



## Arsenic removal from groundwater by membrane technology: Advantages, disadvantages, and effect on human health

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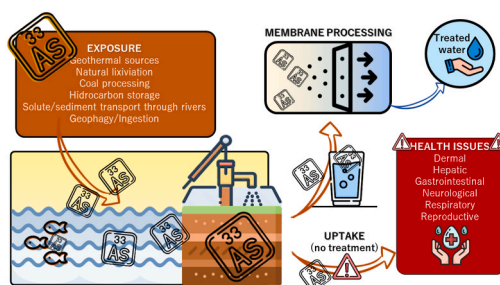
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### HIGHLIGHTS

- Sources and toxicity of arsenic species.
- Arsenic effect on human health have been analyzed.
- Advantages of membranes in arsenic removal have been reviewed.
- Fouling phenomena was discussed and mitigation measure drawn.
- ENFMs for arsenic removal in groundwater was summerized.

### GRAPHICAL ABSTRACT



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### ABSTRACT

The presence of different heavy metals such as arsenic in groundwater is evident and can be attributed to environmental processes and anthropogenic activities. Arsenic is considered one of the most toxic chemical elements in nature; therefore, many studies proposed valid processes for groundwater remediation. In this review, the primary arsenic sources are explored. It also has provided an interesting discussion of how arsenic impurities can be removed from the groundwater using various processes while highlighting the advantages and disadvantages of each of them. Particular attention has been focused on the membrane process. Nanofiltration applications at large scales are obstructed by the difficulty of As(III) removal (which is the most toxic As form) and fouling issues. Application of nanofiber membranes in arsenic remediation is also described: these membranes, characterized by high surface area, uniform pore-size distribution, and improved pore connectivity, exhibit excellent adsorption capacity. Although the research activities in this field have made progress, several problems need to be solved, such as improvement of the porosity and the size of the pores, and the mechanical strength for promoting their use in industrial operating conditions.

### 1. Introduction

Groundwater is widely employed as a water resource for domestic,

agricultural, and industrial purposes, especially in arid regions, due to the scarcity of surface water and rainfalls (Li, 2020). The continuous growth of the worldwide population, industrialization, agriculture

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production and climate change determined a detrimental effect on groundwater quality (Abdelkader et al., ). Some chemical species such as mercury (Hg), zinc (Zn), chromium (Cr), lead (Pb), cadmium (Cd), Fluoride (F), and metalloid as selenium (Se) and arsenic (As) are present in amounts higher than the WHO limit in groundwaters. Most of them has severe effects on human health (Ab Razak et al., 2015; Maity, Vithanage, 2021) These metals and metalloids are natural elements on the earth, and their concentration can change according to the local geology. Table 1 are reported the highest level of heavy metals found in drinking water according to EPA (Kurniawan et al., 2006).

A higher level of arsenic in groundwaters hurts the quality of drinking water, being extremely poisonous to humans and animals (Oliveira et al., 2021), (Chakraborti et al., 2013). Arsenic occurs in distinct inorganic and organic forms and different oxidation states (-3, 0, +3, and +5) (Smedley et al., 2002; Ahmad et al., 2018); in particular, some forms, trivalent (AsIII) and pentavalent (AsV) are hazardous for human and environmental health (Bhattacharya et al., 2007).

The main chemical groups containing As retraceable in the environment are arsenic acids ( $H_3AsO_4$ ,  $H_3AsO_4^-$ ,  $H_3AsO_4^{2-}$ ), arsenious acids ( $H_3AsO_3$ ,  $H_3AsO_3^-$ ,  $H_3AsO_3^{2-}$ ), arsenites, dimethylarsinic acid, arsine, arsenates and methyl arsenic acid. As reported in Table 1, the latter, a metalloid, is extremely dangerous for humans and animals. Elevated concentrations of arsenic (>10 µg/L) are observed in different countries, including the USA, Argentina, Germany, Vietnam, Chile, Mexico, Bangladesh, Pakistan, China, and India (Aftabtalab et al., 2022). Traditional methods to remove arsenic from groundwater include flocculation, precipitation-filtration, coagulation, ion exchange, oxidation-reduction, and carbon adsorption (M and S, 2019). However, these methods present different disadvantages: high energy demand, reduced efficiency (particularly for slightly polluted groundwater), andic compounds (M and M, 2015a), (Mani and Kumar, 2013). Membrane technology is appropriate for cleaning groundwater due to the reduced energy consumption, high efficiency in retaining pollutants of different sizes, reduced waste production, and the easiness of integration with different traditional processes (Kehrein et al., 2020). Membrane processes typically used for water and wastewater treatment are microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO). In most of the membrane operation the driving force is the pressure difference across the membrane (Saleh et al., 2022). For arsenic removal, NF and RO membrane processes are appropriate due to the minimal pore size of the membranes (Ezugbe and Rathilal,

**Table 1**  
Maximum contaminant level (MCL) for various heavy metals and their effect on human health. They were adapted with permission from reference [(Kurniawan et al., 2006)].

Contaminant	MCL <sup>1</sup> (mg/L)	Long term of exposition > MCL effects	Contaminant source
As	0.010	Skin damage; circulatory problems; cancer development.	Rock erosion
Cd	0.005	Kidney disease.	Natural deposit erosion; Corrosion of galvanized pipes; metal refineries discharge
Cr	0.10	Allergy.	Rock erosion. Steel and pulp mill discharge.
Pb	0.015	Physical or mental Delays for infants and children, nephropathy (adults); high blood pressure (adults).	Natural deposit erosion; Corrosion of domestic plumbing.
Hg	0.002	Nephropathy	Natural deposit erosion; Refinery and factory discharge.
Se	0.05	Loss of hair or fingernails; circulatory problems.	Natural deposit erosion; Refinery discharge.

<sup>1</sup> MCL = Maximum permissible level of a contaminant in drinking water.

2020). Those methods see polymeric membranes as the primary membrane type used, and this is true for the whole water sector on an industrial level, to their ease of manufacturing and remarkable efficiency (Al Aani, Mustafa and Hilal, 2020); (Gharsallah et al., 2022). Suffer disadvantages such as fouling, chemical stability, and the trade-off between permeability and selectivity (Hofs et al., 2011a) (Ahmad et al., 2022; M.G De Paola and Lopresto, 2021). The accumulation of organic and inorganic materials (present in the wastewater) on the surface and in the membrane, pores causes fouling, which determines a reduction of the permeate flux and quality and a consequent increase in process costs (Guo et al., 2012). Different routes are followed for fouling mitigation as wastewater pretreatments and cleaning (either physical or chemical). The hydrophobic character of the membranes also influences the fouling, so the development of membranes with high hydrophilicity permits a fouling lessening 1 (Editorial Board, 2019).

This review discusses the source of arsenic and its adverse impact on human and environmental health. In addition, the pros and cons of the different conventional technologies used to remove As are briefly discussed. Subsequently, nanofiltration (NF) membrane technology for heavy metal removal, the advantages, and drawbacks of nanotechnology are presented, with a look to future perspectives.

## 2. Arsenic sources

Arsenic is an essential element in hundreds of minerals, such as sulfides, arsenates, arsenides, and arsenites; furthermore, it is an ever-present component in the atmosphere, soils, minerals, natural waters, and organisms (Matschullat et al., 2000). The primary means of diffusion of arsenic are usually: water streams, which are discharged into the environment by biological and artificial sources, and volcanic activity, aided by windblown dust clouds (Merian, 2008). Artificial emissions in the air occur from the smelting of metals, the discharge of fuels - mainly brown coal - and pesticides (Merian, 2008) (Chakraborty et al., 2012). However, arsenic-based pesticides were gradually replaced by other preparations, and consequently, the production of arsenic decreased, even if arsenic, in different amounts, is still utilized in agricultural chemicals. Today, arsenic pollution has different sources, and its diffusion has been documented worldwide; Particularly in South (Argentina, Chile), North American, and South Asian countries (Vietnam, Myanmar, India, Nepal, Cambodia, Bangladesh, and Indonesia) are regarded as the most Arsenic polluted regions (Ahoulé et al., 2015; Ahmad and Bhattacharya, 2019b; Shaji et al., 2021a). More than 100 countries are affected by arsenic pollution in groundwater (above the Maximum Contaminant Level MCL limit of 0,010 mg/L (Herath et al., 2016)), and most of the arsenic contaminated zones are located near mines, mountain belts, and river delta areas (Shaji et al., 2021b, 2021c; Bundschuh et al., 2022). The summary of Arsenic Distribution around the world is reported in Fig. 1.

Arsenic is essentially embedded as sulfide salt within ores complexes and, in particular, can be found in mineralized areas integrated with metals, particularly silver, lead, and gold. With 46% by mass, Arsenopyrite, FeAsS, and orpiment are considered the most significant arsenic source among all ferrous sulfates; moreover, under oxidizing conditions, it decomposes to discharge acids of As and S into the environment, contributing to polluting the acid mine sewerage with high concentrations of dissolved As (Corkhill and Vaughan, 2009). Once oxidized and then dissolved, Arsenopyrite represents a crucial element of pollution in the environment and the oxidoreduction process affects the geochemical behavior of As (Mandaliev et al., 2014; Hong et al., 2020) The chemical structure and molecular structure of Arsenopyrite are shown in Fig. 2.

As an integral part of the ore structure and assimilated components, significant amounts of Arsenic are present in several hydrated metal oxides and oxide minerals. Usually, arsenic concentrations in iron oxides could reach weight % values, whereas one weight % is equivalent to 10,000 mg/kg; essentially, they result from primary iron sulfide minerals oxidation, which contains an abundant presence of Arsenic (Punshon et al., 2017). Arsenate adsorption to iron (III) oxide-hydroxides



Fig. 1. Arsenic Distribution around the world (adapted from Smedley and Kinniburgh, 2002; Bernard, 2007).

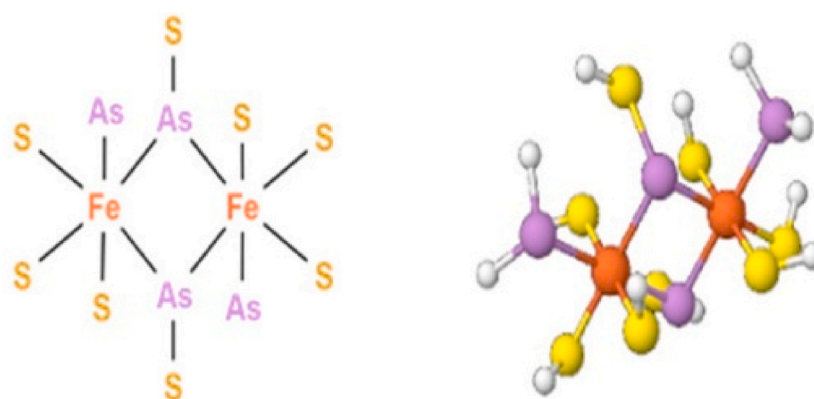


Fig. 2. Chemical structure and molecular structure of Arsenopyrite.

(even to hydrous aluminum and manganese oxides if in abundant presence) is a robust process, and the amount of sorbed mass could be noticeable even at low arsenic concentrations in solution. Most usual silicate ores enclose about 1 mg/kg or smaller Arsenic, and carbonate alloys generally have below 10 mg/kg of Arsenic (Smedley and Kinniburgh, 2002). The concentration and the structure of arsenic depend on multiple aspects, such as the existence of oxygen in the water, the degree of biological activity, the type of water source, and the closeness of the water supply to arsenic-bearing geological formations (Howe et al., 2001). The Arsenic presence in underground water can vary widely; in terms of concentration, Arsenic amount is comprised from 0.5 µg/L to 5000 µg/L. Higher arsenic concentrations are detected in spring water in various environments, such as oxidizing (alkaline pH values) and reducing aquifers; arsenic is also noticeable in regions affected by drilling and industrialized activity (Smedley and Kinniburgh, 2002). Arsenic is essentially present in water in Arsenate [As (V)] form, which is considerably less toxic than its trivalent form. This form can easily create solid interactions with the sulfhydryl group in protein cysteine residues and small molecules of organosulfur compounds such as glutathione (GSH). However, aquatic micro-organisms may degrade the arsenate to arsenite under precise reducing conditions. In an aqueous phase, the methylation of organic arsenic to methyl- and dimethyl arsenic acids is strictly related to biological activity (“WHO air quality guidelines for Europe, 2nd edition, 2000 (CD ROM version),” 2017).

Moreover, arsenate is the chemical analog of phosphate (P). It can

inhibit energy and metabolism by substituting P in phosphorylation reactions, As(III) attaches to the SH-containing (sulfhydryl) groups of proteins and polypeptides that interfere with their activity (Chakraborty et al., 2021). As(V) absorption via phosphate carriers had a high impact on human health, as previously shown in several studies, which reveal a connection between arsenic exposure and the predisposition to Mellitus type diabetes in high As presence areas. Arsenic is present in many oxidation states and different environmental forms, as shown in Table 2.

High concentrations of Arsenic have been detected as an effect of leaching and natural weathering of Arsenic from geological materials containing drainage from thermal sources and geysers, arsenopyrite, base metal sulfides, and atmospheric precipitate. Globally, pyrite, arsenic sulfides, and iron(III) hydroxide are groundwater’s most well-known raw sources of arsenic pollution. Several studies have

Table 2  
As species and oxidation states (Raju, 2022).

As species	Formula
Ferric arsenite	$\text{FeAs}^{3+}\text{O}_3$
Ferric arsenate	$\text{FeAs}^{5+}\text{O}_4$
Monomethyl arsonous acid	$\text{CH}_3\text{AsO}(\text{OH})_2$
Dimethyl arsonous acid	$(\text{CH}_3)_2\text{AsO}(\text{OH})$
Trimethyl arsine oxide	$(\text{CH}_3)_3\text{AsO}$
Arsenious acid	$\text{H}_3\text{As}^{3+}\text{O}_3$
Arsenic acid	$\text{H}_3\text{As}^{5+}\text{O}_4$

suggested how groundwater, originating from the dissolution of sulfur minerals rich in Arsenic or desorption from Iron oxides, can be considered the primary source of Arsenic (Garelick et al., 2008; Barral-Fraga et al., 2020; Björklund et al., 2020). The origin of global Arsenic groundwater pollution in natural sources has been accredited to several processes of geochemical nature, for example, Arsenic release from geothermal waters, desorption from oxides and hydroxides, oxidation of arsenic-containing sulfides, evaporative concentration, leaching of Arsenic from sulfides, and reductive dissolution from oxides/hydroxide (Welch et al., 2000); (Coppola et al., 2021). Arsenic groundwater pollution is estimated to involve more than 200 million people worldwide (Samal et al., 2021) The overview of the regions affected by Arsenic and the possible sources is reported in Table 3.

The main anthropogenic actions accountable for arsenic delivery are smelting of non-ferrous metals, ore dressing, electronic industries, mining, glass manufacture, chemical industries, pesticides, tanning industries, dye industries, burning fossils fuels, paints, pigments, cosmetics, fungicides, insecticides. Several anthropogenic sources of

**Table 3**  
Details of Worldwide As-contaminated groundwater and probable origins (Shaji et al., 2021d).

Region/ Continent	Natural/environmental conditions
<b>Africa</b>	<ul style="list-style-type: none"> <li>Sedimentary rocks containing gold minerals (Bretzler et al., 2017; Irunde et al., 2020);</li> <li>Alluvial residues/sediments (Edet et al., 2004);</li> <li>Volcanic origin Sulphide minerals (Bretzler et al., 2017).</li> </ul>
<ul style="list-style-type: none"> <li>Burkina Faso;</li> <li>Cameroon;</li> <li>Nigeria;</li> <li>Ghana.</li> </ul>	
<b>Europe</b>	<ul style="list-style-type: none"> <li>Superficial groundwater (Tamasi and Cini, 2004) and Hydrothermal (Vivona et al., 2007);</li> <li>Geothermal waters (Bundschuh et al., 2013)</li> <li>Geothermic arsenic around the volcanic canthers (Vivona et al., 2007);</li> <li>Limestone, sandstone, estuarine alluvium, mining (Millward et al., 1997);</li> <li>Alluvium sediments, mineralized sandstone (Heinrichs and Udluft, 1999);</li> <li>Sedimentary basin, ore deposits containing arsenopyrite.</li> </ul>
<ul style="list-style-type: none"> <li>Italy;</li> <li>Germany;</li> <li>UK;</li> <li>Turkey</li> <li>France;</li> <li>Spain.</li> </ul>	
<b>Asia</b>	<ul style="list-style-type: none"> <li>Holocene alluvial sediments (Hossain, 2006);</li> <li>Marine sediments trapped within continental aquifers (Maity et al., 2017)</li> <li>Mining activities (Roy et al., 2022);</li> <li>Groundwater and geothermic activities (Maity and Liu, 2011; Kar et al., 2012; Samal et al., 2013);</li> <li>High presence of hydrated ferric oxides (Rodríguez-Lado et al., 2013);</li> <li>Oxidation of arsenic-bearing pyrite or anoxic reduction of ferric iron hydroxides to ferrous iron (Chakraborti et al., 2009; Bhowmick et al., 2018);</li> <li>Material and volcanic ash (Liu et al., 2011; Ijumulana et al., 2021), Holocene seaside clays (Mitsunobu et al., 2013).</li> </ul>
<ul style="list-style-type: none"> <li>China;</li> <li>Taiwan</li> <li>Japan;</li> <li>Bangladesh;</li> <li>India;</li> <li>Cambodia;</li> <li>Pakistan;</li> <li>Russia.</li> </ul>	
<b>Australia/ Oceania</b>	<ul style="list-style-type: none"> <li>Pyrite residues, hydroxides and Iron hydroxides, gold mining (Smith and Smith, 2004; Appleyard et al., 2006);</li> <li>Geothermic water.</li> </ul>
<ul style="list-style-type: none"> <li>Australia;</li> <li>New Zealand;</li> <li>Guam.</li> </ul>	
<b>North America</b>	<ul style="list-style-type: none"> <li>Thermal spring, Holocene and basin-fill, Sulphide mineralization in volcanic rocks sediments (Dummer et al., 2015a; Bondu et al., 2017), Volcanic residues (Dummer et al., 2015b);</li> <li>Sulphide minerals dissolution (Flanagan et al., 2015).</li> </ul>
<ul style="list-style-type: none"> <li>USA;</li> <li>Mexico;</li> <li>Canada.</li> </ul>	
<b>South America</b>	<ul style="list-style-type: none"> <li>Volcanic and Sulphide-rich rocks (Bidone et al., 2016; Ciminnelli et al., 2017; Lima et al., 2019; Bundschuh et al., 2021);</li> <li>Volcanic deposits from tertiary and quaternary periods (Panigatti et al., 2014; Robles et al., 2016; Lima et al., 2019);</li> <li>Volcanogenic sediments from the quaternary period (Corradini et al., 2018);</li> <li>Adsorption-desorption of As from different sediments and the oxidation of sulfide minerals rich in As (Ormachea Munoz et al., 2015; Quino-Lima et al., 2020; Quino Lima et al., 2021).</li> </ul>
<ul style="list-style-type: none"> <li>Brazil;</li> <li>Argentina;</li> <li>Chile.</li> <li>Bolivia</li> </ul>	

Arsenic have been widely used during the last century; their use has been restricted today due to the damaging impact on health and the environment (Bundschuh et al., 2017). In particular, Chromated copper arsenate, a well-known wood chemical preserving agent containing a mixture of Arsenic, chromium, and copper, is regarded as the most significant anthropogenic arsenic pollutant. Anti-corrosive agents, leather tanning agents, glass and ceramic products, agricultural products, pesticides, textile chemicals, and paints that contained this compound were widely utilized in the past; however, nowadays, usage is prohibited in most of those chemicals. The Arsenic emitted from several anthropogenic activities differs significantly in chemical nature and bioavailability (Mahimairaja et al., 2005). An overview of the primary uses and chemical types of arsenic is shown in Table 4 (Jang et al., 2016).

Several environmental reactions involve arsenic as biotransformation, ligand exchange, precipitation, and oxide-reduction (Welch et al., 1988; Pongratz, 1998). Those dynamic processes strongly depend on the pH, temperature, salinity, metal sulfide, sulfide ion concentrations, iron concentration, and oxidation-reduction potential. Arsenic affects even the composition and the distribution of the complex of organisms that occupy a specific space in an ecosystem. Once extracted, the mineral, stable ores containing Arsenic are strongly exposed to oxidizing conditions ensuing in the release of Arsenic in surface waters and consequently to the environment; for this reason, the control of mining operations implemented by government departments is rigorous and increasingly strict. The EPA (Environmental Protection Agency) has defined the MCL (Maximum Contaminant Level) as the maximum acceptable limit of arsenic concentration to prevent prolonged arsenic exposure in public water systems and environments.

### 3. Effect of arsenic on human health

Arsenic is an abundant element in the environment; exposition to this compound is possible by air, groundwater and even by food consumption (Kar et al., 2012). As discussed, the leading causes of contamination can be traced in soils and various ordinary life actions (Bhattacharyya S. et al., 2022) (Itanna 2021). Arsenic has been proved to affect many body organ systems: its effects are evident on the skin, the respiratory (Kapaj et al., 2007), nervous and cardiovascular systems, plus it can affect the immune and endocrine systems and have adverse effects on liver functionality, kidneys, and have repercussions on the reproductive system (Mohammed Abdul et al., 2015). All the adverse effects of As on human health, including those mentioned above, are represented in Fig. 3.

Skin anomalies are the most recognizable mark of arsenic exposure.

**Table 4**  
Primary uses and chemical types of arsenic (Jang et al., 2016).

Field	Application	Chemical Type and formula
Agriculture	Insecticides, Herbicides, larvicides	<ul style="list-style-type: none"> <li>Monosodium arsenate (<math>\text{NaCH}_3\text{HASO}_3</math>)<sub>3</sub>,</li> <li>Disodium arsenate (<math>\text{Na}_2\text{CH}_3\text{AsO}_3</math>)<sub>3</sub></li> <li>Diethyl arsenic acid (<math>(\text{CH}_3)_2\text{AsOCOH}</math>)</li> <li>Arsenic trioxide <math>\text{As}_2\text{O}_3</math></li> </ul>
Wood	Chromated copper arsenate (CCA) wood preservative, decolorized glass	<ul style="list-style-type: none"> <li>Arsenic trioxide <math>\text{As}_2\text{O}_3</math></li> </ul>
Livestock	Feed additives, disease prevention, aquatic weed, cattle dips	<ul style="list-style-type: none"> <li>Sodium arsenite <math>\text{NaH}_2\text{AsO}_4</math></li> </ul>
Medicine	Treatment trypanosomiasis, Antisyphilitic medications, amebiasis, sleeping disease	<ul style="list-style-type: none"> <li>Arsine <math>\text{AsH}_3</math></li> <li>Arsenic acid <math>\text{H}_3\text{AsO}_4</math></li> </ul>
Industry	Catalysts, Glassware, electrophotography, dyes and soaps, Pharmaceutical substances, antifouling paints,	<ul style="list-style-type: none"> <li>Arsenic acid <math>\text{H}_3\text{AsO}_4</math></li> </ul>
Metallurgy	Battery plates, Alloys	<ul style="list-style-type: none"> <li>Arsine <math>\text{AsH}_3</math></li> </ul>



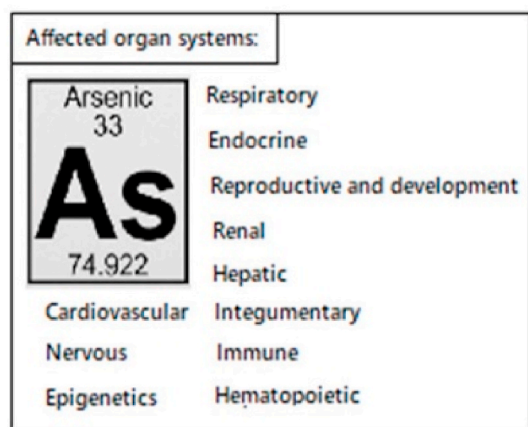


Fig. 3. As an effect on human health.

It is because human skin is considered as highly affected by arsenicosis, which is a chronic disease due to As contaminated water drinking, while highlighting its early manifestations in the body, like melanosis (Maity et al., 2012), pigmentation and keratosis (Rahman et al., 2009). These skin modifications typically do not appear 5 years after exposure (DN et al., 1998).

Skin pigmentation and palm and sole thickening were evident in 65% of patients exposed to arsenic water from the Indian region (DN et al., 1998). Another case study was conducted in Mongolia and China. Around a fifth of the studied population was exposed skin lesions and palm/sole hyperkeratosis, along with hyperpigmentation or depigmentation on chest skin for exposure to high amounts of arsenic in water (JX et al., 2007).

Arsenic can deposit in keratin-rich body parts, with evident effects like white lines on nails, called Mee's lines, and commonly even alopecia (RN, 2003; E et al., 2007). Neurological issues have been widely reported to acute or chronic arsenic exposure, often regarding sensorimotor polyneuropathy. Arsenic is more effective on nerves of the sensory system than on the nerves of the motor system. At the same time, some recurrent symptoms are ache and paresthesia in feet soles (Vahidnia et al., 2007; Spencer and Palmer, 2021); this is due to a reduced ability of neurons in glutathione formation and Reactive Oxygen Species (ROS) detoxification (Aoyama et al., 2008; Chou et al., 2021). Arsenic-driven neurotoxicity is mainly caused by oxidative stress (Mundey et al., 2013; Prakash et al., 2016). Fig. 4 shows how cytoskeletal disorganization and the consequent alteration of the protein composition and hyper-phosphorylation cause arsenic-driven neurotoxicity.

Arsenic exposure is found to cause cardiovascular diseases (CVD).

Smelting workers are at high risk of CVD because of their exposure to high Arsenic levels (Wang et al., 2007; Xu et al., 2013). High levels of arsenic in drinking water sources have severe effects on the cardiovascular system and other organs such as kidneys, liver, and lung (Ahmad and Bhattacharya, 2019a). Healthiness effects of Arsenic Longitudinal Study (HEALS) have established the arsenic effects on 20,000 people in Bangladesh; the resulting data showed how moderate and even low concentrations of As in the water resulted in increased mortality and chronic diseases, including CVD, among other reasons (Ahsan et al., 2006; Chen et al., 2009). Table 5, the baseline features of the study participants with incident CVD, heart disease and stroke (Chen et al., 2013).

In the broader study, the Environmental Protection Agency (EPA) has been working on cancer risk evaluation on the effect of inorganic arsenic (iAs). CVDs linked with exposure to iAs were the foundation for a reference dose (RfD) evaluation, the core of the risk assessment. Low-level exposure to iAs, ranging from 100 to 150 µg/L well water concentrations, and CVD studied on multiple cohorts in Bangladesh, China, Taiwan, and United States, and an RfD was evaluated. No adverse effect was observed under 100 µg/L arsenic concentrations in water, the equivalent to 0.009 mg/kg-day iAs dose. Population resulted sensitive to As effects due to dietary deficiencies acting on arsenic methylation and one-carbon metabolism while increasing the risk of CVD (Tsuji et al., 2014).

Table 5  
Features of the participants of the study.

Characteristic	CVD	Heart disease	Stroke
Participants (n)	369	211	148
Male (%)	74.0	70.6	77.7
Age (years)	48.5 ± 9.3	46.3 ± 9.5	51.5 ± 8.4
BMI (kg/m <sup>2</sup> )	20.1 ± 3.7	20.7 ± 4.0	19.4 ± 3.2
Education level (years)	3.9 ± 4.2	4.3 ± 4.3	3.2 ± 4.0
Cigarette smoking status (%)			
men smokers	88.6	88.6	89.6
Women smokers	19.8	24.2	12.1
Systolic blood pressure (mmHg)	130.7 ± 26.5	126.4 ± 24.2	137.6 ± 28.8
Diastolic blood pressure (mmHg)	81.7 ± 14.7	80.2 ± 13.3	84.5 ± 16.3
Diabetes status (%)	7.5	5.7	9.9
Well-water arsenic (µg/L)	104.2 ± 113.8	110.5 ± 123.4	95.5 ± 100.4
Total urinary arsenic (µg/g creatinine)	259.9 ± 235.7	268.0 ± 247.5	249.4 ± 220.6
Urinary iAs%	14.3 ± 6.1	14.5 ± 6.1	14.1 ± 6.2
Urinary MMA%	14.4 ± 5.3	14.5 ± 5.4	14.3 ± 5.3
Urinary DMA%	71.3 ± 8.1	71.0 ± 8.6	71.7 ± 7.6
PMI [MMA/(As <sup>III</sup> + As <sup>V</sup> )]	1.2 ± 0.6	1.2 ± 0.6	1.2 ± 0.7
SMI (DMA/MMA)	5.8 ± 2.9	5.8 ± 3.1	5.8 ± 2.7

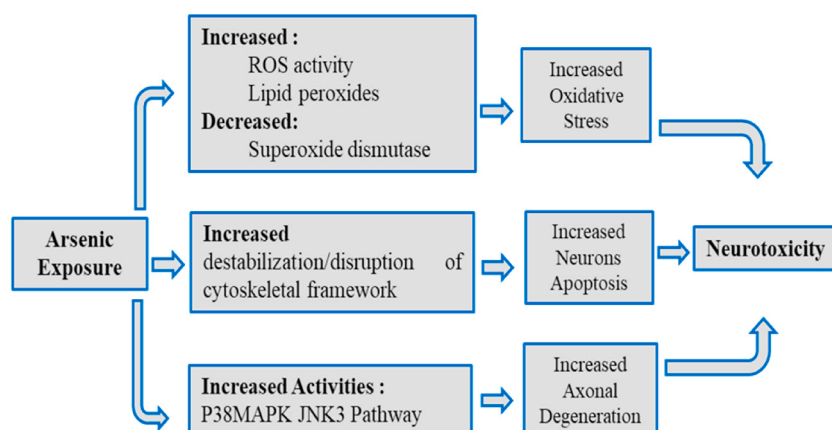
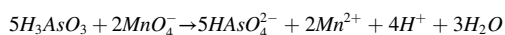
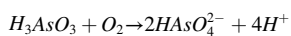


Fig. 4. Cytoskeletal disorganization and consequence of Arsenic neurotoxicity.

#### 4. Arsenic removal: membranes-based processes

Different methods are used for the As removal, and each feature is reported in Table 6.

Soluble arsenite is converted to arsenate Soluble arsenite is converted to arsenate Masscheleyn (2002) (Saha et al., 2019). This process does not permit the removal of arsenic, so other adsorption, coagulation, or ion exchange techniques are coupled to it. Different oxidizing agents used for oxidizing the arsenite to arsenate are used (Kim and Nriagu, 2000; Maity and Kar, 2011; Maity and Chen, 2021a). The most effective oxidants are permanganate, NaOCl, KMnO<sub>4</sub> chlorine, and ozone (Michael et al., 2006; Ahmad et al., 2019), and the reactions involved are:



Among these, ozone is frequently used being a good oxidant and a potent disinfectant. In developing countries, permanganate is used for its long shelf life, ease to recovery, and disinfectant property against common enteric bacteria (Mohanty, 2017).

The adsorption method is primarily used for the nanomaterials being easy to perform and highly efficient (Singh et al., 2021). Usually, activated carbon has been widely used for the removal (Mohan and Pittman, 2007), and the adsorption capacity depends on carbon characteristics, temperature, pH, and ionic strength. However, it is less selective for arsenic removal (a few milligrams of metalloid per gram of activated carbon) (Carneiro et al., 2021), and its recovery is complicated. As a result, this process is costly or it is arduous to utilize in developing countries (Ochedi et al., 2020). Activated alumina is also used for arsenic removal. Different study evidenced as the arsenite is adsorbed in the pH range 7–8, whereas the arsenate in the range 5–6 (Singh and Pant, 2004). In addition, nanomaterials prepared using iron and copper oxides exhibit arsenic adsorption capacity at pH values close to the neutral (M and M, 2015b). Other innovative materials, such as organic metal frameworks, graphene oxide, and carbon nanotubes, represent a good alternative for the As(V) and As(III) treatment showing better properties in terms of reuse and recovery and high adsorption capacity and so are an excellent route to follow for the As removal from wastewater (Liu et al., 2020). Another method efficient for the As removal from soil and groundwater is coagulation because it does not require pretreatment or preparation of the wastewater and the use of chemicals (Ge et al., 2020), (Cheng et al., 1994; Maity et al., 2019). Only the

pre-oxidation and pH adjustment are necessary. Initially, a metal-based coagulant (such as ferric chloride (FeCl<sub>3</sub>)) is added to the wastewater (Sancha, 2006a). The water hydrolyzes FeCl<sub>3</sub> for forming the Fe(OH)<sub>3</sub>, and the arsenite (As (III)) is oxidized to As(V). Arsenate has a negative charge, which is adsorbed by the Fe(OH)<sub>3</sub> particles being positively charged. Finally, the sedimentation and filtration processes permit the As remediation (Sancha, 2006b). Other coagulants used are zirconium (IV) oxy chloride, titanium sulfate titanium (III) chloride, zirconium (IV) chloride, and titanium (IV) chloride, and titanium (IV) oxy chloride (Rathi and Kumar, 2021).

Ion exchange is a process in which the ionic species are exchanged by means of the resins (solid phase) (Comstock and Boyer, 2014; Francesco et al., 2021). The ion exchange process allows obtaining an As concentration below 10 mg/L (Jain and Singh, 2012). The resins used exhibit high affinity towards As(V) because they are negatively charged, while the As (III) is neutral (SV et al., 2015a). Therefore, the pre-oxidation technique is required (SV et al., 2015b). This process is affected by several parameters such as the resin used, the concentration of the metalloid, the pH of the contaminated water, and the presence of other anions (i.e. Cl<sup>-</sup>, CO<sub>3</sub><sup>2-</sup>, SO<sub>4</sub><sup>2-</sup>). Electro-coagulation is an emerging electrochemical technique used to treat potable water, urban wastewater, wastewater containing pollutants, heavy metals, and dyes (Elazzouzi et al., 2019). Anode and cathode are used and connected to an external power supply in an electro-coagulation reactor. In the beginning, the oxidation occurs to the anode (i.e., aluminum and ferrous electrodes) with the generation of metallic cations. At the same time, the reduction of the water happens with the formation of H<sub>2</sub> (gas bubbles) and hydroxide ions (at the cathode) (Sandoval et al., 2021a). The metallic ions combine with hydroxyl groups to form amorphous metallic hydroxides (M(OH)<sub>n</sub>) in the water, which are very good adsorbents for pollutants (Maity and Chen, 2021b). The M(OH)<sub>n</sub> species make chemical bonds with the pollutants forming flocs (PV and TSA, 2017). Then, the flocs are removed from water by physical processes such as flotation, precipitation, and filtration (Sandoval et al., 2021b). Membrane processes used in water treatment are microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO). In these processes, a pressure difference between the two sides of the membrane is the driving force (Bruggen et al., 2003) (Dasgupta et al., 2015). In these processes, the membranes exhibit various pore sizes and can remove pollutants with dimensions from suspended particles (MF) to monovalent ions (RO). The characteristics of the pressure-driven membrane processes are reported in Table 7.

Low-pressure membrane processes do not permit the permeation of arsenic due to its ionic radius (0.24 nm), which is much smaller than the pore size range (10,000–1 nm). In addition, nanofiltration (NF) and

**Table 6**  
Arsenic removal methods (adapted from [(Yadav et al., 2021)]).

Method	As oxidation state	Efficacy	Advantages	Disadvantages
Oxidation-Precipitation	As (III)	High	- Simple	- Process very slow
	As(V)	Low	- Inexpensive - Kill microbes	- Coupled with other methods - Possibility to produce toxic compounds
Adsorption	As (III)	Low	- Ecofriendly	- Regeneration cost
	As(V)	High	- Cheap - Commercial available	- Need to operate at optimum pH
Coagulation-flocculation	As (III)	Low	- Cheap	- Toxic sludge produced
	As(V)	High	- Simple - Possibility to treat large volume of water	- Oxidation treatment required before
Ion-Exchange	As (III)	Low	- Commercial available	- Beads are fouled by suspended particles and organic species
	As(V)	High	- No Toxic sludge produced - pH independent	
Electro-coagulation	As (III)	High	- Cheap	- Anode changed periodically
	As(V)	High	- No chemical addition - No pollutant production	- Oxidation film on the electrode blocks the flow of the current.
Membrane	As (III)	High	- Very high efficiency	- Expensive
	As(V)	High	- No chemical addition - No pollutant generation	- Fouling

**Table 7**

Membrane separation processes used for water and wastewater treatment (Adapted from (Cheryan, 1998), (Barjoveanu and Teodosiu, 2006)).

Membrane Process	Driving force	Applied pressure (bar)	Membrane characteristics	Pore size (nm)	Species removed
MF	Pressure Gradient (Low)	1–3	Porous; Asymmetric or symmetric	100–10,000	Suspended particles
UF	Pressure Gradient (Low)	2–5	Microporous; Asymmetric	1–100	Macromolecules
NF	Pressure Gradient (High)	5–15	Finely porous Asymmetric and thin-film composite	0.5–2	Divalent ions Sugars
RO	Pressure Gradient (High)	15–75	Dense Asymmetric and thin-film composite	<0.5	Monovalent ions

reverse osmosis (RO) membranes easily remove the arsenic due to the minimal pore size of the membrane concerning its ionic radius (Mólgora et al., 2013). In NF membranes, the rejection combines steric, Donnan, dielectric, and transport effects. The transport of neutral solutes is due to the steric (mechanism (size-based exclusion) mechanism. The Donnan mechanism refers to the equilibrium and the interaction between the charged chemical species and the charge present on the membrane (Rodrigues et al., 2021). The ionizable groups on the surface and in the pores of the membranes generates the charges (Ernst et al., 2000). These chemical species are acids or bases, and their dissociations are affected by the solution's pH contact with the membrane (Ernst et al., 2000).

The first works on arsenic removal with membranes take place in the 90s (Waypa et al., 1997), (Uruse et al., 1998), (Seidel et al., 2004), (Kang et al., 2000). In a paper published in 2002, three different nanofiltrations (NF) membranes were used, and the characteristics and experimental data are reported in Table 8 (Y, 2002a). In particular, the rejection for the As (V) ( $H_2AsO_4^-$  or  $HAsO_4^{2-}$ ) is higher than 95%, while the trivalent arsenic ( $H_3AsO_3$ ) is very difficult to remove. This different behavior of the membranes is due to their negative charge and so can reject the arsenic (V) existing in monovalent and divalent forms.

Some years later, Saitua et al. used the NF membrane process for As (V) removal from different model solutions (Saitúa et al., 2005). The NF membrane module used was a thin film composite polyamide membrane (192-NF 300) in a spiral wound configuration (produced by Osmonic Inc.) and characterized by a molecular weight cut-off of 180 Da. The experimental results evidenced the possibility of removing the As(V), and the arsenic concentration in the permeate is 5–10 µg/L. The rejection of As(V) was between 93 and 99% for an initial arsenic concentration of 100 and 382 mg/L. The rejection resulted independent of the feed-flow rate, temperature, and transmembrane pressure. The arsenic removal mechanism, has been studied using a commercial NF membrane (Nguyen et al., 2009). In particular, in this research work, a negatively charged polyamide membrane (NE 90; manufactured by Woongjin Chemical, South Korea) with a nominal MWCO of 220 Da and resistant in the pH range 3–10. The zeta potential measurements on the membrane evidenced an isoelectric at the value of pH 3.3. Considering this aspect, the experiments were performed in the pH range 5–8 to ensure a negative charge on the membrane surface. The experimental results evidenced as the As(V) and As(III) are removed even if the arsenate is more rejected. This indicated that both ions experienced the steric exclusion but the arsenate for its negative charge is also subjected to the Donnan exclusion mechanism. In addition, the authors demonstrated that the arsenate removal increased with  $Cl^-$  and  $HCO_3^-$  being more mobile ions, and it decreased with  $SO_4^{2-}$  due to its low mobility. The rejection of As (V) from the model solution raised from 89% to 96% by enhancing the As(V) concentration from 20 to 100 µg/L. In addition,

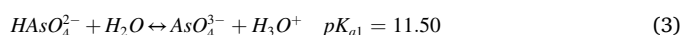
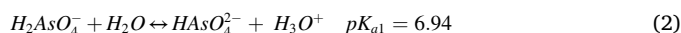
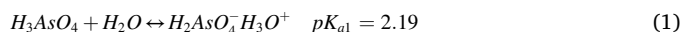
**Table 8**

As removal with three different NF membranes. Adapted from (Y, 2002b).

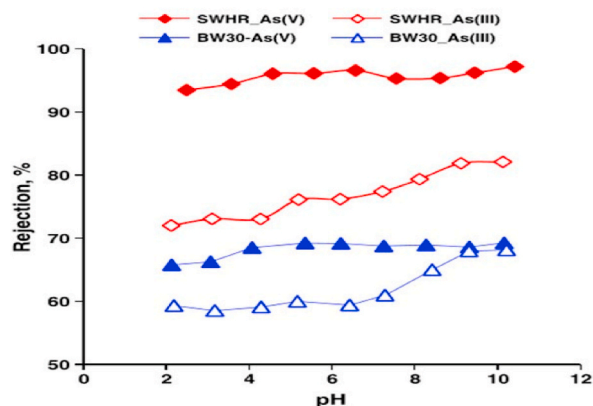
Membrane-type	Polymeric Skin layer	NaCl rejection (%)	As (V) rejection (%)	As(III) rejection %
ES-10	Aromatic polyamide	99.6	>95	>75
NTR-729HF	Polyvinyl alcohol	93.0	>95	<22
NTR-7250	Polyvinyl alcohol	70.0	>95	<22

the As (V) removal increased with the pH because the membrane's zeta potential became more hostile, increasing the electric exclusion. In 2015, Akin et al. studied the effect of different parameters such as pH and operating pressure on arsenic rejection using the SWHR and BW-30 membranes (FILMTEC) membranes (Akin et al., 2011). The influence of the feed water pH on the arsenic removal is illustrated in Fig. 5.

The experimental data evidenced as the rejection of the As(V) and As (III) rejection are heavily influenced by the pH of the feed water. For the considered membranes (SWHR and BW-30 membranes), As(V) and As (III) are rejected at pH = 4 and pH = 9, respectively. These results are due to the dissociation of the arsenic and arsenous acids at the different pH values (see equations 1-4) (Fischer, 1965).



The rejection of As(V) and As(III) increases with the transmembrane pressure being the driving force in the NF process. Finally, groundwater containing a total arsenic concentration of 62 µg/L was treated with the membrane. The total arsenic concentration in the permeate was about 2.86 µg/L, which was lower than WHO and EPA recommendation limit (10 µg/L). Song et al. prepared composite membranes with a hollow fiber configuration and a thin active layer in sulfonated poly(ether-ether ketone) (SPEEK) and a PES UF membranes. They are used for the arsenic removal (Song et al., 2015). The coating is permitted to reduce the zeta potential due to the deprotonation of the sulfonate group of SPEEK. A water permeability of 11.1 L/m<sup>2</sup>h/bar has been obtained, and the As(V) rejection enhanced from 89% to 96.4% by increasing the pH value from 4 to 9. This result is due to the anionic nature of the SPEEK membrane and the different dissociation states of the of As(V) by varying the pH of the feed solution. Finally, a techno-economical evaluation indicated that the treatment cost of groundwater containing arsenic using the proposed SPEEK coated hollow fiber membrane for a plant of 1000 m<sup>3</sup>/h was about 0.15 US\$ per m<sup>3</sup>. However, further improvement could be obtained by reducing both membrane and pretreatment costs. The fouling represents a huge problem in membrane technology because it causes

**Fig. 5.** The effect of feed water pH on the rejection of As(V) and As(III).

flux decline in time and a decrease of productivity, and a rising in energy consumption for the accumulation of organic, biological, and inorganic constituents on the surface and into the pores of the membranes (Algieri and Drioli, 2022). Different studies have explored the chemical modification of the membrane to enhance hydrophilicity and improve the antifouling capacity and separation performance (Kamaludin et al., 2022). Polysulfone (PSf) is one of the most polymeric materials used to prepare UF membranes, even if it does not have a hydrophilic character and surface charges. Blending the PSf with hydrophilic materials permits applying these membranes to remove small dangerous ions as the arsenic. In 2017, Nayak et al. functionalized the polyvinyl chloride (PVC) with L-cysteine (Cys) to obtain a zwitterionic polymer. Subsequently, the Cys-PVC polymer was blended with PSf, and TiO<sub>2</sub> nanoparticles have also been added to enhance the membrane charge further. Mechanical stability, the different composition of all synthesized membranes, is reported in Table 9 (Nayak et al., 2017a).

The best permeation performances obtained in the cited study are relative to an applied pressure of 500 kPa with a permeate flux of 35 L/m<sup>2</sup>h. The pure water flux decreased with the TiO<sub>2</sub> amount from 300 mg to 500 mg. In addition, the composite membranes showed higher arsenic rejection in the pH range investigated (from 4 to 9). Furthermore, for sample A<sub>1</sub>, the rejection is almost 100% in basic conditions because of the present negative charges provided by the cysteine and TiO<sub>2</sub> (Imhof, 2002). Increased TiO<sub>2</sub> concentration reduced the rejection for forming clusters in the membrane structure. A zwitterionic copolymer (P [MPC-co-AEMA]) soluble in water has been prepared via free radical polymerization (He et al., 2018), that presents a zwitterionic segment (containing both positively charged ammonium cations and negatively charged phosphate anion) and (2) a poly(AEMA) segment with amino terminal groups. Subsequently, the co-polymer has been incorporated into the polyimide selective layer of a thin film composite membrane via interfacial polymerization. The modified membrane has been used for the As removal. The prepared membranes exhibited higher water flux and rejection values than those obtained with the pristine membrane. In particular, the membrane containing 50% co-polymer exhibited an As (V) rejection of 99.8% and water permeability of 8.5 L/m<sup>2</sup>h/bar. The modified membrane exhibited stable performance for 180 h. Tanne et al. have studied the influence of pore dimension and surface properties of the membranes on the As removal (Tanne et al., 2019). In particular, three commercially NF membranes (NF90 (Dow FilmTec, USA), ESNA1, and ESNA1-LF2-LD (both from Nitto Denko, Japan) and a non-commercially available (M#1, Origin Water, China) have been used. The NF90 membranes displayed the highest rejection values for the arsenate. Using an arsenate concentration of 70 µg/L in the feed water and operating pressure of 5 bar, the rejections of arsenate are 91.1% for NF90, 86.2% for M#1, and 81.8% for ESNA1. These results are attributable to the negative surface charge of the membrane being very high for the sample NF90, followed by the M1 and the others. However, the M#1 sample has been used for the As removal from natural groundwater, which is less prone to membrane fouling than NF90.

Recently, PA blended membranes were prepared by varying the chitosan-clay nanoparticles (C-SBF) concentrations of (10–50 mg)

**Table 9**

Composition of the differently prepared samples (Nayak et al., 2017b).

Sample	Membrane	TiO <sub>2</sub> particles (mg)	Composition (%)		
			Cys-PVC	PSf	NMP <sup>(*)</sup>
A <sub>0</sub>	Cys-PVC/PSf	–	4	16	80
A <sub>1</sub>	TiO <sub>2</sub> (300 mg)/Cys-PVC/PSf	300	4	16	80
A <sub>2</sub>	TiO <sub>2</sub> (400 mg)/Cys-PVC/PSf	400	4	16	80
A <sub>3</sub>	TiO <sub>2</sub> (500 mg)/Cys-PVC/PSf	500	4	16	80

\* NMP stands for the solvent N-Methyl-2-pyrrolidone.

(Zeeshan et al., 2020). The composite membranes exhibited increased hydrophilicity that grew with the chitosan-clay content. The composite membranes showed higher permeation fluxes than the control due to the presence of the C-SBF hydrophilic nanoparticles (Jabur et al., 2016). The modified membranes showed the highest rejection values for the As(III) at pH = 9, while the lowest rejection has been found in an acidic environment. This result is interesting considering that AS(III) is removed with enormous difficulty by NF and RO membranes being neutral species. Recently, Ma and coworkers have fabricated highly electrically conducting ultrafiltration membranes to remove the As(III) (Ma et al., 2021). The UF membranes present a hydrophilic nickel-coated CNT ((Ni-CNT) layer deposited on PSF membranes that acts as a support. The application of cathodic potential to the surface of the membrane considerably determined an increase of the As(III) rejection in synthetic and natural tap water (R = 72.6% ± 5.1% when 7 V has been applied). The application of the negative potentials determined an increase of the local pH (production of OH<sup>-</sup> ions) that determined the transformation of the neutral arsenic species (H<sub>3</sub>AsO<sub>3</sub>) in its ionic forms (H<sub>2</sub>AsO<sub>3</sub><sup>-</sup>/HASO<sub>3</sub><sup>2-</sup>). So these last species are rejected by the negative charges present on the membrane surface. In Table 10, the application of various membranes for the As removal has been reported.

This section is evidenced as various nanofiltration membranes (some of these commercial and other synthesized) exhibited exciting performance in the arsenic removal (Algieri et al., 2021). However, a large-scale application is still in the early stage for the water flux decrease and membrane degradation owing to the fouling (Leonzio, 2018). Electrospinning is an exciting process for preparing nanofiber porous membranes for drinking water production and wastewater purification (Zhu et al., 2021a). This process has the advantages of being economical, easy to use, and high efficient (Pereao et al., 2019a). In addition, the nanofibers exhibit 3D interconnected pore structure, high

**Table 10**

Arsenic removal efficiency (%) by commercial and synthesized membranes.

Membrane Used	As removal efficiency (%)	Ref.
ZVI-Kaolin and PES	As (III): 50	[(Selvan et al., 2022)]
Cu/CuO/poly(ethylene terephthalate) composites	As(III): 48.70	[(Mashentseva et al., 2020)]
5% Zn/Al <sub>2</sub> O <sub>3</sub> /Polysulfone	As(V): >87	[(Sherugar et al., 2021)]
P-60S-EDTA membrane <sup>°</sup>	As(V): >96.75	[(Roy et al., 2020)]
PPSU/ZrO <sub>2</sub> /CA hollow fiber membrane**	As(V): 87.24	[(Kumar et al., 2020a)]
PPSU/ZrO <sub>2</sub> /CAP hollow fiber membrane**	As(V): 70.48	[(Kumar et al., 2020b)]
NF- flat sheet membrane	As(III) 72	[(Elcik et al., 2015a)]
RO-membrane	As(III) 99	[(Elcik et al., 2015b)]
Composite membrane with aromatic polyamide Selective layer	As(V): 40	[(Chang et al., 2014)]
NF-300 TFC polyamide	As(V): >95	[(Saitua et al., 2011)]
NF90-4040 Polyamide TFC <sup>°</sup>	As(V): 94	[(Harfoush et al., 2018a)]
NF90-4040 Polyamide TFC	As(III): 90	[(Harfoush et al., 2018b)]
Dow/FilmTec NF90 Polyamide	As(V): 98	[(Jadhav et al., 2016)]
TFN <sup>°</sup>	As(V): 98.6	[(He et al., 2017)]
AF-NF <sup>°</sup>	As(V): >72	[161]
NF-90 membrane	As(V): 98.4	[(Figoli et al., 2010)]

\* Amino functionalized ceramic-supported-polymeric composite NF membranes; \*\*PPSU = polyphenylsulfone, CA = cellulose acetate, CAP = cellulose acetate phthalate; ° NF-90 = Polyamide thin film composite membrane (Dow Chemical); ° TFC = Thin film composite membrane; TFN = Thin film nanocomposite membrane (0.15 wt% UiO-66 (MOF)); AF-NF = TFN aliphatic amino membranes -functionalized with multiwalled carbon nanotubes.



specific surface area, ease of surface functionalization, and recycling (Pereao et al., 2019b). An electrospinning set-up, illustrated in Fig. 6, presents high voltage power supply; an electrospinning setup is constituted by a high-voltage power source, a syringe pump for generating the polymer solution jet, a grounded collector, a spinneret electrically conductive, and a polymer solution.

The properties of the prepared nanofibers depend on voltage, feed rate and air gap (processing parameters), temperature and humidity (environmental parameters) (Sanders et al., 2019) and polymer solution parameters (concentration, solution viscosity, polymer's molecular weight, and solvent's evaporation) (Medeiros et al., 2008). The researchers have focused on developing mixed matrix electrospun nanofibrous membranes (ENMs) inorganic fillers dispersed into polymeric nanofibers owing to their improved Physico-chemical properties and separation performance.

Nanofiber membranes made in polyvinylidene fluoride and loaded with specific adsorbents (nanoparticles of titanium dioxide-halloysite nanotubes) have been used for the removal of As(III) from contaminated water (Moslehyan et al., 2020). A maximum amount of As (31.2 mg/g) has been adsorbed by using a mixed ENM loaded with a 0.5 wt % of adsorbents.

In some cases, accomplishing consistent removal rates over sustained periods requires a regeneration-free process, like ion exchange and adsorption. It is worth mentioning that these procedures can be slowed down by high salinity and the presence of organic matter. This, said the treatment choice should consider the ability to eliminate either salinity and organic matter. This, is why NF/RO membranes could well serve many As contaminated waters (Boussouga et al., 2021). Recently, novel adsorbent mixed matrix membrane consisting of a polycaprolactone matrix with iron-intercalated mont-morillonite filler has been synthesized (Peña et al., 2021). The prepared fibers had diameters of  $212.04 \pm 98.48$  nm and showed attractive adsorption capacity towards the arsenic from wastewater.

Recently, Torasso et al. have developed electro spun membrane comprising super paramagnetic iron oxide nanoparticles (SPIONs) loaded in PVA nanofibers (PVA-SPIONs) [(Torasso et al., 2021)]. The experimental data confirmed a maximum adsorption capacity of 52.5 mg/g for the sample containing the 0.14 wt % of SPIONs. PVA-SPIONs membrane did not present defects at the microscopic level, and it is constituted of crisscrossed bead-free nanofibers. After the adsorption tests, a change in the fiber morphology was detected (see Fig. 7).

Recently, another innovative technology for arsenic removal explored graphene-based materials due to its physical and chemical properties, high surface area, and structure (Yoon et al., 2017), (Su et al., 2017), (La et al., 2017). Leaper et al. demonstrated the possibility to remove arsenic by utilizing electro spun polyvinylidene difluoride

membranes loaded with reduced graphene oxide (Leaper et al., 2021). The membrane loaded with an amount of GO equal to 2 wt % exhibited higher flux than the pure polymer, with an As(III) rejection of about 99.9%. In this field, some challenges should be overcome, such as the pore size, the porosity, and the mechanical strength of the ENM improvement for ensuring to their large-scale production and application.

## 5. Membrane fouling

Membrane fouling represents the main disadvantage of the membrane technology because it determines a flux decrease, a permeate deterioration, and an increase of the costs due to the increase energy demand, chemicals, and numerous cleanings (Mohammad et al., 2015). It is caused by the accumulation on the surface and into the membrane pores. In particular, it determines the deposition of organic (colloids, proteins, polysaccharides etc.), biologic (viruses, algae, bacteria, and other microorganisms), and inorganic constituents (salts) on the membrane and into its porous structure. Different factors influence the fouling wastewater chemical composition, membrane properties, and operating conditions (for example, pH and temperature) (Manawi et al., 2016). A possible way to reduce the fouling is the pretreatment of the water before the membrane process to remove most of the foulant. Usually, as membrane processes are chosen microfiltration and ultra-filtration enable the removal of larger pollutant that can clog the membrane used in NF and RO processes (Abdelkader et al.,). The fouling reduction is obtained utilizing physical and chemical cleaning. One of the most used physical cleanings is used during the process of water flushing (Wu et al., 2018). Carnevale et al. studied the treatment of the olive milling wastewater by DCMD and VMD processes. The authors demonstrated that cleaning the membrane with deionized water at 60 C for 15 min permitted a flux recovery up to 92%. Physical cleaning is used for treating reversible fouling, while, for irreversible fouling, chemical cleaning is required (Poojammong et al., 2020). The chemical agents used are acid, base, chelating, and surfactants. Zhao and coworkers studied the effect of three different chemical agents (NaOH, EDTA, and sodium dodecyl sulfate) for cleaning hollow fiber membranes used for wastewater treatment (Zhao et al., 2017). They found that cleaning with SDS is more efficient than NaOH and DTA one. Recently, Ren et al. demonstrated that the separation of monosaccharides and divalent salts by the NF process has improved post chemical cleaning. In this study, different chemicals were used as H<sub>2</sub>SO<sub>4</sub>, NaOH and NaClO, and the use of NaClO showed a better result regarding membrane regeneration (Ren et al., 2022).

Hydrophobic membranes are more prone to fouling than hydrophilic ones. Considering this aspect, improving the membrane surface hydrophilicity has been considered an important route to follow fouling mitigation (Yuan et al., 2014). Various grafting, coating, and blending techniques are applied to modify the membrane surface (Díez and Rosal, 2020). The grafting permits hydrophilic chains on the membrane surface by forming chemical bonds (Lee et al., 2018). This method is easy to perform and with the possibility to have high chemical stability, but at the same time is determined an increase of the membrane cost, and it is not accessible to scale up (Lee et al., 2018; Wang et al., 2018; Russo et al., 2021). The surface coating is a simple, cheap, and environmentally friendly method for obtaining a hydrophilic membrane (Li et al., 2014). In addition, polymers with high molecular weight are utilized to avoid the penetration of the coating into the membrane pores. Du et al. compared the performance of two membranes, a hydrophobic PVDF and PVDF coated with a PVA (hydrophilic polymer) (Du et al., 2018). They demonstrated as hydrophilic-coated membrane exhibited better performance for various treatment cycles. Polymeric materials or inorganic fillers, characterized by hydrophilic properties, are used as additives during membrane formation to manipulate the membrane properties (Alenazi et al., 2017). In this technique, the main drawbacks are the less compatibility between the additives and the polymeric materials and the

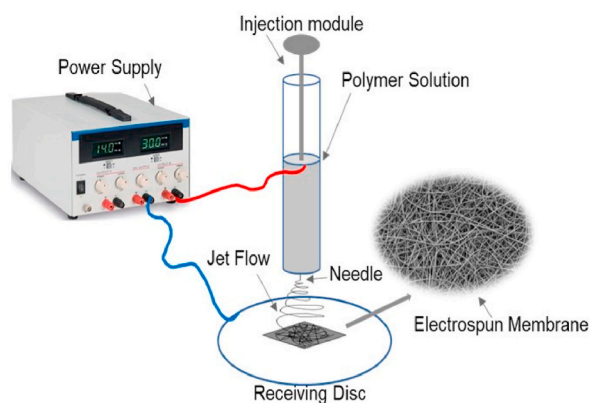


Fig. 6. Electrospinning set-up and SEM picture of the electrospun nanofibrous membrane (Zhu et al., 2021b). Reproduced with permission of Elsevier. B. V. (2021).

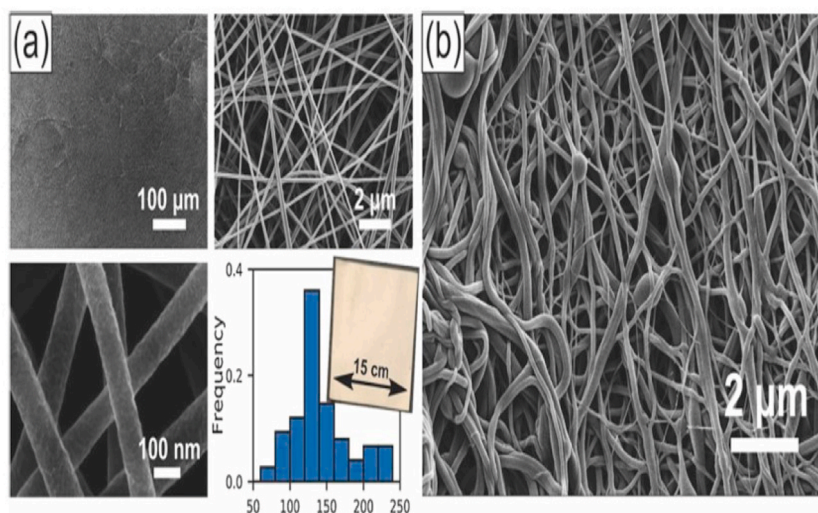


Fig. 7. (a) SEM images of the PVA-SPIONs membrane; (b) PVA-SPIONs fibers after arsenic adsorption.

leaching of the blended materials during long-term operations (Shen et al., 2020; Asiri et al., 2021). Usually, PVP or PEG are added to the polymeric solution to improve the membrane performance (Malik et al., 2019). The prepared membranes exhibit an increased water flux because these two pore-forming polymers induce a finger-like structure (Malik et al., 2019). In addition, different inorganic fillers (TiO<sub>2</sub>, MOFs, Zeolite, carbon molecular sieve, porous silica) are added to the polymeric membranes for preparing the mixed matrix membranes (Malik et al., 2019). The fillers modify the polymer-free volume by altering the packing of the polymeric chains.

## 6. Conclusions

The presence of arsenic in groundwater represents a problem for human health worldwide for its high toxicity. Inorganic As in drinking water causes different cancer types but also cardio-vascular, pulmonary, immunological, neurological, and endocrine diseases. NF membrane process shows high As rejection values and water flux operating at sustained pressure difference values. In particular, the rejection values obtained using commercial and synthesized NF membranes are very high for the As (V). However, removing the arsenite (As(III)) is complicated, meaning low rejection values. This different behavior is due to the membrane's negative charges that can reject the arsenic (V) by existing as monovalent and divalent ions; arsenite is a neutral species. Commercial NF membranes present different drawbacks (the most relevant are the fouling, the efficiency for long-term, and the reduced arsenite removal). The fouling mitigation is possible by following different ways as the cleaning and physical cleaning, wastewater pre-treatment and the utilization of more hydrophilic membrane. In addition, in the last years, the researchers have focused on developing the electrospun nanofiber membranes that seem to be very promising for arsenic removal from the groundwater. It is due to their 3D interconnected pore structure, high specific surface area, and ease of surface functionalization and recycling. However, their production at a large scale and the electrospinning process improvement represents the key drawbacks in this field.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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