

Electrodialytic removal of arsenic from wastewater: a mini review of the present state of research

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Arsenic is among the most damaging pollutants found in wastewater and as such requires special attention. It is highly toxic, carcinogenic and exposure to it has been linked to several respiratory, gastrointestinal, cardiovascular, renal and dermal conditions in humans. As such, strict environmental regulations are in place to maintain arsenic levels in groundwater to the lowest amount possible. There has been extensive research into strategies to remove arsenic from wastewater. Membrane separation processes have been an important part of research into the implementation of wastewater treatment at a scale large enough to meet the demand for human consumption. Among membrane separation processes, electrodialysis is a relatively new, yet especially appealing process due to significantly lower energy costs in comparison to the more commonly used reverse osmosis process, combined with high separation efficiencies for charged ions, making it more economically feasible for implementation. This paper gives an overview of arsenic, its sources, effects on human physiology, as well as an overview of the strategies in place for arsenic removal from wastewater. Further, the paper reviews the most recently published research into the area of arsenic removal from wastewater using electrodialysis, in order to provide a holistic view of the present state of research in the area.

Keywords: Electrodialysis; Arsenic; Membrane separation; Wastewater treatment.

INTRODUCTION

Arsenic is a highly toxic metal that can exist in water in two forms: arsenites, As(III), and arsenates, As(V). In either form, it is highly toxic and detrimental to both human and animal health. Out of those two forms, As(III) is more prevalent and also more toxic, but can easily be converted to As(V) *via* oxidation. The speciation of As(V) with pH is outlined in Fig 1 [1].

Under oxidizing conditions, such as surface water bodies, a commonly encountered compound is arsenic acid (H_3AsO_4) which can be further dissociated into H_2AsO_4^- , HAsO_4^{2-} , or AsO_4^{3-} . The trivalent form (H_3AsO_3), however, is mostly observed in reducing conditions, like deep sections of groundwater, where it is likely to dissociate into H_2AsO_3^- , HAsO_3^{2-} or AsO_3^{3-} [2].

Arsenic has been reported to cause vomiting and diarrhea on short-term exposure, and in the long term has been reported to cause neuropathy, impairment of central nervous system functions, failure of kidneys and liver, as well as deficits in locomotion, learning ability, and other cognitive functions, and even cancer [2].

As such, its concentration has to be controlled and restricted to as low a value as possible, especially when it comes to water consumed by humans and animals. It has in fact been classed as a

group 1 human carcinogen by the WHO [2]. Arsenic can affect humans either by ingestion or by inhalation, but given that it is oral ingestion that is the main source of arsenic toxicity in humans, it is the removal of arsenic from water that takes precedence when it comes to controlling arsenic-based toxicity [3].

The fact that arsenic is present in several industrial effluents, including dye industry, coal mining, thermal power plants, gold mining, and glass manufacture, makes it a difficult task to constrain arsenic and its effects downstream from such industries [4].

This paper outlines the sources of arsenic, their effects on human physiology, and strategies for their removal, as well as gives a review of the most recently published research on the use of electrodialysis as a viable method for arsenic removal from wastewater.

SOURCES OF ARSENIC

Arsenic can make its way into the water meant for human consumption *via* several sources, some being natural and some as a direct result of human activity. The concentration of arsenic can vary anywhere from 10 $\mu\text{g/L}$ to 150 $\mu\text{g/L}$ depending on a number of factors [1].

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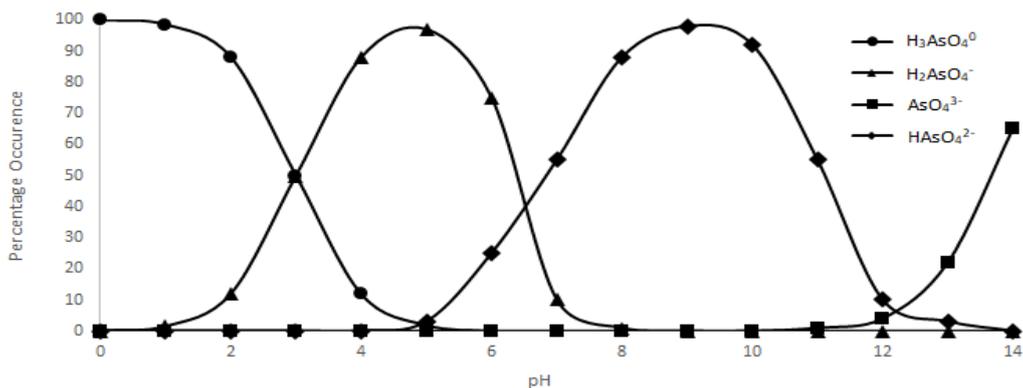


Fig. 1. As(V) speciation vs pH at 298 K [1]. Reproduced with permission from the copyright holder.

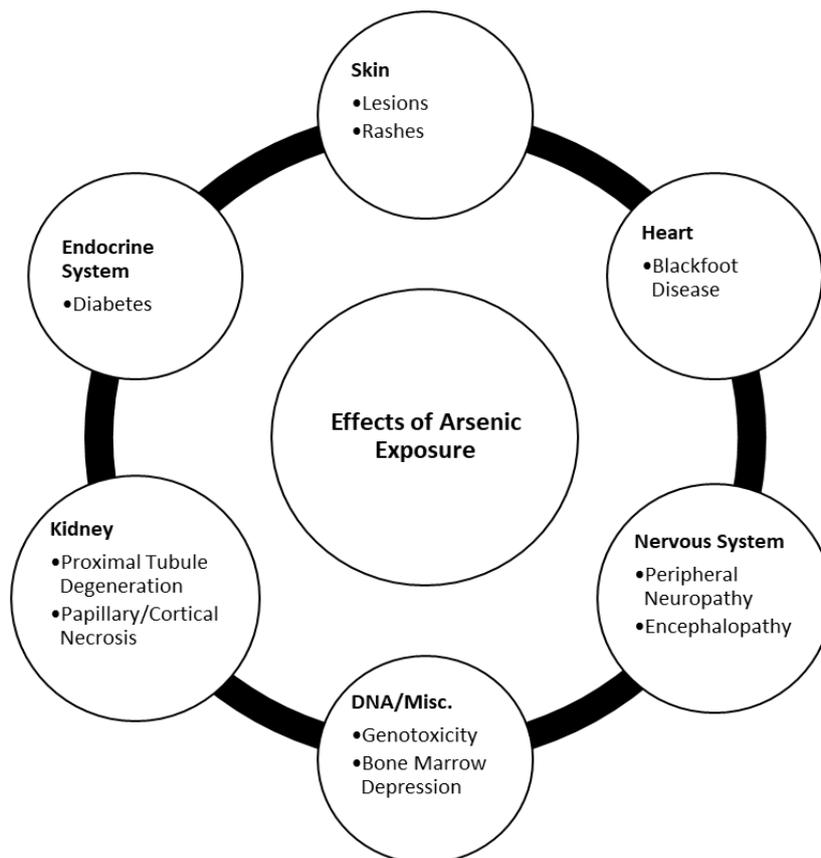


Fig. 2. Effect of arsenic exposure on human physiology.

Natural sources of arsenic

Arsenic is present in sedimentary rocks, especially in mountain ranges such as the Himalayas, as well as in igneous rocks and sulfidic ores [5, 6]. From these sources, by the mechanism of desorption from mineral rocks under alkaline conditions or release under anaerobic environments, the arsenic can reach water sources by a number of pathways [6]. The level of arsenic, as well as the concentration released varies significantly depending on the unique mineral makeup of the specific geographical location, for instance, rock deposits with high organic matter content are likely to contain a higher concentration of arsenic [7].

Other than this, geothermal water deposits, as well as locations close to frequent volcanic activities have also been observed to contain significantly high arsenic concentrations.

Anthropogenic sources of arsenic

Industrial activities. While it is clear from the previous section that arsenic does exist naturally, anthropogenic sources significantly multiply the severity of arsenic contamination and are the primary cause of arsenic levels increasing above recommended safe levels in water sources [8].

Industries responsible for the release of arsenic in effluents include mining industries, where arsenic mineral deposits are dissolved and leak into the

water table as a consequence of mining operations, and metallurgical industries, where smelting operations produce significant levels of arsenic which is released as effluent into water bodies [9–11]. Among these, mining operations are considered especially harmful not only due to arsenic release but also due to the release of several other heavy metals such as Zn, Cd and Ni [12].

Apart from these, tanning industry, as well as dye industries have also been observed to significantly contribute to arsenic toxicity. In both industries arsenic is used in various steps of the manufacturing processes and as a result the effluents from these industries usually contain high levels of arsenic unless the effluent is properly treated before release [13].

Agricultural activities. In the agricultural industry, arsenic toxicity has been historically caused by blatant overuse of pesticides, as well as insecticides containing arsenic as an ingredient. The arsenic from these sources can easily seep into the ground water, or can get absorbed by the crops and make its way up the food chain. These activities, however, have largely been phased out and replaced with less toxic alternatives [8]. Other agricultural activities such as farming of tobacco at scale are still widely associated with significant arsenic contamination to nearby soil and water bodies [14]. Further, the end product use, i.e. smoking of tobacco cigarettes, has been associated with increased

arsenic levels in the user’s lungs and an increase in the likelihood of tumour growth as a result of arsenic exposure [15].

Strategies for arsenic removal

Fig. 3. outlines the presently available methods of arsenic remediation and removal.

Pre-oxidation

Among the available methods, pre-oxidation is the simplest and easiest to implement method, wherein As(III) is converted to As(V). As(V) is significantly less toxic in comparison to As(III) and is also easily adsorbed onto solid surfaces, and thus is easily removed. Pre-oxidation is carried out in a number of ways, such as aeration, application of UV-rays, microbial activity, or use of chemicals such as chlorine and hydrogen peroxide.

Major parameters to control in this process are the oxidant used, its concentration, the presence of a catalyst, the temperature and the reaction time. [16]. This method is fairly easy to implement, however is held back due to difficulty in scaling the process as per requirement, slow reaction kinetics making the process take even weeks to achieve sufficient levels of purification, and production of toxic by-products such as bromate, iodate, products of organic matter oxidation, etc.

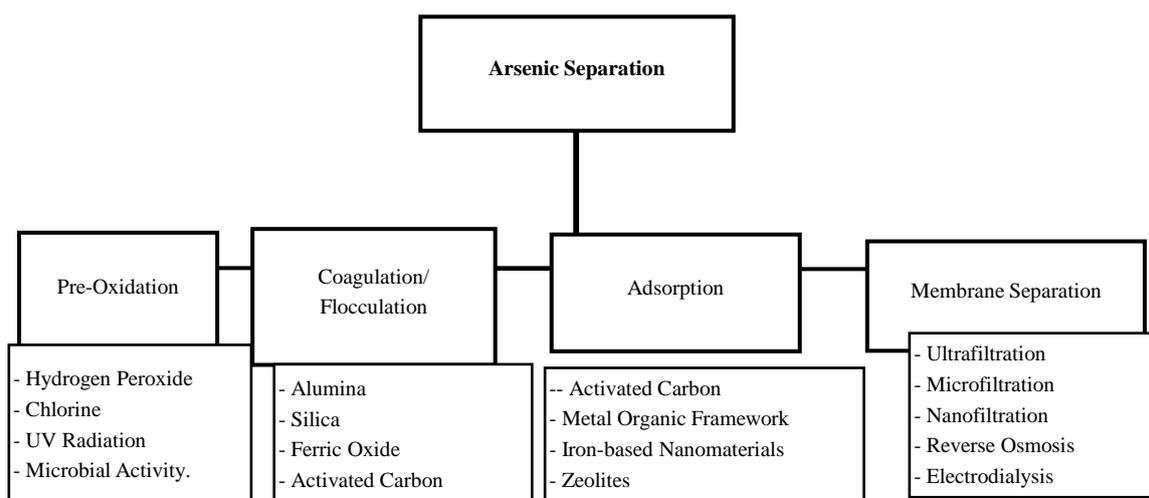


Fig. 3. Available methods for arsenic removal.

Coagulation-flocculation

Coagulation-flocculation is a much more industrially viable method; it involves addition of coagulant to the feed, causing agglomeration and subsequent precipitation into floc, resulting in removal of arsenic from the water [17, 18].

Coagulants such as alum and ferric chloride are the most commonly employed for this process. Apart from ferric salts, aluminium-based coagulants are among the ones most commonly used commercially [18, 19]. However, the separation efficiencies reported vary widely between plants (anywhere from 6% to 74%), depending on the coagulant used, dosage, and intensity of stirring and temperature [20]. This process is fairly efficient in arsenic removal from soil and wastewater, however, the method requires pre-oxidation, as well as pH control of the process in order to work as needed.

Adsorption

Adsorption and ion exchange are scalable and widely used methods for arsenic removal. Activated carbon is the most commonly employed adsorbent in arsenic separation applications, however the cost considerations of using activated carbon necessitate the need for research into cheaper or easily reusable adsorbent materials [21, 22]. These include metal organic frameworks, iron-based nanomaterials, graphene-oxide and iron oxide [23, 24]. This process is highly efficient, but is held back by cost considerations due to the need to constantly cycle adsorbents into the system [25]. Further, many materials such as commercial activated carbon are extremely cost restrictive, especially in the context of use in developing countries [18]. Due to this, extensive research efforts have been concentrated on low-cost sources of activated carbon with varying results [26, 27]. These include activated carbon derived from agricultural wastes such as rice husks, coconut husks, carbonized wood powder, sawdust, by-products of juice production, etc. [28-34]. Although a significant reduction in cost is achieved, due to the nature of the source materials, the availability of raw material is insufficient to sustain a scaled-up process.

Presently, metal oxides appear to be the most commercially viable options for this process due to easy access and lower costs. This has, in fact, been commercially applied to arsenic removal by Bayer AG, a German chemical company [18]. A simplified scheme of the SORB-33 treatment process is given in Fig. 4 [35]. The feed is introduced through the top

of the tower, passes the proprietary adsorbent media (G33 EFO), and purified product is obtained from the bottom of the setup. Adsorption processes report 95%+ efficiencies when the nature of adsorbent used, residence time, as well as temperature and pressure conditions are optimized.

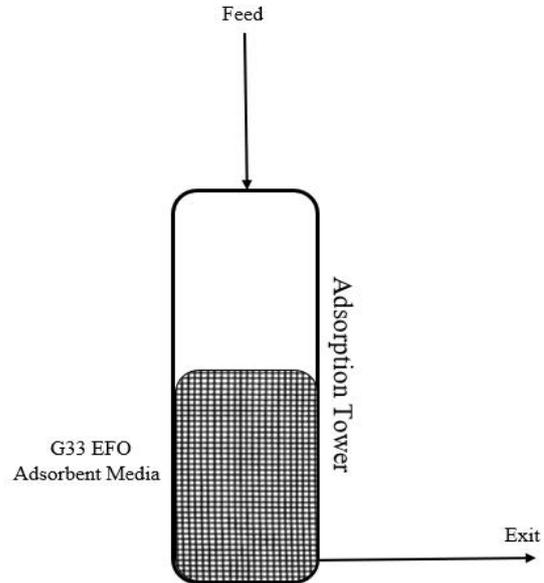


Fig. 4. Scheme of SORB-33 treatment process for separation of arsenic from wastewater.

Membrane separation

Membrane separation for arsenic removal has been gaining increased industrial adoption and research interest over time. It presents an environmentally benign, scalable and easy to implement alternative to the more conventionally employed methods, while maintaining high separation efficiencies. Several membrane separation processes are employed on a commercial scale, such as ultrafiltration, microfiltration, nanofiltration and reverse osmosis [16]. However, NF processes are considered the most competitive due to charge exclusion mechanisms that are unique to that process, resulting in high separation efficiencies. In case of arsenic removal, both NF and RO have been observed to achieve arsenic rejection of above 99%, with comparable separation efficiencies for both As(III) and As(V) [36, 37]. Further, NF has also widely substituted RO based on the significantly lower energy costs of NF processes compared to RO [38, 39]. An overview of the comparative effectiveness of all membrane separation processes towards As(III) and As(V) removal is outlined in Table 1.

Table 1. Comparison of different MSPs for arsenic removal from drinking water.

	Reverse osmosis	Nanofiltration	Ultrafiltration	Microfiltration
As(III)	Very effective	Possibly effective	Not effective	Not effective
As(V)	Very effective	Very effective	Possibly effective	Not effective

Further, more innovative membrane separation techniques such as electro-ultrafiltration, forward osmosis, as well as electrodialysis have been employed, with promising results [40-45].

PRESENT STATE OF RESEARCH INTO ELECTRODIALYSIS FOR SEPARATION OF ARSENIC INTO WASTEWATER

Research into membrane separation technology

The use of electrodialysis for arsenic removal is still a developing field, and hence the research into the area is scarce in comparison to other methods. Electrodialysis has either been used as a stand-alone process [46-52], or in conjunction with other processes [53-55].

The electrodialysis stacks used in research vary anywhere between 3 compartments to 20 compartments. Fig. 4 outlines the ED setup used by Rathi *et al.*, consisting of three compartments, with the middle one being the feed compartment where the feed is introduced. As electric potential is applied to the system, arsenic moves across the AEM towards the adjacent compartment, and hence can be collected separately from the purified product [55].

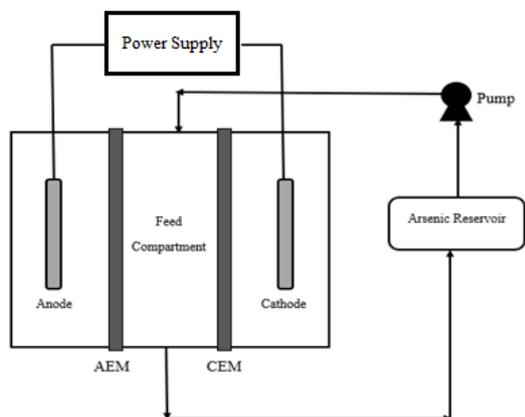


Fig. 5. Schematic setup used for electrodialytic separation of arsenic [4].

Among membranes used for separation, majority of the research has been carried out using commercially available membranes, most notably Neosepta (ASTOM Corp.) AMX and CMX membranes.

Aliaskari *et al.* in their publication used a 20-compartment stack of Neosepta AMX and CMX membranes for the electrodialytic removal of nitrate,

fluoride and arsenic. Effects of a wide range of operational parameters such as flow rate, operational voltage, feed pH and salinity were also tested. Arsenic removal increased with increasing pH but decreased significantly with an increase in salinity. The system was able to reduce the arsenic levels to below the governmental guidelines [46]. Another paper, by Onorato *et al.* used an electrodialytic stack, consisting of 6 anion exchange membranes and 7 cation exchange membranes, for the removal of trace contaminants from brackish water, while studying the effect of applied voltage, as well as solution pH. The membranes were commercial, manufactured by ASTOM Corp. The separation performance was enhanced with increasing applied voltage. As for pH, the separation performance for monovalent ions was found to be pH dependent, while the separation performance of divalent ions was independent of pH. [47].

Studies have also been carried out for enhancement of electrodialysis performance using different substances, in case of Babilas *et al.*, EDTA was used to enhance separation performance of a 5-compartment electrodialysis cell *via* metal complex formation [51]. Whereas the work by Xu *et al.* studied the removal of arsenic from a reverse-osmosis concentrate product, studying the impacts of coagulants, dosing, and pH on the separation performance, with highest separation achieved at 190 A/m². As a bonus, despite expected fouling, none was observed over the course of the research [48]. Another publication by Choi *et al.* used electrodialysis for removal of heavy metals including arsenic from groundwater using an 11-compartment electrodialysis cell, achieving up to 99.2% separation efficiency in the system [40].

Another publication by Pham *et al.* studied the effects of operational conditions on arsenic removal in an ED stack consisting of 10 cell pairs. The removal efficiency achieved was upwards of 96% when the initial concentration of As(V) was kept at 60 mg/L, while it reached 92% when the initial concentration of As(III) was kept 5 mg/L. The mass-transfer coefficient was observed to increase with increase in the discharged voltage, and the mass-transfer coefficients of As(V) were always higher in comparison to As(III) [56].

In terms of electrodialysis being used in conjunction with other processes, a publication by

Rathi *et al.* attempted a hybrid electro dialysis and ion exchange process, using a 3-compartment stack of one AEM and one CEM, with the middle (feed) compartment filled with anion exchange resin. The setup was able to achieve up to 100% removal of arsenic with the combination of the two processes [55]. Another research by Oehmen *et al.* studied a hybrid ED-coagulation process for arsenic removal. The system consisted of a two-compartment cell separated by an AEM. FeCl₃ was used as a coagulant to trigger precipitation of arsenic in the concentrate cell. The system was capable of achieving as low as 6.6 ppb arsenic levels [54]. Yet another attempt at a hybrid process was made in a publication by Ortega *et al.* wherein a hybrid ion exchange resin-electrodialysis system was used for arsenic removal from water. The system used was a five-compartment cell, with ion exchange resins in the middle compartment – similar to the work by Rathi *et al.* The system was applied to a feed containing up to 15 mg/L of arsenic, and was able to reduce the arsenic levels to below 10 µg/L, reaching current efficiencies of up to 47.7% and energy consumption as low as 7.5 kWh per kg of arsenic separated [53]. While studies on use of commercial membranes are available, ones with new membrane materials are rare. One such paper, by Bhadja *et al.* used polythene interpolymer membranes lab- fabricated for arsenic separation. The fabricated membrane showed superior fluoride removal capabilities compared to commercially available membranes and competitive arsenic separation performances as well [52]. Further, modelling has also been attempted on electro dialytic systems being used for arsenic removal. Honarparvar *et al.* developed a 2-D model based on Nernst-Planck equation and electroneutrality conditions. The results indicated an improvement in the selective removal of divalent ions with decreasing the cell length, potential, and ionic concentration of feed water. Enhanced mixing in spacer-filled cell was also observed to promote selective divalent ion removal. Further, higher concentrations of fixed charges on the membranes resulted in increased selectivity towards divalent ions [49]. A summary of papers with the membranes used is given in Table 2.

Research into membrane fabrication and novel membrane materials

As can be seen in Fig. 5, the research regarding new membrane materials for use in arsenic separation is scarce, while there are several publications using commercially available membranes. Research into original membrane materials tailor made for separation or arsenic ions

is hence an area that can be looked into further. Membranes with better separation efficiency, lower costs of operation, higher temperature resistance, wider operational pH range, higher current efficiency, ion exchange capacity must be looked into.

Table 3 provides a summary of the properties of commonly used commercial membranes, and Table 4 provides a summary of the most recent research work undertaken regarding fabrication of novel anion exchange membranes. As can be seen from the table, several different materials can be chosen for such an operation, each offering different properties that can be useful for electro dialytic separation operations.

Table 2. Summary of recently published work on arsenic removal using electro dialysis.

Ref.	Operational parameters	Membrane
[46]	Membrane area: 210 cm ² Voltage: 0 - 60 V pH: 2 – 12	Commercial Membranes
[47]	Membrane area: 58 cm ² Voltage: 12 & 18 V pH: 3 – 11	Commercial Membranes
[51]	Membrane area: 64 cm ² Current Density: 100 – 250 A/m ² pH: 2 – 4	Commercial Membranes
[48]	Membrane area: 100 cm ² Operating Pressure: 12 – 27 kPa pH: 6 – 8	Commercial Membranes`
[40]	Membrane area: 47 cm ² Voltage: 12 & 18 V pH: 3 – 11	Commercial Membranes
[56]	Membrane area: 55 cm ² Voltage: 10 - 30 V pH: 4 – 10	Commercial Membranes
[55]	Membrane area: 49 cm ² Voltage: 5 - 25 V pH: 3 – 10	Commercial Membranes
[54]	Membrane area: 11.3 cm ² pH: 7.8 – 8	Commercial Membranes
[53]	Membrane area: 10 cm ² Voltage: 10 - 20 V pH: 6 – 10	Commercial Membranes
[52]	Membrane area: 200 cm ² Voltage: 45 V Feed TDS: 200 ppm	Fabricated Membranes.
[49]	Membrane area: 47 cm ² Voltage: 0 – 1.4 V pH: ~7	Commercial Membranes

Table 3. Properties of commonly used commercial membranes

Property	Range
pH tolerance	0 – 14
Temperature range	< 40° C
Ion exchange capacity	2.2 mmol/g
Water uptake	30%

Further, those materials once fabricated were only tested on common salts and hence their effectiveness for a more complex operation of heavy metal removal can be looked into.

Table 4. Summary of recent research on ion-exchange membrane fabrication

Ref.	Material	Properties of membranes
[52]	Polyethylene interpolymer-based ion-exchange membranes	Energy density (W) = 1.24 kWhr/Kg Current efficiency (CE) = 74.5% Ion exchange capacity (IEC) = 1.30 mmeq/g Temperature: 25° C
[57]	Pore-filling anion exchange membranes (PAEM). Trimethylammonium chloride mixed with piperazine diacrylamide as a crosslinking agent.	IEC: 1.645 mmeq/g, Water uptake (WU) = 34.47% Tensile strength: 148 Mpa, Resistance: 0.661 Ohm/cm ² Temperature: 25° C
[58]	Photocured membrane made using N-dimethoxymethylsilylpropyl-N, N, N-trimethylammonium chloride, 50 wt% in MeOH) and 3-acrylamidopropyl trimethoxysilane	IEC: 2.2 mmol/g Temperature: 75 ° C WU: 128.18% Resistance: 0.224 Ohm/cm ²
[59]	Graphene oxide (rGO) crosslinked polysulfone-based AEM.	IEC: 33.5 mmol/g WU: 33.5% Tensile strength: 1730 MPa Temperature: 80° C
[60]	Brominated poly (2,6-dimethyl-1,4-phenylene oxide) as the polymer base and 4-methylpyridine as an ion exchanger.	Tensile strength: 33 MPa IEC: 2.2 mmol/g Temperature: 70° C
[61]	Imidazole-based anion exchange membranes created by photopolymerization.	Temperature: 100°C Tensile strength: 29.5 MPa IEC: 2.69 mmol/g, Resistance 1.78 Ohm/cm ² Limiting current density: 13.23 mA/cm ² W: 9.1 kWh/kg, CE: 81.9%
[62]	Membrane fabricated by single step crosslinking of porous brominated poly (phenylene oxide) membrane substrate with 1, 4-diazabicyclo [2.2.2] octane.	WU: 151% Temperature: 110° C IEC 1.7 mmol/g

One important factor for any fabricated membrane that decides whether it will achieve commercial viability is its stability. Maintenance is a major issue in electrodialysis processes. Hence, thermal and chemical stability of membranes are vital parameters for a membrane, which are assessed by studying the weight loss percentage of the membrane when subjected to thermal and chemical stress. However, not all publications have reported the stability of the membranes fabricated in their research. Bai *et al.* reported a 55% weight percentage over a course of 72-hour experimentation

for oxidative stability tests and for thermal stability tests, a weight percentage of 80% at 200°C and 60% at 800°C [59]. Yang *et al.* in their thermal stability tests recorded a weight percentage ranging from 70–80% at 300°C and 10–20% at 700°C, depending on the membrane [58]. Thermal stability tests by Khan *et al.* showed a weight percentage ranging from 90–95% at 200°C and 10–45% at 600°C for the various membranes fabricated in the research [60]. Lin *et al.* recorded an impressive near 100% weight percentage at 300°C and nearly 50% at 500°C [62].

These are consistent with commercially used membranes and even surpass them in some cases.

Additionally, depending on the selectivity of the membrane being used, information regarding speciation of arsenic as described earlier in Fig 1 can be used to control the number of monovalent and divalent arsenic ions in the solution.

CONCLUSIONS

The paper briefly summarized the sources of arsenic, the effects – both short- and long-term of exposure on humans, the strategies in place for arsenic removal from water, as well as the place of electrodialysis among those processes. Further, the paper reviewed and summarised the research regarding separation of arsenic from wastewater using electrodialysis undertaken over the past decade.

The majority of work has been carried out on commercial membranes, hence a need for research into newer membrane materials is crucial for the commercial viability of the process for wastewater treatment. Membranes with better separation efficiency, lower costs of operation, higher temperature resistance, wider operational pH range, higher current efficiency, ion exchange capacity can be researched and developed.

Further, a summary of recent research on fabricated ion exchange membranes, as well as their properties was given. Based on the information, membranes can be tailor-made for the specific purposes of arsenic separation in particular or heavy metals in general, with properties superior to the ones found in commercially available membranes.

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