *et al.* 2019; Verduzco *et al.* 2019) were used with GO or rGO to synthesis nanocomposites. The arsenate rejection rate of these nanocomposites reaches 100% in some cases.

Titanium, silicon, or zirconium are an emerging class of nanoparticles, which due to their unambiguous chemical signal can facilely be deposited on a membrane surface using the information provided by scanning electron microscopy in association with energy dispersive spectrometry. Thus, new NF membranes could be fabricated for a specific purpose. However, because of the high surface energies of these nanoparticles (NPs), they tend to aggregate in aqueous media, thus leading to a drastic decrease in the surface area and therefore reduced capacity, selectivity, and long-term utilization of the resulting membrane. Despite the increasing number of studies realized regarding this class of NPs, little attention was given to them in sciences and engineering, while in other fields these nanoparticles are gaining more attention due to their outstanding properties.

Biomimetic membrane process-drawing, which was inspired by natural methods of water transport and purification, has recently been a motivation for the development of novel efficient separation membranes to purify streams water, not only quickly but also efficiently. Over thousands of years, nature has developed essential and excellent proteins in water channels, such as aquaporins, which are very important for the life of all organisms. These protein layers facilitate the rapid and highly selective transport of water across the cell membrane (Balaban *et al.* 2006; Yang *et al.* 2018; Yalcinkaya *et al.* 2020).

2D-materials, for example, graphene, have recently attracted scientists worldwide because they were a potential novel class of NFMs since they showed a minimum resistance in water transportation. Computational investigations have justified excellent predicted salt rejection rates and high permeate flux through a nanoporous graphene membrane (Cohen-Tanugi & Grossman 2015).

For smaller treatment plants, NF is a more cost-effective method, as reported in the literature (Ferndale 2013). If highly colored water is being treated, NF membrane systems produce water at a cheaper price compared to technologies using ozonation, lime soda, and granular-activated carbon (GAC).

Although NF membranes have proved their effectiveness in many fields of applications including arsenic removal, a remaining large-scale application is still in the premature stage due to water flux decrease with time, low long-term durability of membranes, rebarbative problems of fouling, and lack of research on membrane materials. Most of the studies carried out focus on commercial NF

membranes available in the market rather than novel synthesized ones for arsenic removal. Almost all of the research conducted is limited to treating arsenate ions, which are easier to process and are not subject to the steric exclusion phenomenon. NF polyamides, the most used membranes for drinking water production and wastewater treatment, are negatively charged under operating conditions, as a result of the balance between amine end groups and dissociated free acid. This negative surface charge greatly favors the membrane fouling activity. Naturally, the membrane fouling reduces the water flux with time, both lower mechanical strength and the swelling tendency of the membrane reduce the NF membranes' lifetime for As removal.

Furthermore, membrane fouling is still a great hindrance to membrane competitiveness compared to other conventional technologies. Fouling limits the competitiveness of membrane operations and cost-effectiveness. Fouling is the source of several deleterious effects such as decline in permeate water flux (decreased productivity), maintenance, membrane degradation, and replacement, increase in energy demand, shorter membrane lifespan, chemicals and frequency for cleaning, and above all the increase in costs.

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## CONCLUSION

Nanofiltration membranes (NFMs) have shown high rejection performance towards arsenate As (V) ions. Although As (III) is more toxic than As (V), most of the works performed so far focused on the arsenate removal since its rejection process is easier due to the Donnan exclusion effect of membranes. Both commercial and synthesized NF membranes were unable to reject more than 70% As (III) from water without a prior pretreatment. Nevertheless, scientists have prepared novel NF membranes, modifying microfiltration (MF) or ultrafiltration (UF) membranes by different methods including sol-gel deposition, in-situ formation, using nanoparticles (NPs). These synthesized membranes, according to the field of use, have shown long-term efficiency, fouling reduction, cost reduction, an increase in separation of multivalent ions, rejection performance, and high flux achievement. Taking into account the features mentioned above, NFMs has the potential to be the most efficient arsenic (As) rejection technology and it could be possible to prefer it for drinking water production in the affected areas, at a household, small plant, and community level. However, problems still persist in arsenite As (III) removal, fouling control, and mitigation. Future investigations should focus on developing better and cost-effective NF membranes under easier operating conditions with improving arsenite rejection, and ability to control, reduce, and mitigate fouling.

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## CONFLICT OF INTEREST

On behalf of all authors, the corresponding author states that there is no conflict of interest.

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## DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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