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## REVIEW OF MEMBRANE TECHNOLOGIES IN THE REMOVAL OF HEAVY METALS IN SURFACE WATERS FOR HUMAN CONSUMPTION

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Several sources contaminate heavy metals in surface waters. Among the main ones are industries and mining, in addition to natural geological pollution. Several international organizations consider heavy metals as toxic and dangerous.

Globally, many bodies of surface water have concentrations of metal ions that exceed the allowed limits. This problem is more significant when water bodies contaminated with metals are used as sources of supply for cities. Therefore, there is an imminent need to apply efficient treatment technologies and with a high metal removal rate. This work makes a bibliographic review of the results obtained in the various experiences of the last 20 years, to identify which of the membrane filtration processes is the most promising for its application, considering different metal ions.

*Keywords: metal; ions; membrane; filtration*

## REVISIÓN DE TECNOLOGÍAS DE MEMBRANAS EN LA ELIMINACIÓN DE METALES PESADOS EN AGUAS SUPERFICIALES PARA CONSUMO HUMANO

Existen diversas fuentes que contaminan con metales pesados en las aguas superficiales. Entre las principales se encuentran las industrias y la minería, además de la contaminación geológica natural. Los metales pesados son considerados por varias organizaciones internacionales como tóxicos y/o peligrosos.

A nivel mundial, muchos cuerpos de agua superficial presentan concentraciones de iones metálicos que exceden los límites permitidos. Esta problemática es mayor cuando los cuerpos de agua contaminados con metales se utilizan como fuentes de abastecimiento para ciudades. Observando lo anterior, existe la necesidad inminente de aplicar tecnologías de tratamiento eficientes y con alta tasa de remoción de metales. Este trabajo hace una revisión bibliográfica de los resultados obtenidos en las diversas experiencias de los últimos 20 años, con el fin de identificar cuál de los procesos de filtración por membranas es el más prometedor para su aplicación, considerando diferentes iones metálicos.

*Palabras clave: iones; metálicos; filtración; membrana*

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## 1. Introduction

The term heavy metal applies to the group of metals and metalloids with an atomic density greater than  $4000 \text{ kg}\cdot\text{m}^{-3}$ , which are natural components of the Earth's crust. Although some act as essential micronutrients for living things, higher concentrations can cause serious toxicity (Srivastava & Majumder, 2008). Owing to their mobility in aquatic ecosystems and their toxicity to higher life forms, heavy metals in surface and groundwater supplies have been prioritized as major inorganic contaminants in the environment (Thayer & Brinckman, 1982). In particular, heavy metals are considered especially dangerous contaminants in freshwater reserves (Alvizuri et al., 2019).

It is known that the origin of the contamination of water bodies by heavy metals is usually geogenic (as is the case of As) or domestic and industrial discharge (Zn, As, Pb, Cd, Ni, Cu, or Cr). However, the greatest anthropogenic sources of heavy metal contamination (As, Pb, Zn, Cu, Cd, Mn, or Hg) are active and abandoned mining areas (Chowdhury et al., 2016). In mining areas, rainwater leaches metals, thereby contaminating surface water bodies. The metals are transported by water currents and are either dissolved or adsorbed by sediments. These can also accumulate in sediments and pass into groundwater (Duruibe, Egwurugwu, & Ogwuegbu 2007).

The widespread concern about removing heavy metals from water has led to the publication of many articles and reports on heavy metal removal methods. In terms of the efficiency of metal removal, ion exchange and membrane filtration (MF) have the best results (Fu & Wang 2011). However, the high separation selectivity ratio, in addition to other advantages such as easy operation and saving of time and space, makes MF technology the most promising method for the removal of metal contaminants from water (Alvizuri et al., 2019).

This study analyzed the results of different MF processes in order to identify the potential of ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) membranes in the remission of different metal ions. In addition, some popular membrane manufacturers in the industry are presented. Finally, opportunities that should be addressed in future research are identified.

## 2. Methodology

This study conducted a bibliographic review of heavy metal remission studies through MF technologies and analyzing procedures of UF, NF, and RO. A systematic review of bibliographic databases (SCOPUS, Web of Science, Google Scholar, and PubMed/MEDLINE) was conducted. For the review, only articles with an impact factor in the Journal Citation Reports of greater than 2 were considered. The review begins with the definition and main characteristics of membrane processes, followed by a discussion and comparison of the yields achieved by the different membrane processes in water purification.

## 3. Synthesis and discussion

MF is a process of pressure separation (Zhao et al., 2016). This technology is widely accepted as a means of producing various qualities of water from surface water, well water, brackish water, and seawater. The choice of a suitable membrane process depends on the size of the contaminants and admixtures removed from the water (Bodzek, Konieczny, & Kwiecińska, 2011). This technology has advantages and disadvantages for its application.

The advantages include high efficiency, high separation selectivity, space and time saving, and easy operation. The disadvantages include the requirement of pretreatment to ensure adequate function, high operating costs, and high energy consumption (Fu & Wang, 2011).

The membrane processes used to remove metals from wastewater are UF, NF, and RO (Fu & Wang, 2011). The descriptions and main features of the processes involved in each method are provided below.

- Ultrafiltration

On the basis of pore size (in the range 2–100 nm) and the molecular weight of the separating compounds (1000–100 000 Da), UF, which is an energy-intensive process, works at low transmembrane pressures for the removal of dissolved and colloidal materials (Zhao et al., 2016). Particles that are larger than the pore size of UF membranes are trapped, while dissolved metal ions (in the form of hydrated ions or low-molecular-weight complexes) pass through easily (Vijayalakshmi et al., 2008).

To obtain a high removal efficiency of metal ions, micellar-enhanced ultrafiltration (MEUF) and polymer-enhanced ultrafiltration (PEUF) processes have been proposed (Fu & Wang, 2011). In the MEUF process, metal ions are captured within surfactant micelles and form large metal-surfactant structures (Liu et al., 2016). In the PEUF process, water-soluble polymers form complexes with metal ions and form macromolecules with higher molecular weights, which are then retained when pumped through a UF membrane (Landaburu-Aguirre et al., 2009).

- Nanofiltration

NF is the intermediate process between UF and RO (Fu & Wang, 2011). In this process, the molecular weights of the separating compounds range between 200 Da and 1000 Da with pore diameters varying from 1 nm to 2 nm (Khedr, 2008). The application of NF processes for acid mine drainage treatment has recently attracted attention owing to its high capacity for salt and metal retention (Kefeni, Msagati, & Mamba, 2017).

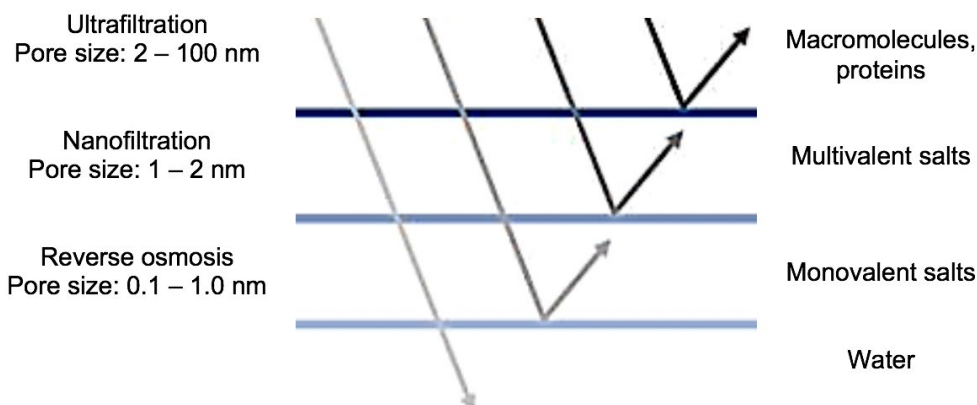
- Reverse osmosis

RO, which involves membranes with a pore size of <1 nm, works on the principle of size exclusion and solution diffusion through a semipermeable membrane (Greenlee et al., 2009). A significant advantage of RO over other traditional water treatment technologies is the ability to reduce the concentrations of other ionic contaminants as well as dissolved organic compounds (Fu & Wang, 2011). Within membrane technologies, RO is the most selective technology because by selecting the appropriate membrane, rejections close to 100% can be obtained for most metal cations (Akpór & Muchie, 2010; Fu & Wang, 2011).

The significant improvements in the technology and design of RO have made it an eco-friendly process that treats water in many regions worldwide, particularly in those where its sources are limited. It has also been seen that other advances, such as the availability of alternative energy sources, the possibility of pretreatment, and applied materials, have helped position RO among the most promising methods in terms of effluent treatment (Meschke et al., 2019).

A description of the membrane processes with the corresponding pore diameter and the retained species is shown in Figure 1.

**Figure 1: Membrane processes**



Source: based on Yang et al., 2019.

### 3.1. Applicability and case studies

MF technology is one of the most commonly used methods for the removal of emerging pollutants from water (Kim et al., 2018), including heavy metals (Fu & Wang 2011). A summary of some cases where MF processes (UF, NF, and RO) were applied for the removal of different heavy metals and the results are given in Tables 1, 2, 3, 4, 5, 6, and 7.

**Table 1: Efficiency of membrane treatments in copper removal**

Heavy metal	Type of technology	Removal efficiency (%)	Reference
Cu <sup>2+</sup>	NF	47.0–66.0	Chaabane et al., 2006
Cu <sup>2+</sup>	NF	96.0–98.0	Tanninen, Mänttari, & Nyström, 2006
Cu <sup>2+</sup>	RO	99.5	Mohsen-Nia, Montazeri, & Modarress, 2007
Cu <sup>2+</sup>	PEUF	94.0	Molinari, Poerio, & Argurio 2008
Cu <sup>2+</sup>	RO+NF	95.0–99.0	Sudilovskiy, Kagramanov, & Kolesnikov, 2008
Cu <sup>2+</sup>	RO	70.0–95.0	Zhang et al., 2009
Cu <sup>2+</sup>	RO+NF	>95.0	Cséfalvay, Pauer, & Mizsey, 2009
Cu <sup>2+</sup>	NF	97.6	Barakat, & Schmidt, 2010
Cu <sup>2+</sup>	PEUF	99.5	Camarilloa et al., 2010

Note: NF: nanofiltration; RO: reverse osmosis; MEUF: micellar-enhanced ultrafiltration; PEUF: polymer-enhanced ultrafiltration.

**Table 2: Efficiency of membrane treatments in arsenic removal**

Heavy metal	Type of technology	Removal efficiency (%)	Reference
As(VII)	MEUF	19	Ferella et al., 2007
As(V)	RO	91–99	Chan, & Dudeney, 2008
As(V)	RO	>90	Yoon et al., 2009
As(V)	NF	>90	Figoli et al., 2010
As(III)	RO	20–55	Chan, & Dudeney, 2008
As(III)	RO	98	Chang, Liu, & Wang, 2014

Note: NF: nanofiltration; RO: reverse osmosis; MEUF: micellar-enhanced ultrafiltration.

**Table 3: Efficiency of membrane treatments in lead removal**

Heavy metal	Type of technology	Removal efficiency (%)	Reference
Pb <sup>2+</sup>	MEUF	>99.0	Ferella et al., 2007
Pb <sup>2+</sup>	NF	93.0	Zhu et al., 2014
Pb <sup>2+</sup>	RO	98.9	Vera et al., 2018
Pb <sup>2+</sup>	NF	98.7	Vera et al., 2018

Note: NF: nanofiltration; RO: reverse osmosis; MEUF: micellar-enhanced ultrafiltration.

**Table 4: Efficiency of membrane treatments in nickel removal**

Heavy metal	Type of technology	Removal efficiency (%)	Reference
Ni <sup>2+</sup>	RO	99.3	Ipek, 2005
Ni <sup>2+</sup>	RO	99.5	Mohsen-Nia, Montazeri, & Modarress, 2007
Ni <sup>2+</sup>	MEUF	98.6	Danisa, & Aydiner, 2009
Ni <sup>2+</sup>	PEUF	99.1	Barakat, & Schmidt, 2010

Note: RO: reverse osmosis; MEUF: micellar-enhanced ultrafiltration; PEUF: polymer-enhanced ultrafiltration.

**Table 5: Efficiency of membrane treatments in cadmium removal**

Heavy metal	Type of technology	Removal efficiency (%)	Reference
Cd <sup>2+</sup>	MEUF	92.0–98.0	Huang et al., 2010
Cd <sup>2+</sup>	NF	95.0	Zhu et al., 2014
Cd <sup>2+</sup>	PEUF	99.0	Ennigrou et al., 2009
Cd <sup>2+</sup>	MEUF	99.0	Landaburu-Aguirre et al., 2010
Cd <sup>2+</sup>	NF	98.1	Vera et al., 2018
Cd <sup>2+</sup>	RO	99.3	Vera et al., 2018

Note: NF: nanofiltration; RO: reverse osmosis; MEUF: micellar-enhanced ultrafiltration; PEUF: polymer-enhanced ultrafiltration.

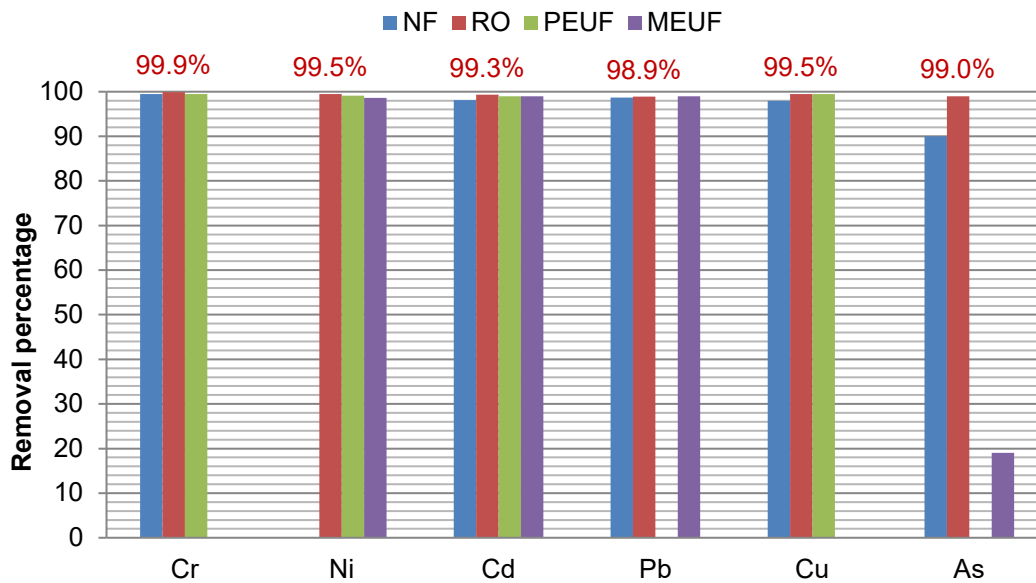
**Table 6: Efficiency of membrane treatments in chrome removal**

Heavy metal	Type of technology	Removal efficiency (%)	Reference
Cr(VI)	RO	99.9	Ozaki, Sharma, & Saktaywin, 2002
Cr(VI)	NF	99.5	Muthukrishnan, & Guha, 2008
Cr(VI)	PEUF	82.0–100.0	Korus, & Loska, 2009
Cr(VI)	RO	>90.0	Yoon et al., 2009
Cr(VI)	RO	96.0	Cimen, Kilicel, & Arslan 2014
Cr(III)	PEUF	82.0–100.0	Korus, & Loska, 2009
Cr(III)	PEUF	99.5	Barakat, & Schmidt, 2010

Note: NF: nanofiltration; RO: reverse osmosis; MEUF: micellar-enhanced ultrafiltration; PEUF: polymer-enhanced ultrafiltration.

Summarizing the information in Tables 1, 2, 3, 4, 5, and 6, Figure 2 denotes that the highest rates of heavy metal removal were obtained by RO. This demonstrates that RO is an optimal technique applicable to the treatment of water contaminated with these metal ions.

**Figure 2: Heavy metal removal rates in different membrane processes**



Note: NF: nanofiltration; RO: reverse osmosis; MEUF: micellar-enhanced ultrafiltration; PEUF: polymer-enhanced ultrafiltration.

There are few studies for other metals such as Zn, Fe, and Mn. Despite this, the removal rates achieved for the metal ions were high and can be improved if further studies are conducted.

**Table 7: Efficiency of membrane treatments in zinc, manganese, and iron removal**

Heavy metal	Type of technology	Removal efficiency (%)	Reference
Zn <sup>2+</sup>	RO	98.9	Ipek, 2005
Zn <sup>2+</sup>	MEUF	92.0–98.0	Huang et al., 2010
Zn <sup>2+</sup>	MEUF	99.0	Landaburu-Aguirre et al., 2010
Mn	NF	96.0	Soares et al., 2005
Mn	UF-RO	95.8	Nasir, Ibrahim, & Arief, 2016
Fe	UF-RO	94.1	Nasir, Ibrahim, & Arief, 2016

Note: NF: nanofiltration; RO: reverse osmosis; MEUF: micellar-enhanced ultrafiltration.

### 3.2. Membrane manufacturers

It is well known that polymeric membranes are currently used more in the seawater desalination and wastewater treatment industries owing to their well-developed and outstanding performance (Bassyouni et al., 2019). Research is still available to solve problems related to performance limitations and the post-treatment process. Fouling is one of the main disadvantages of polymeric membranes. The surface structure and materials have been

modified to suppress the embedding effect. Table 8 provides a list of membranes with their popular manufacturers in the market.

**Table 8: Membrane manufacturers**

Membrane	Manufacturer	Maximum temperature (°C)	pH range	Salt rejection (%)
SW30HRLE-400	Dow Filmtec, USA	45	2–11	99.8 NaCl
NF270-400/34i	Dow Filmtec, USA	45	3–10	>97.0 NaCl
SWC4+	Hydranautics, USA	45	3–10	>99.7 NaCl
TM820C-370	Toray, USA	45	2–11	>99.5 NaCl
HB10255	Toyobo, Japan	40	3–8	>99.4 NaCl
TS80	Microdyn-Nadir, USA	45	1–12	80.0 NaCl >98.5 MgSO <sub>4</sub>
AD-90	GE-Osmonics, USA	50	4–11	>99.5 NaCl 95.0 B
AG4040C	GE-Osmonics, USA	50	4–11	>99.0 NaCl
8040-SW-400-34	Koch, USA	45	4–11	>99.5 NaCl
4040-HR	Koch, USA	45	4–11	>99.2 NaCl
NFX	Synder, USA	50	2–11	40.0 NaCl >99.0 MgSO <sub>4</sub>
NFW	Synder, USA	50	2–11	20.0 NaCl >97.0 MgSO <sub>4</sub>

Source: Yang et al., 2019.

In recent years, hybrid processes have appeared, such as combined bioreactors with MF technologies that have optimized heavy metal removal (Malamis et al., 2012; Mulyati & Syawaliah, 2018). However, these hybrid models are still poorly studied owing to aspects such as high economic costs and energy costs that directly affect their efficiency.

#### 4. Conclusions

The RO technique had the highest values of removal efficiency of heavy metals (from 98.0% to 99.9%). RO is highly efficient, but as observed in many studies, it requires a large amount of energy for its operation. It is important to maintain high efficiency in the RO process, but work must also be done to determine moderate critical conditions that can ensure the sustainability of the process. Determining the best-operating conditions for RO will also help



hybrid systems where RO is applied, which would eliminate the limitations of these new systems.

Membrane manufacturers are launching new products on the market with different characteristics and operating specifications. These peculiarities must be studied to make an adequate selection of membranes and guarantee their implementation in water treatment processes at an industrial level.

Further research should study the integration of membranes with bioreactors for the treatment of wastewater. Initially, the hybrid models have presented attractive features that conventional treatment processes do not have.

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**Communication aligned with the Sustainable Development Objectives**

