



## New trends of heavy elements removal in wastewater: A comprehensive review on inorganic membrane technology

Neamatalla M. Azzam <sup>1,2</sup>, Sahar S. Ali <sup>3</sup>, Gehad G. Mohamed <sup>1,4</sup>, Mohamed M. Omar <sup>1</sup>,  
Shereen K. Amin <sup>3\*</sup>



<sup>1</sup>Chemistry Department, Faculty of Science, Cairo University, Giza, 12613, Egypt.

<sup>2</sup>Quality Department, Egyptian Organization for Standardization and Quality (EOS), Cairo, 11813, Egypt.

<sup>3</sup>Chemical Engineering and Pilot Plant Department, Engineering & Renewable Energy Research Institute, National Research Centre (NRC), Affiliation ID: 60014618, Giza, 12622, Egypt.

<sup>4</sup>Nanoscience Department, Basic and Applied Sciences Institute, Egypt-Japan University of Science and Technology, New Borg El Arab, Alexandria, 21934, Egypt.

### Abstract

All life on earth, even non-living things, depends on water. It is well known that the availability of clean water is depleting daily and that the fast development of industry and technology has increased the amount of hazardous wastewater released into the environment. Before being released into the environment, wastewater from industries, farms, and communities shall be treated since it may contain dangerous contaminants such as heavy metal ions, inorganic compounds, organic dyes, and pharmaceutical waste. Heavy metals contamination in water resources significantly threatens human health and the environment. Several processes have been developed to eliminate heavy metals from water, among which membrane-based technologies have gained considerable attention due to their effectiveness, selectivity, and scalability. This review paper focuses on the application of inorganic membrane technology for the removal of heavy elements from wastewater. It provides a comprehensive overview of diverse categories of inorganic membranes utilized for heavy metal removal from water, highlighting their principles of operation, advantages, limitations, and recent advancements.

**Keywords:** Wastewater; Heavy metals; Water resources; Membrane-based technology; Inorganic membranes.

### 1. Introduction:

Water is a precious natural resource that is abundant around the world. It is necessary for all living things, including plants and animals, to survive [1]. Even though water surrounds 70% of the earth, but just 2.5 percent is drinkable, with the other percent being saline. Approximately 1% of this fresh portion is available overall, providing for roughly 7.6 billion persons. Regrettably, industrial operations, wastewater treatment plants, and agricultural residues from pesticides and fertilizers pollute a portion of this water [2-4].

Worldwide biodiversity and humankind are seriously threatened by contamination of the water. The release of industrial effluents into the environment affects the quality of the water, atmosphere, and soil. The most prevalent pollutants thrown into waterways such as water bodies, and streams include runoff from municipal, domestic, manufacturing, and agricultural residue, as well as sewage from wastewater treatment plants and rainfall [5]. The textiles, petrochemical manufacturing, medicinal products, refinery, and mineral extraction industries all directly release toxic substances into water bodies, which pollutes the water and has negative effects on aquatic life, plants, animals, and people. Wastewater contains an extensive number of microbes that are pathogenic and xenobiotic compounds that are directly disposed of, even though minimal physiochemical changes in contaminated water can have a serious negative impact on aquatic life and human health [6]. Every day, a massive amount of metal is thrown into the surroundings as wastewater, industrial effluent, and solid waste. Exhaust gas from automobiles, metal smelting factories such as copper and zinc, pesticide and insecticide industries that manufacture chemicals, the combustion of fossil fuels, paint industries, household product industries, cosmetic industries, and so on are all human-induced sources of heavy metals in the surrounding environment [7]. Pollutants from these sources are eventually released into the environment and either wash into water bodies or are released as wastewater. Numerous metals with economic significance and practical use in daily life can be found in the wastewater produced by homes and businesses. These metals can be classified as highly hazardous elements (Hg, Pb, Cd, Ag, Au, Pd, Bi, As, Pt, Se, Sn, Zn), macronutrients (Co, Fe), and micronutrients (Cu, Ni, Cr, Fe, Mn, Mo) [8]. Depending on the intrinsic characteristics, they may be beneficial

\*Corresponding author e-mail: [dr.shereenkamel@hotmail.com](mailto:dr.shereenkamel@hotmail.com); [sheren51078@yahoo.com](mailto:sheren51078@yahoo.com); (Shereen Kamel Amin).

Receive Date: 16 September 2024, Revise Date: 12 November 2024, Accept Date: 24 November 2024

DOI: <https://doi.org/10.21608/ejchem.2024.321481.10441>

©2025 National Information and Documentation Center (NIDOC)

nutrients or cause chronic toxicity in living organisms due to their capacity to accumulate in biomass [9]. Heavy metals accumulate in organisms as one progresses up the food pyramid; hence, species at the top are more prone to face toxic effects than those at the bottom [10]. Humans are the ecosystem's top predators; thus, they are exposed to high levels of heavy metals through their diet. Metals are well recognized to attack important organs such as the kidneys, lungs, brain, liver, and skin, causing a variety of diseases such as organ failure, neuropathies, cancer, ulcers, and so on [11, 12].

As a result, various government organizations place a strong premium on remediation and preventing further poisoning of the environment with these harmful metals. Water purification and wastewater treatment are two critical processes for improving water quality and removing the great majority of pollutants from wastewater and therefore, for alleviating water scarcity. The main scope of this review is to critically examine the current state and future prospects of inorganic membrane technology in the removal of heavy elements from wastewater, with a particular focus on its efficiency, challenges, and potential for addressing the growing global water contamination crisis. The results demonstrate that inorganic membrane technology offers a promising approach to addressing heavy metal pollution in water, aligning with sustainable practices within environmental engineering.

## 2. Heavy Metals (HMs):

The term "heavy metals" refers to metallic elements having a relatively high density. HMs can cause toxicity even at low exposure levels [13]. The atomic weight range for heavy metals is 63.5-200.6 Da, and their density is larger than 5 g/cm<sup>3</sup> [14-15]. Contrary to organic contaminants, which degrade over time, heavy metals do not, which means that after being released into the environment, they tend to accumulate in living organisms. When treating industrial wastewater, the following harmful heavy metals are primarily considered Fe, Cr, Cd, Hg, As, Ni, Cu, and Zn. Numerous studies have been conducted on their detrimental effects on both human and animal health. A reduction in memory and attention, hypertension, cardiovascular blockage, speech impairment, tiredness, sleep problems, altered behavior (aggression, irritability, mood swings, sadness), increased allergic reactions, and autoimmune illnesses are just a few of the symptoms [16]. Typically exchange and coordination are the mechanisms that cause poisoning. When heavy metals are ingested into the body, they mix with the proteins and enzymes already there to create stable bio-toxic chemicals [17]. Heavy metals like Mg, Fe, Cu, and Zn are necessary for the body, but their presence in large amounts is harmful [18-19].

### 2.1. Sources of heavy metal pollution: [20-24]

Heavy metal pollution in water resources stems from various anthropogenic activities. The primary sources include:

- 1) **Industrial activities:** Industries such as mining, smelting, and manufacturing are major contributors to heavy metal pollution. The metal extraction processes can release significant quantities of lead, cadmium, and mercury into the environment, contaminating nearby soil and water sources.
- 2) **Agricultural practices:** The use of fertilizers and pesticides can lead to the accumulation of heavy metals in agricultural soils. For example, phosphate fertilizers are often a source of cadmium contamination, which can subsequently enter the food chain.
- 3) **Urban runoff:** Urban areas contribute to heavy metal pollution through stormwater runoff, which can carry metals from road surfaces, buildings, and industrial sites. The runoff in urbanized watersheds often contains elevated levels of zinc, copper, and lead.
- 4) **Waste disposal:** Improper disposal of electronic waste and other materials can lead to significant heavy metal contamination. The e-waste recycling processes release harmful metals such as lead and mercury into the environment.
- 5) **Natural sources:** While anthropogenic sources are predominant, natural processes such as volcanic eruptions and weathering of mineral deposits also contribute to the presence of heavy metals in the environment. Natural weathering can release metals like arsenic and chromium into surrounding ecosystems.

### 2.2. Dangers of heavy metal pollution to human health: [25-26]

Heavy metal pollution poses severe risks to human health due to their persistence in the environment and tendency to bioaccumulate. Some of the primary health effects include:

- 1) **Neurological disorders:** Lead and mercury can cause cognitive impairment, developmental delays in children, and neurodegenerative diseases.
- 2) **Kidney damage:** Cadmium and lead are known to cause renal dysfunction and chronic kidney disease.
- 3) **Cardiovascular problems:** Arsenic exposure is linked to increased risk of hypertension and cardiovascular diseases.
- 4) **Respiratory issues:** Inhalation of chromium and nickel can lead to respiratory tract cancers and other lung disorders.
- 5) **Reproductive problems:** Many heavy metals, including lead and mercury, can cause reproductive issues and birth defects.
- 6) **Cancer:** Several heavy metals, such as arsenic, chromium, and cadmium, are classified as carcinogens.

Addressing heavy metal pollution is critical to protecting human health, necessitating stringent regulations and public awareness initiatives to mitigate exposure risks.

### 2.3. Toxicity and regulatory limits:

Heavy metals, including lead (Pb), cadmium (Cd), mercury (Hg), and arsenic (As), are toxic elements commonly found in industrial wastewater and pose significant risks to human health and the environment. Understanding their toxicity and regulatory standards is crucial for effective remediation strategies. The toxicity of some heavy metals and their regulatory limits are shown in Table (1) [27-28]. The toxicity of heavy metals like lead, cadmium, mercury, and arsenic highlights the

critical need for effective removal technologies. Regulatory standards established by the EPA serve as benchmarks for safeguarding public health and preventing environmental degradation from contaminated wastewater.

**Table (1): The toxicity of some heavy metals and their regulatory limits**

Metal	Toxicity	Maximum Contaminant Level (MCL)	Standards for Wastewater Disposal
<b>Lead (Pb)</b>	Lead is a potent neurotoxin, particularly harmful to children, affecting cognitive development and leading to behavioral issues. Chronic exposure can cause hypertension, kidney damage, and reproductive problems in adults.	The U.S. Environmental Protection Agency (EPA) sets the MCL for lead in drinking water at 0.015 mg/L (15 µg/L).	The National Pollutant Discharge Elimination System (NPDES) establishes a limit of 0.1 mg/L (100 µg/L) for lead in industrial wastewater discharges.
<b>Cadmium (Cd)</b>	Cadmium is highly toxic and is known to cause kidney damage, bone fragility, and has carcinogenic effects. It can also disrupt cellular metabolism and affect the endocrine system.	The EPA has set the MCL for cadmium in drinking water at 0.005 mg/L (5 µg/L).	For cadmium, the allowable limit for wastewater discharge is typically 0.01 mg/L (10 µg/L), according to federal standards.
<b>Mercury (Hg)</b>	Mercury is a potent neurotoxin that can lead to neurological and developmental damage. It affects the brain, kidneys, and immune system. Methylmercury, an organic form, bioaccumulates in fish, posing risks to wildlife and humans.	The MCL for mercury in drinking water is set at 0.002 mg/L (2 µg/L) by the EPA.	The limit for mercury in wastewater discharges is 0.05 mg/L (50 µg/L).
<b>Arsenic (As)</b>	Arsenic is a carcinogen linked to skin, bladder, and lung cancers. It can cause various health issues, including skin lesions, developmental effects, and cardiovascular disease.	The EPA's MCL for arsenic in drinking water is 0.010 mg/L (10 µg/L).	In the context of wastewater treatment, the limit for arsenic generally ranges from 0.05 mg/L (50 µg/L) for certain industrial discharges.

#### 2.4. Factors affecting the removal of heavy metals: [29-32]

The efficiency of heavy metal removal from wastewater is influenced by several factors:

- 1) **pH:** The acidity or alkalinity of the solution affects the solubility and speciation of heavy metals, impacting their removal efficiency.
- 2) **Temperature:** Higher temperatures generally increase the rate of adsorption and ion exchange processes.
- 3) **Initial metal concentration:** The initial concentration of heavy metals in the wastewater affects the removal efficiency and the choice of treatment method.
- 4) **Contact time:** Longer contact times between the wastewater and the treatment medium often result in higher removal efficiencies.
- 5) **Presence of other ions:** Competing ions in the wastewater can interfere with the removal of target heavy metals.
- 6) **Particle size of adsorbents:** Smaller particle sizes generally provide larger surface areas for adsorption, improving removal efficiency.
- 7) **Flow rate:** In continuous flow systems, the flow rate affects the contact time and thus the removal efficiency.

### 3. Strategies for Remediating Toxic Metals:

Technical improvements have made it achievable to remove heavy metals using a combination of physical, chemical, and biological approaches, or by combining more than one treatment method.

#### 3.1. Techniques for eliminating toxic heavy metals: [33-38]

The removal of heavy metals from contaminated environments is crucial for protecting human health and ecosystems. Various techniques have been developed, each with distinct mechanisms and efficiencies. This overview discusses the primary methods: physical, chemical, and biological.

##### 3.1.1. Physical methods:

- a) **Adsorption:** This technique involves the adhesion of metal ions onto the surface of solid adsorbents, such as activated carbon, clay minerals, and zeolites. The activated carbon can effectively remove heavy metals like lead and cadmium from aqueous solutions, achieving removal efficiencies greater than 90%, adsorption mechanism is shown in Figure (1) [39].

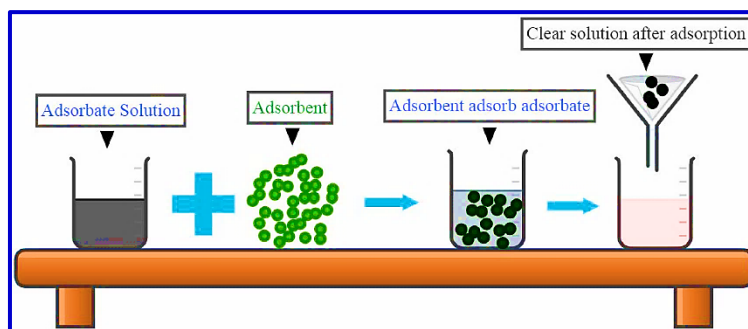


Fig. (1): Adsorption mechanism [39]

- b) **Membrane filtration:** This method uses selective barriers to separate metals from solutions. Techniques such as microfiltration, ultrafiltration, and reverse osmosis can effectively remove dissolved metals from wastewater. The membrane bioreactors can achieve high removal rates for metals and pathogens, making them suitable for treating industrial effluents, membrane filtration process is shown at Figure (2) [40].
- c) **Electrokinetic remediation:** This technique employs an electric field to mobilize heavy metals within soils or sediments, facilitating their extraction. The effectiveness of electrokinetic processes in remediating contaminated sites by enhancing the transport of heavy metals toward collection electrodes.

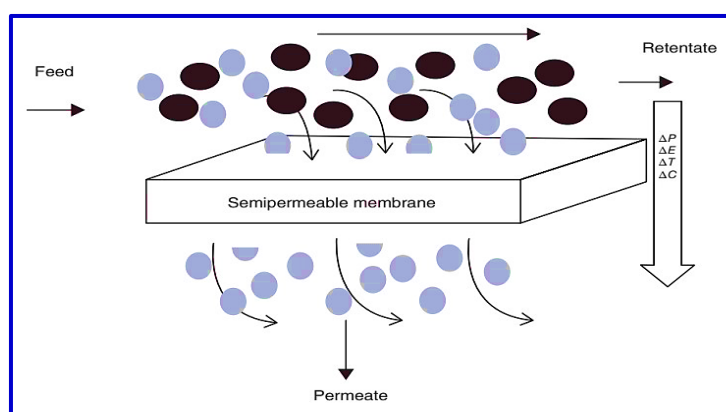


Fig. (2): Membrane filtration process [40]

### 3.1.2. Chemical methods:

- a) **Chemical precipitation:** This widely used technique involves adding reagents that react with dissolved metals to form insoluble compounds, which can then be filtered out.
- b) **Oxidation-reduction reactions:** Chemical oxidation and reduction processes can convert toxic metal species into less harmful forms. For instance, the use of oxidizing agents like hydrogen peroxide can effectively oxidize certain metal ions, enhancing their removal.
- c) **Complexation:** This method involves adding ligands that bind to metal ions, forming stable complexes that can be removed from the solution. Researches indicated that using biodegradable chelating agents can significantly enhance the removal of heavy metals from contaminated water.

### 3.1.3. Biological methods:

- a) **Bioremediation:** This approach utilizes microorganisms to degrade or accumulate heavy metals. Certain bacteria and fungi can bioaccumulate metals, effectively removing them from contaminated environments.
- b) **Phytoremediation:** This technique employs plants to absorb, accumulate, and detoxify heavy metals from soil and water. Plants such as mustard greens and sunflowers have shown potential for heavy metal uptake.
- c) **Mycoremediation:** This is a subset of bioremediation that uses fungi to degrade or sequester heavy metals. Fungi have unique metabolic pathways that can transform toxic metals into less harmful forms.

### 3.1.4. Others:

- a) **Ion exchange techniques:** Utilize ion exchange resins that selectively attract and bind heavy metal ions from wastewater. For example, when wastewater containing cadmium ions is passed through a column filled with a cation exchange resin, cadmium ions are exchanged for sodium ions, effectively removing cadmium from the solution, ion exchange technique is shown in Figure (3) [41].

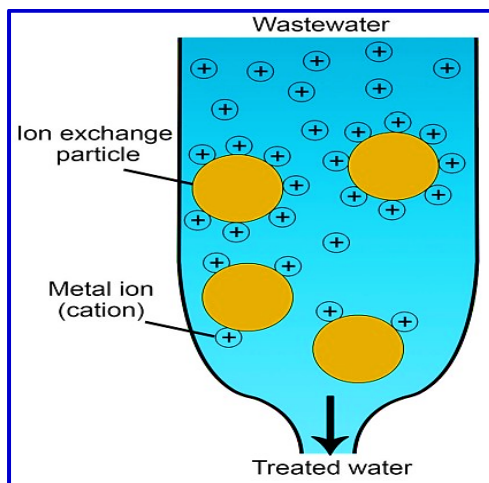


Fig. (3): Ion exchange [41]

- b) **Electrochemical treatment:** In electrochemical treatment, an electric current is passed through contaminated water, promoting the electrochemical reactions that lead to the precipitation of heavy metals at the electrodes. For example, electrocoagulation can be applied to treat wastewater containing arsenic. The application of an electric current results in the oxidation of aluminum anodes, forming aluminum hydroxide, which can scavenge arsenic ions, facilitating their removal from the water.

Due to their remarkable selectivity, minimal space requirements, and high removal effectiveness membrane-based approaches have recently gained popularity in removing heavy metals from water sources [42]. In recent years, nanoparticles have been used to make efficient membranes for filtering contaminants [19]. Table (2) outlines the benefits and drawbacks of the various technologies employed for toxic metal rejection from wastewater [43-45].

The selection of an appropriate metal removal method depends on various factors, including the type of metal, concentration, and the specific environmental context. Combining these techniques can also enhance overall efficiency, leading to more effective remediation strategies.

### 3.2. Efficiency of different removal techniques: [32-46]

In recent years, the efficiency of different removal techniques has gained significant attention in environmental management and industrial applications. Studies have highlighted a range of methods, including chemical, biological, and physical techniques, each with unique advantages and limitations. For instance, the advanced oxidation processes such as photocatalysis can achieve removal efficiencies exceeding 90% for certain contaminants, making them highly effective for wastewater treatment. Conversely, bioremediation techniques demonstrate the potential for sustainable and cost-effective removal of heavy metals, although their efficiency can be influenced by environmental conditions. Additionally, physical methods like adsorption have shown versatility and rapid removal rates, particularly when utilizing novel materials such as activated carbon and biochar. Understanding these varied approaches allows for the optimization of removal strategies tailored to specific contaminants and contexts.

Various techniques have been developed for heavy metal removal from wastewater, each with different efficiencies:

- a) **Chemical precipitation:** Can achieve 90-99% removal efficiency for many metals but may produce large volumes of sludge [47].
- b) **Ion exchange:** Offers 90-99% removal for most metals with the advantage of metal recovery [48].
- c) **Adsorption:** Activated carbon can achieve 80-99% removal efficiency, while bio-adsorbents show variable efficiencies from 50-99% depending on the metal and biosorbent used [49].
- d) **Membrane filtration:**
  - Reverse Osmosis (RO): >99% removal efficiency for most heavy metals [50].
  - Nanofiltration (NF): 95-99% removal efficiency [51].
  - Ultrafiltration (UF): 90-99% when combined with complexation or other pretreatment methods [52].

**Table (2): Benefits and drawbacks of various methods of separating metals from wastewater [43-45]**

Approach	Benefits	Drawbacks
Chemical precipitation	<ul style="list-style-type: none"> <li>Low pH levels for operation and effective in removing copper.</li> <li>Minimal time spent in custody.</li> <li>Improved filtering properties are exhibited by carbonate sludge.</li> </ul>	<ul style="list-style-type: none"> <li>The process requires more than stoichiometric amounts of reagent.</li> <li>Significant amount of sludge production.</li> <li>Inefficient when the level of ionic substances is low.</li> </ul>
Ion exchange	<ul style="list-style-type: none"> <li>Minimal expenses.</li> <li>Not much energy is needed.</li> <li>The cost of regenerating chemicals is low.</li> </ul>	<ul style="list-style-type: none"> <li>Adsorption of organic materials.</li> <li>Contamination by bacteria.</li> <li>The resin may have contaminated organic matter.</li> </ul>
Ion flotation	<ul style="list-style-type: none"> <li>Promotes its wider application for industrial use.</li> <li>A high level of selectivity without interference from complexing agents.</li> </ul>	<ul style="list-style-type: none"> <li>An extremely high initial capital expense.</li> <li>Operating costs include energy.</li> </ul>
Phyto-extraction	<ul style="list-style-type: none"> <li>Economically viable.</li> <li>Excellent for extracting at low concentrations.</li> <li>Effective in the soil.</li> </ul>	<ul style="list-style-type: none"> <li>Slow pace, extending over several months or years.</li> <li>Needs auxiliary technologies from the point of ultimate recovery.</li> </ul>
Bio-extraction	<ul style="list-style-type: none"> <li>It is inexpensive.</li> <li>Minimal carbon emissions.</li> <li>Appropriate for applications related to mining as well.</li> </ul>	<ul style="list-style-type: none"> <li>The capacities of adsorption a topic of the chemical characteristics of water.</li> <li>The development stage of cells impacts the results.</li> </ul>
Application of biochemistry – electrochemistry	<ul style="list-style-type: none"> <li>A small energy consumption and energy efficiency.</li> <li>Organic compounds can be treated in wastewater immediately.</li> <li>Multiple uses and sustainable technology.</li> <li>Metal deposited on the electrode after being obtained.</li> </ul>	<ul style="list-style-type: none"> <li>The use of technology in its early phases of development.</li> <li>It isn't easy to scale up.</li> <li>The reactor's initial and ongoing maintenance costs.</li> </ul>
Technology using membranes	<ul style="list-style-type: none"> <li>Requires minimal space.</li> <li>Minimal production of solid waste materials.</li> <li>Despite high concentration levels, it remains straightforward, quick, and effective.</li> </ul>	<ul style="list-style-type: none"> <li>Small and medium-sized industries regularly face prohibitively expensive costs for investment.</li> <li>Demands for high energy.</li> <li>The flow rates are restricted.</li> </ul>
Adsorption	<ul style="list-style-type: none"> <li>A large selection of targeted pollutants and a broad range of commercial adsorptive materials.</li> <li>Because of the rapid kinetics, there is a need for less time.</li> <li>They can be inexpensive and cost-effective sorbents.</li> </ul>	<ul style="list-style-type: none"> <li>The cost of regeneration can be high.</li> <li>The need for multiple kinds of adsorbents.</li> </ul>

### 3.3. Cost comparison of membrane technique with other techniques: [53-56]

Cost comparisons of membrane separation techniques versus other separation technologies (like distillation, adsorption, and extraction) can vary significantly based on the specific application, feed composition, scale of operation, and local economic conditions. Below is a summary of how membrane techniques generally compare with other methods.

#### 3.3.1. Membrane techniques:

##### a) Advantages:

- *Lower Energy Costs:* Membrane processes often require less energy compared to thermal methods like distillation. They operate at ambient temperatures and pressures.
- *Modularity:* Membrane systems can be easily scaled up or down, making them versatile for different capacities.
- *Chemical Stability:* Membranes can be resistant to fouling and chemical degradation depending on the material used.



b) **Disadvantages:**

- *Capital Costs:* Initial investment in membrane technology can be high due to the cost of membrane materials and module construction.
- *Fouling:* Membranes can foul, leading to increased maintenance and replacement costs over time.

**3.3.2. Cost comparison with other techniques:**a) **Distillation:**

- *Energy Intensive:* Distillation is highly energy-intensive due to the need for heating and cooling. This can significantly raise operational costs.
- *Capital Costs:* While distillation units can have lower capital costs than some membrane systems, the overall operational costs (energy + maintenance) can be higher.

b) **Adsorption:**

- *Regeneration Costs:* While adsorption can be cost-effective for specific applications, the cost of regenerating adsorbents can offset initial savings.
- *Capital Costs:* Adsorption systems may require significant capital investment for large-scale operations, particularly for materials like activated carbon or zeolites.

c) **Liquid-Liquid Extraction:**

- *Solvent Costs:* The use of solvents can be expensive, and their recovery adds to operational complexity and cost.
- *Environmental Concerns:* Disposal of solvents can lead to additional regulatory and cost implications.

d) **General Findings:**

- **Membrane Technology vs. Distillation:** In many cases, membrane technology offers lower operational costs, especially for large-scale separation processes where energy savings can be substantial.
- **Membrane Technology vs. Adsorption:** While adsorption may be cheaper for small-scale or specific separations, membranes tend to be more cost-effective for continuous processes and large volumes.
- **Membrane Technology vs. Extraction:** Membranes generally outperform extraction in terms of sustainability and operational simplicity, particularly when considering the costs associated with solvents.

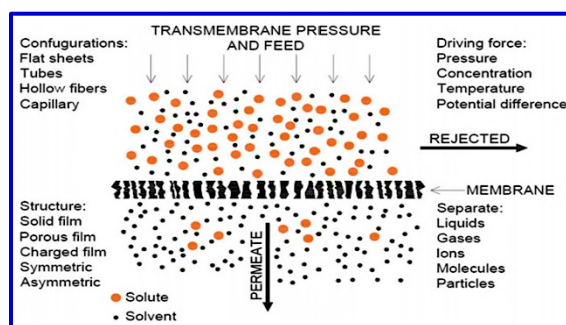
The choice of separation technology should consider both capital and operational costs, as well as the specific requirements of the application. Membrane technology can be a more economical and efficient solution in many cases, especially where energy costs are a major concern.

While membrane techniques, especially RO and NF, have higher initial and operational costs, they offer superior removal efficiencies and can treat a wide range of contaminants simultaneously. The choice of treatment method depends on factors such as the specific heavy metals present, their concentrations, the volume of wastewater to be treated, and local regulations.

**3.4. Membrane science and technology:****3.4.1. Definition and main principles of membrane separation:** [57]

**Definition:** Membrane separation is a technology that selectively separates materials via pores and/or minute gaps in the molecular arrangement of a continuous structure. The membrane acts as a selective barrier, allowing some components to pass through while retaining others based on properties such as size, shape, or chemical structure.

**Main Principle:** The principle behind membrane separation lies in the differential transport of species through the membrane. Depending on the membrane's characteristics and the driving force, such as pressure, concentration gradient, or electrical potential, certain ions and molecules can permeate while others are rejected. The effectiveness of membrane separation depends on the membrane's pore size, structure, material properties, and the characteristics of the feed solution, as shown in Figure (4) [58].



**Fig. (4): The principle of membrane separation** [58]

The gradient of pressure, concentration, or electrical potential can create a transfer force that will cause permeation across the membrane [59]. In the past, synthetic membranes were used for filtering and separation processes and many other smaller-scale applications, including medical equipment, biosensors, controlled release mechanisms, and battery separators. A systematic approach to the processes controlling the efficiency of this significant variation of membrane materials, structural

features, and mechanisms is necessary because of the massive advancements in polymeric membranes and membrane-based technologies and devices in the 1960s and 1970s, which made it possible for the industrial implementation of a vast number of membrane processes [60]. Fast treatment is made possible by membrane technology; however, biofouling and high cost remain drawbacks.

### 3.4.2. General classification of membrane processes:

A membrane morphology, geometry, chemical composition, and separation regime are the most common classifications, as shown in Figure (5) [59].

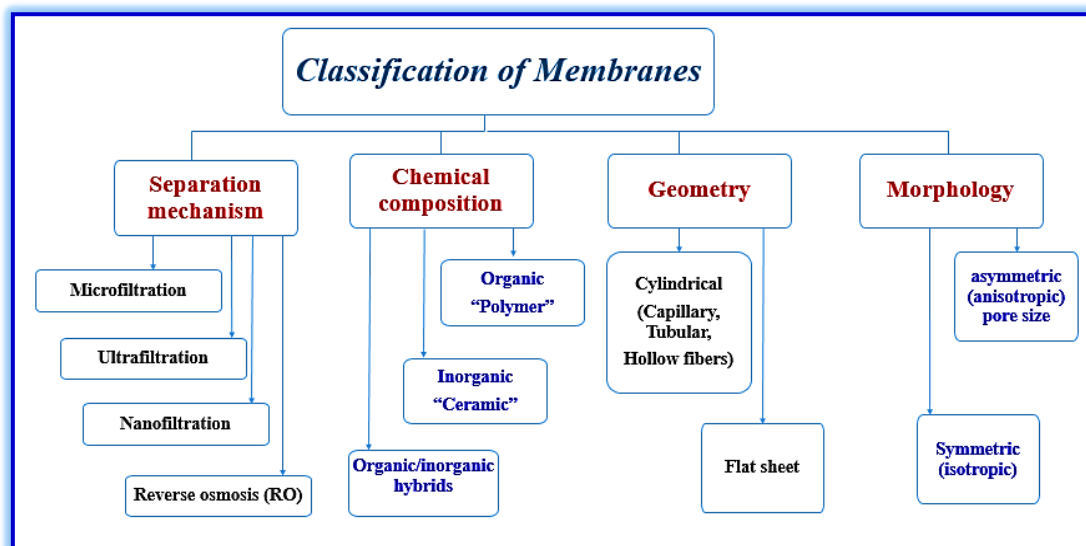


Fig. (5): Classification of membranes [59]

Membrane processes can be classified based on various factors. Membrane processes can be broadly classified into two categories based on their operational conditions: [61]

#### a) Pressure-driven processes:

Membrane types according to Pressure-driven processes are shown in Figure (6) [51].

- *Microfiltration (MF)*: Removes particles and bacteria larger than 0.1  $\mu\text{m}$ . Often used for pre-treatment.
- *Ultrafiltration (UF)*: Separates larger solutes and macromolecules (1-100 nm), often used for concentrating solutions and removing larger organic compounds.
- *Nanofiltration (NF)*: Targets smaller organic molecules and divalent ions (0.5-1 nm); effective for softening water and removing specific contaminants.
- *Reverse Osmosis (RO)*: Provides the highest level of purification, removing nearly all ions and small molecules (size < 0.5 nm).

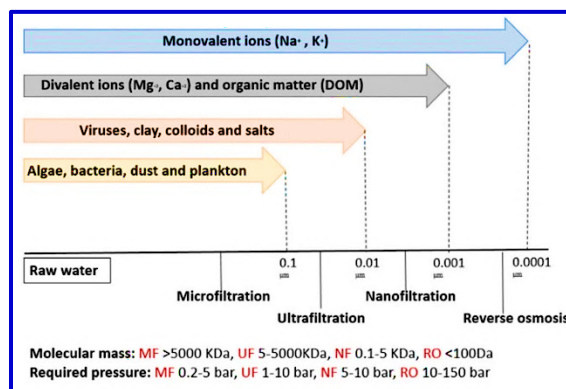


Fig. (6): The principle of membrane separation [51]



b) **Diffusion-driven processes:**

Electrodialysis (ED): Uses an electric field to drive ions through selective cation and anion exchange membranes, effectively separating charged species.

**3.4.3. Classification of membrane processes according to membrane material:**

Membranes can be classified according to their material properties into the following categories:

- a) *Organic (polymeric) membranes*: Made from organic polymers, these are the most common membranes in wastewater treatment. They can be designed for various separation processes, such as ultrafiltration and nanofiltration.
- b) *Inorganic membranes*: Composed of materials such as ceramics or metals, these membranes are more resistant to high temperatures and aggressive chemicals. Inorganic membranes often exhibit higher flux rates and longer lifespans but are costlier and fragile.
- c) *Composite (hybrid) membranes*: These membranes are made of multiple layers of different materials designed to combine the advantages of both polymeric and inorganic membranes, providing enhanced performance and durability.

**3.4.4. Definition and types of inorganic membranes:**

Inorganic membranes are separation devices made from non-carbon-based materials. They can be classified into several types:

- a) *Ceramic membranes*: Made from materials like alumina, zirconia, or titania.
- b) *Metallic membranes*: Composed of metals like palladium or silver.
- c) *Carbon membranes*: Derived from the pyrolysis of organic precursors.
- d) *Zeolite membranes*: Made from crystalline aluminosilicates.
- e) *Glass membranes*: Composed of silica or other glass-forming materials.

**3.4.5. Definition and types of organic membranes:**

**Definition:** Organic membranes are composed of polymeric materials and are widely utilized due to their versatility, processability, and adequate separation performance.

**Types of Organic Membranes:**

- i. *Homogeneous membranes*: These are made from a uniform polymer material and exhibit consistent properties throughout their thickness.
- ii. *Heterogeneous membranes*: These consist of different materials or phases, often being structured to achieve specific separation properties, such as surface modifications to enhance selectivity.
- iii. *Thin-film composite membranes*: These have a thin selective layer on a porous support, improving permeate quality while maintaining high flux.

**3.4.6. Principle and mechanism of heavy metal removal by membrane process:** [37-38, 62-63]

The removal of heavy metals through membrane processes is based on several mechanisms:

- 1) *Size exclusion*: Heavier metal ions, such as lead ( $Pb^{2+}$ ) or cadmium ( $Cd^{2+}$ ), are larger than water molecules. In processes like microfiltration and ultrafiltration, the membrane's pore size can effectively block these larger ions, resulting in their retention while allowing water and smaller molecules to pass through.
- 2) *Charge interaction*: Many metal ions exhibit positive charges in aqueous solutions. Hence, in ion-selective membranes like nanofiltration, negatively charged membranes can repel certain anions while attracting positively charged metal ions. This selective separation enhances overall removal efficiency.
- 3) *Diffusion*: In reverse osmosis, heavy metal ions are driven through a semi-permeable membrane under high pressure. Smaller ions, such as arsenic, are unable to pass through because of the size of the membrane's pores, leading to their retention in the feed solution.
- 4) *Electrodialysis*: The application of an electric current in the presence of selective ion exchange membranes allows for the separation of charged heavy metal ions from neutral or oppositely charged ions. Positively charged membranes (cation-exchange) facilitate the passage of cations while rejecting anions and vice versa for anion-exchange membranes.
- 5) *Adsorption*: Some inorganic membranes can adsorb metal ions onto their surface.
- 6) *Ion Exchange*: Certain membranes can exchange ions with the metal ions in the solution.

The efficiency of removal depends on factors such as membrane material, pore size, surface charge, pH of the solution, and the specific properties of the metal ions. In summary, membrane separation is an advanced method for removing toxic heavy metals from wastewater. By utilizing the properties of membranes and understanding the underlying principles of separation, effective solutions can be applied to achieve regulatory compliance and protect public health. The diverse types of membrane processes used for heavy metals removal are displayed in Table (3).

**Table (3): Types of membrane processes for heavy metals removal**

Membrane process	Characteristics	Application	References
<b>Ultrafiltration</b>	It involves forcing liquid through semi-permeable barriers. High molecular-weight compounds and suspended matter stay on the retentate side of the barrier, while water and low molecular-weight substances flow to the permeate side.	It was provided for the purification and recycling processes of industrial effluents and wastewater, and the elimination of macromolecules and particles.	[64-67]
	Most chemical substances, viruses, and many different kinds of salts can all be removed by UF. It can eliminate 90–100% of pathogenic organisms, produce consistent water quality independent of the source of water, have a minimal physical footprint, and require none of the chemicals other than membrane maintenance that led to its increasing popularity.		
	Pore diameters range from 0.1 to 0.01 $\mu\text{m}$ , and "molecular weight cut-off" (MWCO) is now one of the best terminologies to describe them. It must be cleaned frequently to prevent fouling from solids, scaling, and microbial contaminants like bacteria and algae.		
<b>Nano-filtration</b>	Between reverse osmosis and ultrafiltration, nanofiltration is a pressure-driven barrier technology that can reject all ionic and molecular contaminants. A common technique for eliminating organic debris, colour, odor, taste, residue disinfectant levels, and minimal herbicides from large water bodies is nanofiltration (NF) membrane technology.	<ul style="list-style-type: none"> <li>Nanofiltration is gaining popularity in industrial sectors such as medicinal products, water purification, sewage treatment, the field of biotechnology, and brine water desalination.</li> <li>The NF method is employed in industry to clean olive mill sewage, recover metals, and separate pigments in the waste from the textile industry.</li> <li>In the oil and petroleum industries, NF has also been employed in the filtration method of effluents from pulp and paper manufacturing, greasy wastewater, coke wastewater, and mine water to remove acid sulfate.</li> </ul>	[68-71]
	Hardness, dyes, and heavy metals are separated from low molecular weight solutes such lactose, glucose, and salt using the characteristics of NF, which include a 1–5 nm pore size and an operating pressure from 7 to 30 bar. Since NF runs at lower pressures of 8–30 bar than RO, it can obtain higher flux at lower process running costs. The Donnan phenomenon and the sieve mechanism are the two operational mechanisms that regulate transport across the NF membrane.		
<b>The reverse osmosis method</b>	Reverse osmosis (RO) membranes are a low-cost option for water purification in wastewater treatment plants. RO membranes have been demonstrated to efficiently decrease the overall concentration of heavy metals, organic substances, pathogenic bacteria, viruses, and other dissolved pollutants.	<ul style="list-style-type: none"> <li>The effluent produced by metal finishing and plating processes is treated and recycled by RO.</li> <li>In circuit boards for electronic devices and semiconductors (recycling and treating rinse fluids utilized in electroplating procedures).</li> <li>In the production of automobiles (cleaning and painting water treatment and recycling).</li> <li>Food and drinks (reduction of biological before discharge and concentration of sewage for reuse).</li> <li>Leachate from landfills and groundwater (which removes salts and heavy metals before being released).</li> </ul>	[72-74]
	The removal of dissolved solids and small particles is accomplished via pressure-driven RO, which only allows water molecules to flow through. For the water to overcome the osmotic pressure, a high enough pressure must be supplied to the RO.		
	Compared to UF barriers, the membranes of RO systems have a much tighter pore structure that is less than 0.1 nm and can effectively remove all particles, germs, and organic molecules while additionally converting hard water to soft water. It needs only minimal maintenance.		

#### 4. Case Studies for Membrane Techniques for the Elimination of Heavy Metals:

##### 4.1. Removal of Cr(VI):

One of the heavy metals that harm the ecosystem is chromium. There are several different oxidation states of chromium, including Cr(III), (IV), (V), and (VI). Trivalent and hexavalent chrome ions are the two chromium oxidation states that are the most stable. Due to its teratogenic and carcinogenic effects on humans and animals, Cr(VI) ions are considerably more harmful than Cr(III) ions. Edema, skin irritation, pulmonary congestion, and liver damage are side effects of prolonged exposure to Cr(VI) ions. Numerous sectors use chromium metal extensively, including leather, plating,

photographic, mineral extraction, paints manufacture, wood preserving, fabric dyeing, and chromate production. Effluents of these industries significantly contribute to water contamination. “The recommended level for drinking water is only 0.05 mg/L, while the tolerance level for Cr(VI) ions for release into inland surface water is 0.1 mg/L, according to the Environmental Protection Agency (EPA) of the United States of America (USA)” [27]. Some of membranes that are used to remove hexavalent chrome ions from water are displayed in Table (4).

**Table (4): Membranes used to remove hexavalent chrome**

Membrane / Material	Characteristics	Modification	Initial Concentration	Removal Efficiency	References
Ball clay, feldspar, kaolin, pyrophyllite, quartz, and calcium carbonate	- Tubular substrate porosity was 53%. - Membrane pore size was 0.272 $\mu\text{m}$ . - Water permeability $4.43 \times 10^{-7} \text{ m}^3/\text{m}^2 \cdot \text{s} \cdot \text{kPa}$ .	Mordenite framework inverted (MFI) zeolite membrane	1000 ppm	78%	[75]
Porous clay-alumina ceramic support tube	- Pore size was 3 nm. - The permeability was $34.99 \text{ L}/\text{m}^2 \text{ h bar}$ .	Hydroxyethyl cellulose and CuO nanoparticles	Not reported	91.44%	[76]
Tubular clay-alumina ceramic support	- Permeability to clean water of $85.51 \text{ L}/\text{m}^2 \text{ h bar}$ .	Catharanthus Roseus leaf extract and CuO nanoparticles	Not reported	88.08%	[77]
Clinoptilolite and a natural zeolite hollow fiber ceramic membrane	- The water permeability was $29.14 \text{ L}/\text{m}^2 \text{ h}$ and mechanical strength was 50.92 MPa.	---	40 ppm	44%	[78]
Waste animal bones / bio-ceramic hollow fiber membrane	- The high flux of $88.3 \text{ L}/\text{m}^2 \text{ h}$ .	---	Not reported	100%	[79]
Clay and sawdust	---	Impregnated with silver nanoparticles	Not reported	57.3%	[80]
Natural zeolite support	- Tubular microporous supports with pore diameter of 0.55 $\mu\text{m}$ and porosity of 43.7%. - Modified membrane with pore diameters of 3 nm - 5 nm and a permeate flux of $59 \text{ L}/\text{m}^2 \text{ h}$ .	Smectite nanoparticles	Not reported	>89%	[81]
Clay and alumina ceramic microfiltration support	- Tubular support with 1.6 $\mu\text{m}$ pore size.	Polyethyleneimine modified with ethylenediaminetetraacetic acid	Not reported	97.22%	[82]
$\alpha\text{-Al}_2\text{O}_3$ ceramic membrane	- The permeate flux for modified membrane was $85.6 \text{ L}/\text{m}^2 \text{ h}$ .	3-aminopropyltriethoxysilane-modified mesoporous silica SBA 16	Not reported	100%	[83]
Clay, perlite, and iron	---	---	Not reported	> 99%	[84]
Kaolin, nano-magnetite, magnesium carbonate, methocel, and starch	---	Polyethersulfone	10 ppm	96.2%	[85]
Industrial rutile, fly ash, and graphite	---	TiO <sub>2</sub> nanofiber	Not reported	97.09%	[86]
Ceramic microfiltration membrane	- Having an average pore size of 1.6 $\mu\text{m}$ . - Tubular membrane.	Copper ions with the polyethyleneimine matrix	Not reported	> 95%	[87]
Kaolin, clay, and starch	- The membrane had a water absorption rate, a compressive strength, and a pure water permeability of 27.27%, 31.05 MPa, and $20.74 \text{ L}/\text{m}^2 \text{ h}$ , respectively. - Chitosan coated ceramic membrane has pores that are 16.24 nm in size.	Chitosan with glutaraldehyde crosslinking	Not reported	71.25%	[88]

#### 4.2. Removal of Cd(II):

Cadmium (Cd), one of the heavy metal contaminants, is present in the water body and is an environmental issue that harms the quality of water resources. The ceramic membrane used for groundwater treatment is printed in a gypsum cylinder mold with dimensions of 4 cm on the inside, 5 cm on the outside, 0.5 cm thick, and 20 cm in length. The ceramic filter is

made of clay, Andisol soil, and flour powder. The groundwater was processed using a ceramic membrane, activated carbon pellets, a carbon block, osmosis filters, and activated carbon powder as the final step. Measurement outcomes demonstrated that the ceramic filter was efficient in lowering cadmium ions concentration in water wells (98.9%) and reducing dissolved solids levels (94%) [89]. Some of membranes that are used to remove cadmium ions from water are displayed in Table (5).

**Table (5): Membranes used to remove cadmium**

Membrane / Material	Characteristics	Modification	Initial Concentration	Removal Efficiency	References
Waste animal bones / bio-ceramic hollow fiber membrane	- The high flux of 88.3 L/m <sup>2</sup> h.	---	Not reported	100%	[79]
Industrial rutile, fly ash, and graphite	---	TiO <sub>2</sub> nanofiber	Not reported	96.38%	[86]
Ceramic microfiltration membrane	- Having an average pore size of 1.6 µm. - Tubular membrane.	Copper ions with the polyethyleneimine matrix	Not reported	> 95%	[87]
Palm oil fuel ash, Polyethersulfone, and metakaolin powder	---	---	Not reported	96.4%	[90]
Alumina-carbon nanotube composite membrane	- Membrane porosity was 31%, and its strength was 15.64 MPa.	---	1 ppm	93%	[91]
Carbon nanotubes, alumina, starch (as a pore-forming agent), and dispersion agents (sodium dodecyl sulfate, and gum Arabic)	- Solid porous nanocomposite membrane has a pure water flux of 37.8 L/m <sup>2</sup> hr bar, compressive strength of 9.5 MP, and mean size of the pore of 0.14 µm.	---	Not reported	97%	[92]
Ceramic support tube	- Pure water flux was 8.09 L/m <sup>2</sup> h bar. - Positively charged nanofiltration membrane.	Cu nanoparticles and Polytetrafluoroethylene	Not reported	95.5%	[93]
Clay and activated carbon	- The pore's diameter reduced from 35.5 nm (for the support) to 14.2 nm (for the modified membrane)	Zeolite Linde type A (LTA)-type	Not reported	88.3%	[94]

#### 4.3. Removal of Zn(II):

Zinc is a trace element that is benign mainly to humans, but excessive amounts can induce stomach pains, irritability, vomiting, nausea, anemia, and even death [95]. Some of membranes that are used to remove zinc ions from water are displayed in Table (6).

**Table (6): Membranes used to remove zinc**

Membrane / Material	Characteristics	Modification	Initial Concentration	Removal Efficiency	References
Waste animal bones / bio-ceramic hollow fiber membrane	- The high flux of 88.3 L/m <sup>2</sup> h.	---	Not reported	100%	[79]
Ceramic microfiltration membrane	- Having an average pore size of 1.6 µm. - Tubular membrane.	Copper ions with the polyethyleneimine matrix	Not reported	> 95%	[87]
Clay and activated carbon	- The pore's diameter reduced from 35.5 nm (for the support) to 14.2 nm (for the modified membrane)	Zeolite Linde type A (LTA)-type	Not reported	81.8%	[94]
Clay, iron powder, and rice husk	- It has a 2.8 µm pore size and a surface area of 45.38 m <sup>2</sup> /g.	---	Not reported	99.80%	[96]
Rice husk ash and Polyethersulfone	- Green hollow fiber and high flux membrane.	---	Not reported	up to 99%	[98]
Mono-tubular ceramic membrane	---	The anionic surfactant sodium dodecyl sulfate	Not reported	up to 99%	[99]
Ceramic substrate	- Hollow fiber. - 1.06 µm in pore size.	Graphene oxide grafted with ethylenediamine solution. A thin film of polyamide	Not reported	93.33%	[100]
Iranian clay	- The membrane porosity ranged from 57 to 58%	SnO <sub>2</sub> /Montmorillonite nanocomposite	5-50 ppm	76.79-92.23%	[101]

**4.4. Removal of As:**

One of the most dangerous heavy metals considered to be arsenic, and the most common environmental toxin in the world, found in abundance in rocks, soils, and groundwater. It is categorized as a Group 1 human malignancy by the International Agency for Research on Cancer (IARC). In the Earth's crust, arsenic is the twenty-ninth most common element; in seawater, it is fourteenth, and in human tissue, it ranks twelve. There are two distinct forms of arsenic: ions of arsenite (As(III)) and arsenate (As(V)). In surface water streams and groundwater, arsenic is often present in trace concentrations [102]. Arsenic metal is a carcinogen and can impair the cardiovascular, dermatologic, neurological, hepatobiliary, renal, gastrointestinal, and respiratory systems [97]. Some of membranes that are used to remove arsenic ions from water are displayed in Table (7).

**Table (7): Membranes used to remove arsenic**

Membrane / Material	Characteristics	Modification	Initial Concentration	Removal Efficiency	References
Clay and alumina ceramic microfiltration support	- Tubular support with 1.6 µm pore size.	Polyethyleneimine modified with ethylenediaminetetraacetic acid	Not reported	96.75%	[82]
Ceramic microfiltration membrane	- Having an average pore size of 1.6 µm. - Tubular membrane.	Copper ions with the polyethyleneimine matrix	Not reported	> 95%	[87]
Silica-based rice husk ash hollow fiber ceramic membranes	- Hydrophobic membrane had fluxes of 50.4 kg /m <sup>2</sup> h and 51.3 kg /m <sup>2</sup> h for As(III) and As(V) ions. - With a 0.5 µm pore size	The 1H,1H,2H,2H-perfluorodecyltriethoxysilane	Not reported	up to 99.6%	[102]
Iron, perlite, and montmorillonite	---	---	Not reported	97%	[103]

#### 4.5. Removal of Ni(II)

Chronic bronchitis, impaired lung function, and lung cancer are among the significant health impacts of nickel, which also causes allergic reactions in humans [97]. Some of membranes that are used to remove nickel ions from water are displayed in Table (8).

**Table (8): Membranes used to remove nickel**

Membrane / Material	Characteristics	Modification	Initial Concentration	Removal Efficiency	References
Ceramic microfiltration membrane	- Having an average pore size of 1.6 $\mu\text{m}$ . - Tubular membrane.	Copper ions with the polyethyleneimine matrix	Not reported	> 95%	[87]
Rice husk ash and Polyethersulfone	- Green hollow fiber and high flux membrane.	---	Not reported	up to 99%	[98]
Ceramic substrate	- Hollow fiber. - 1.06 $\mu\text{m}$ in pore size.	Graphene oxide grafted with ethylenediamine solution. A thin film of polyamide	Not reported	90.45%	[100]
Iranian clay	- The membrane porosity ranged from 57 to 58%	SnO <sub>2</sub> /Montmorillonite nanocomposite	5-50 ppm	24.97-64.74%	[101]
Alumina particles, N-methyl-2-pyrrolidone, polyether sulfone, and a surfactant	- Hollow ceramic fiber.	---	Not reported	63%	[104]
Sayong ball clay, methacrylamide, and N,N'-methylenebisacrylamide	---	---	Not reported	59%	[105]

#### 4.6. Removal of Cu(II):

Cu(II) ions are present in wastewater diffuse into the soil and water streams. It will eventually accumulate along with the food chain, endangering human health. Food pollutants like copper ions are typically found in mushrooms, liver, chocolate, almonds, and seafood. Consuming too much copper ions damages blood capillaries, irritates the intestinal tract, causes significant mucosal irritation, and causes necrotic changes in the liver and kidney. Conventional techniques typically include the reaction of copper ions with the compounds to produce insoluble precipitates that are separated through sedimentation or filtration [95]. Some of membranes that are used to remove copper ions from water are displayed in Table (9).

**Table (9): Membranes used to remove copper**

Membrane / Material	Characteristics	Modification	Initial Concentration	Removal Efficiency	References
Waste animal bones / bio-ceramic hollow fiber membrane	- The high flux of 88.3 L/m <sup>2</sup> h.	---	Not reported	100%	[79]
Clay and alumina ceramic microfiltration support	- Tubular support with 1.6 $\mu\text{m}$ pore size.	Polyethyleneimine modified with ethylenediaminetetraacetic acid	Not reported	99.82%	[82]
Industrial rutile, fly ash, and graphite	---	TiO <sub>2</sub> nanofiber	Not reported	90.15%	[86]
Ceramic substrate	- Hollow fiber. - 1.06 $\mu\text{m}$ in pore size.	Graphene oxide grafted with ethylenediamine solution. A thin film of polyamide	Not reported	92.73%	[100]
Iranian clay	- The membrane porosity ranged from 57 to 58%	SnO <sub>2</sub> /Montmorillonite nanocomposite	5-50 ppm	97.88-99.26%	[101]
Sayong ball clay, methacrylamide, and N,N'-methylenebisacrylamide.	---	---	Not reported	87%	[105]
Kaolin, silica, starch, graphite, and sodium silicate	---	Combining the electrolysis procedure with the created ceramic membrane	350 ppm	97%	[106]



#### 4.7. Removal of Pb(II):

Lead impairs brain and nervous system development, which is especially hazardous to fetuses and young children. Lead harms to the kidneys, reproductive organs and neurological systems [97]. Some of membranes that are used to remove lead ions from water are displayed in Table (10).

**Table (10): Membranes used to remove lead**

Membrane / Material	Characteristics	Modification	Initial Concentration	Removal Efficiency	References
Porous clay-alumina ceramic support tube	- Pore size was 3 nm. - The permeability was 34.99 L/m <sup>2</sup> h bar.	Hydroxyethyl cellulose and CuO nanoparticles	Not reported	97.14%	[76]
Ceramic microfiltration membrane	- Having an average pore size of 1.6 µm. - Tubular membrane.	Copper ions with the polyethyleneimine matrix	Not reported	> 95%	[87]
Clay and activated carbon	- The pore's diameter reduced from 35.5 nm (for the support) to 14.2 nm (for the modified membrane)	Zeolite Linde type A (LTA)-type	Not reported	99.7%	[94]
Rice husk ash and Polyethersulfone	- Green hollow fiber and high flux membrane.	---	Not reported	up to 99%	[98]
Ceramic substrate	- Hollow fiber. - 1.06 µm in pore size.	Graphene oxide grafted with ethylenediamine solution. A thin film of polyamide	Not reported	88.35%	[100]
Clay-alumina ceramic substrate	- The membrane nominal pore size was 2.8 nm	Hydroxyapatite nanoparticles	5 ppm	99.6%	[107]
Activated zeolite powder and polyethylene glycol	---	---	200 ppm	87%	[108]

### 5. Limitations of Membrane Separation Processes:

#### 5.1. Limitations in inorganic membrane technology: [109-110]

While membrane separation processes are increasingly recognized for their efficacy in removing heavy metals and other contaminants from wastewater, they face several challenges and limitations that can hinder their performance and applicability. This section explores these limitations in detail and discusses strategies for controlling fouling across different types of inorganic membranes.

##### 1) **Fouling:**

- *Definition:* Fouling is the accumulation of unwanted materials on the membrane surface or within its pores, which leads to a decline in membrane performance, including reduced permeate flux and increased transmembrane pressure, different types of membrane fouling are shown in Figure (7) [51].
- *Types of Fouling:* Fouling can primarily be classified into three categories:
- *Particulate fouling:* Due to the deposition of suspended solids and colloids.
- *Organic fouling:* Caused by the adsorption of macromolecules and organic substances onto the membrane surface.
- *Inorganic fouling (scaling):* The deposition of minerals such as calcium carbonate and sulfate salts, usually resulting from supersaturation conditions in the feed solution.
- *Impact:* The presence of fouling not only reduces the efficiency of the membrane but also necessitates frequent cleaning and maintenance, increasing the operational costs.

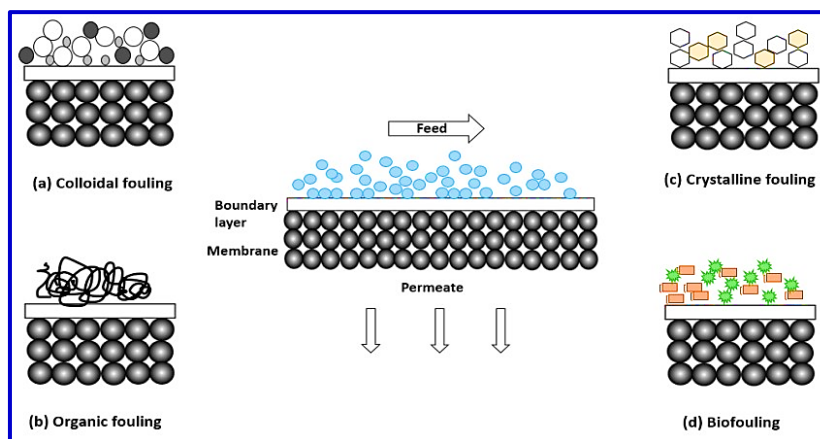


Fig. (7): Different types of membrane fouling [51]

- 2) **Membrane Material Limitations:** Durability and Chemical Resistance: Although inorganic membranes, including those made of ceramics, typically demonstrate superior durability and chemical resistance compared to polymeric membranes, they can still be susceptible to specific solvents or extreme pH levels, limiting their applicability.
- 3) **Membrane Selectivity:** Membrane processes may struggle to achieve effective separation if the feed solution contains a broad spectrum of contaminants, as the selectivity of the membrane may not discriminate well among similarly sized ions and molecules. This is particularly relevant when addressing mixtures of heavy metals at low concentrations.
- 4) **Reduced Flux Over Time:** Over extended periods, the permeate flux through the membrane typically declines due to fouling, concentration polarization, and the membrane's aging or degradation.
- 5) **Limited Flow Rates:** The design and size of certain inorganic membranes may limit their flow rates, impacting the overall operational efficiency for large-scale applications.
- 6) **Concentration Polarization:** The build-up of rejected solutes near the membrane surface, reducing effective driving force.
- 7) **Mechanical Strength:** Brittleness of some ceramic membranes can lead to cracking under high pressures.
- 8) **Cost Issues:** Inorganic membranes are often more expensive than their polymeric counterparts, owing to their complex manufacturing processes and materials. This high initial cost can be a barrier for widespread adoption, especially in financially constrained applications.
- 9) **Scalability:** Challenges in producing large-scale, defect-free inorganic membranes.

## 5.2. Controlling fouling in inorganic membranes: [111-112]

To mitigate fouling, a variety of strategies can be employed across the range of inorganic membranes, including ceramic membranes as shown in Table (11).

Despite the considerable advantages of membrane separation processes in treating water contaminated by heavy metals, challenges such as fouling, material limitations, and suboptimal flux rates remain significant hurdles. Understanding and employing effective strategies for fouling control, along with optimizing operational conditions, will enhance the performance and longevity of inorganic membranes. Continued research and development in membrane technology hold promise for overcoming these limitations and facilitating the broader application of membrane systems in environmental remediation and industrial wastewater treatment.

**Table (11): Different strategies for fouling control [111-112]**

Name of Strategy	Details
<b>1. Pre-treatment of Feed Solutions</b>	
a) Coagulation and Flocculation	The addition of coagulants can enhance the agglomeration of suspended solids, leading to easier removal before the feedwater reaches the membrane. This method helps in minimizing particulate fouling.
b) Microfiltration	Using microfiltration as a pre-treatment step can effectively remove larger particles and colloids, thereby reducing the fouling load on downstream membranes like ultrafiltration or reverse osmosis.
c) Filtration Systems Design	Implementing appropriate sedimentation tanks or clarification systems prior to the membrane unit can significantly decrease the particulate load.
<b>2. Membrane Surface Modification</b>	
a) Hydrophilization	Modifying the membrane surface to increase its hydrophilicity can enhance its resistance to organic fouling by improving water flux and reducing the adsorption of organic substances. Techniques include graft polymerization and surface coatings.
b) Functionalization	Incorporating specific functional groups that repel certain foulants can also be effective. For instance, the introduction of charged groups can enhance the membrane's selectivity toward certain ions.
c) Antimicrobial modifications to prevent biofouling.	
<b>3. Operational Adjustments</b>	
a) Flux Management	Adjusting the operational flux can significantly reduce fouling rates. Maintaining lower operating pressures and fluxes can minimize shear stress on the membrane surface, thereby reducing the likelihood of fouling.
b) Backwashing	Regular backwashing (reverse flow) of the membrane can dislodge foulants that have adhered to the membrane surface, restoring flux without major chemical cleaning.
<b>4. Chemical Cleaning</b>	
a) Cleaning Protocols	Routine chemical cleaning with appropriate cleaning agents (e.g., sodium hypochlorite, citric acid) can help remove fouling layers that have formed on the membrane surface. Establishing a comprehensive cleaning strategy based on the nature of fouling is crucial.
b) Use of Antifouling Agents	Adding antifouling agents to the feed solution can prevent the adhesion of foulants and facilitate easier maintenance.
c) Acid cleaning for inorganic scaling	
d) Alkaline cleaning for organic fouling	
e) Enzymatic cleaning for specific foulants	
<b>5. Hybrid Systems</b>	
Combining membrane processes with other separation methods (such as adsorption, advanced oxidation processes, or biological treatments) can be beneficial in managing fouling. These hybrid systems allow for considerable overall removal of contaminants while lowering fouling potential.	

**6. Conclusion and Suggestions for the Future Research Needs:**

One of the most pressing issues confronting the planet is the demand for safe drinking water. Since the majority is contaminated and salty, and only 2.5 percent are classified as clean water. The demand for safe and pure water is rising globally as a result of the world's rapid population growth, growing manufacturing, growing cities, and extensive agricultural practices. Numerous techniques are now being used to purify and disinfect the water. Furthermore, since heavy metals and some bacteria cannot be removed by traditional processes, it is important to treat wastewater using innovative techniques. The different forms of membrane technologies will become progressively more important to the industry's ability to handle wastewater and water. After a thorough assessment of the literature, it was discovered that ceramic membranes (CMs) outperformed traditional techniques in the elimination of heavy metals. To create more valuable and affordable membranes for eliminating hazardous heavy metals from effluents and solutions, a variety of materials have been employed, including waste materials, modified carbon materials, clay minerals, and modified clays. Lately, waste materials management in the production of ceramic membranes has been the focus of research. As a result, more investigation must be conducted in this area. Therefore, the implementation of membrane-based methods in sewage treatment seems highly promising and may have a bright future; nevertheless, scientific societies and government bodies must make an extensive and coordinated investment in this field.

By adding nanomaterials, the effectiveness of NF and RO in removing heavy metals might be improved. By using nanocomposite membranes, water flow, and heavy metal rejecting can both be enhanced. Carbon nanoparticles are thought to be the perfect technology needed for wastewater treatment in future processes involving membrane separation because of their many exceptional qualities. Carbon nanomaterials, with their huge surface areas, size, and unique optical, electrical, and catalytic features, have demonstrated considerable potential for developing more effective wastewater cleanup methods. Nonetheless, other obstacles related to toxic effects, fouling, and other issues persist, which clarifies the need for nanoparticle modifications or functionalization, primarily concerning carbon nanotubes (CNTs). Additionally, modified groups that aid in excellent dispersion in solutions and polymer matrix can be generated by the functionalization of CNTs and graphene oxide (GO) sheets. Membrane efficiency and characteristics have improved as a result of innovation. The most promising technologies for sewage treatment and many additional sectors will emerge from the exceptional efficacy and ongoing development of barriers based on enhanced or functionalized carbon nanoparticles.

When producing carbon-based nanomaterials, accuracy and a deeper understanding of the surface shape and characteristics are necessary for functionalization. Although these carbon nanoparticles are thought to be cost-effective, creating an effective carbon nano-membrane is a labour-intensive procedure that calls for personnel with more qualifications in this area as well as competent employees. Scientists and investigators need to be informed on the long-term effects of CNM concentrations on our surroundings, as well as the hazards of specific CNMs. This raises the cost challenge as well. Therefore, additional research is required to successfully synthesize these nano-based materials and use them in membrane filtration techniques. Certainly, the use of CNMs in membrane separation for treating wastewater has shown to be beneficial and not economical; nevertheless, these applications are limited to smaller-scale, non-mass operations. To determine their effectiveness for large-scale uses, more investigation is required.

#### Conflict of Interest:

The authors declare they have no competing interests.

#### Formatting of Funding Sources:

This study received no explicit financing from public, commercial, or non-profit organizations.

#### Acknowledgments:

The authors would like to acknowledge the Academy of Scientific Research and Technology (ASRT), Egypt, for their financial support and for all the facilities they offered to perform this work through the "Locally Fabrication of Industrial Ceramic Membranes for Water Treatment", ASRT-APPLE Program, Energy, Environment and Water Nexus.

#### References and Bibliography:

- [1] He Z, Lyu Z, Gu Q, Zhang L, Wang J. Ceramic-based membranes for water and wastewater treatment. *Colloids Surf. A*. 2019; 578:123513. <https://doi.org/10.1016/j.colsurfa.2019.05.074>.
- [2] Le NL, Nunes SP. Materials and membrane technologies for water and energy sustainability. *Sustain. Mater. Technol.* 2016; 7:1-28. <https://doi.org/10.1016/j.susmat.2016.02.001>.
- [3] El-Gawad, Heba A., Ghada Kadry, Hamdy A. Zahran, and M. H. Hussein. "Chromium disarmament from veritable tanneries sewer water utilizing carbonic rice straw as a sorbent: Optimization and carbonic rice straw characteristics." *Water, Air, & Soil Pollution* 234, no. 10, 2023: 659.
- [4] Issaoui M, Jellali S, Zorpas AA, Dutournie P. Membrane technology for sustainable water resources management: Challenges and future projections. *Sustain. Chem. Pharm.* 2022; 25:100590. <https://doi.org/10.1016/j.scp.2021.100590>.
- [5] Badejo AA, Omole DO, Ndambuki JM. Municipal wastewater management using *Vetiveria zizanioides* planted in vertical flow constructed wetland. *Appl. Water Sci.* 2018; 8. <https://doi.org/10.1007/s13201-018-0756-0>.
- [6] Almuktar SAAAN, Scholz M, Al-Isawi RHK, Sani A. Recycling of domestic wastewater treated by vertical-flow wetlands for watering of vegetables. *Water Pract. Technol.* 2015; 10:445-464. <https://doi.org/10.2166/wpt.2015.052>.
- [7] Masindi V, Muedi K. Environmental Contamination by Heavy Metals. 2018; Chapter 7, 115-133. <http://dx.doi.org/10.5772/intechopen.76082>.
- [8] Selvi A, Rajasekar A, Theerthagiri J, Ananthaselvam A, Sathishkumar K, Madhavan J, Rahman PK. Integrated remediation processes toward heavy metal removal/recovery from various environments-a review. *Front. Environ. Sci.* 2018; 7:66. doi:10.3389/fenvs.2019.00066.
- [9] Jaishankar M, Tseten T, Anbalagan N, Mathew BB, Beeregowda KN. Toxicity, mechanism and health effects of some heavy metals. *Interdiscip Toxicol.* 2014;7(2):60-72. doi:10.2478/intox-2014-0009.
- [10] Garai P, Banerjee P, Mondal P, Saha NC. Effect of heavy metals on fishes: Toxicity and bioaccumulation. *J. Clin. Toxicol.* 2021; 11(S18):1-10.
- [11] Mahurpawar M. Effects of heavy metals on human health. *Int. J. Res. Granthaalayah.* 2015; 530:1-7. DOI:10.29121/granthaalayah.v3.i9SE.2015.3282.
- [12] Engwa GA, Ferdinand PU, Nwalo FN, Unachukwu MN. Mechanism and health effects of heavy metal toxicity in humans. *Poisoning in the Modern World - New Tricks for an Old Dog.* Intechopen. 2019; 10:70-90. DOI:10.5772/intechopen.82511.
- [13] Bhateria R, Singh R. A review on nanotechnological application of magnetic iron oxides for heavy metal removal. *J. Water Process. Eng.* 2019; 31. <https://doi.org/10.1016/j.jwpe.2019.100845>.

- [14] Talukdar D, Jasrotia T, Sharma R, Jaglan S, Kumar R, Vats R, Kumar R, Mahnashi MH, Umar A. Evaluation of novel indigenous fungal consortium for enhanced bioremediation of heavy metals from contaminated sites. *Environ. Technol. Innov.* 2020; 20. <https://doi.org/10.1016/j.eti.2020.101050>.
- [15] Zamora-Ledezma C, Negrete-Bolagay D, Figueroa F, Zamora-Ledezma E, Ni M, Alexis F, Guerrero VH. Heavy metal water pollution: A fresh look about hazards, novel and conventional remediation methods. *Environ. Technol. Innov.* 2021; 22. <https://doi.org/10.1016/j.eti.2021.101504>.
- [16] Rasheed T, Bilal M, Nabeel F, Iqbal HM, Li C, Zhou Y. Fluorescent sensor-based models for the detection of environmentally-related toxic heavy metals. *Sci. Total Environ.* 2018; 615:476-485. <https://doi.org/10.1016/j.scitotenv.2017.09.126>.
- [17] Zarenezhad M, Zarei M, Ebratkhahan M, Hosseinzadeh M. Synthesis and study of functionalized magnetic graphene oxide for Pb<sup>2+</sup> removal from wastewater. *Environ. Technol. Innov.* 2021; 22. <https://doi.org/10.1016/j.eti.2021.101384>.
- [18] Nezhadheydari H, Tavabe KR, Mirvaghefi A, Heydari A, Frinsko M. Effects of different concentrations of Fe<sub>3</sub>O<sub>4</sub>@ZnO and Fe<sub>3</sub>O<sub>4</sub>@CNT magnetic nanoparticles separately and in combination on aquaculture wastewater treatment. *Environ. Technol. Innov.* 2019; 15. <https://doi.org/10.1016/j.eti.2019.100414>.
- [19] Vasseghian Y, Dragoi EN, Almomani F, Le VT, Berkani M. Graphene-based membrane techniques for heavy metal removal: A critical review. *Environ. Technol. Innov.* 2021; 24. <https://doi.org/10.1016/j.eti.2021.101863>.
- [20] Alloway BJ. Sources of Heavy Metals and Metalloids in Soils. In: Alloway, B. (eds) *Heavy Metals in Soils*. *Environ. Pollut.* 2013; 22. Springer, Dordrecht. [https://doi.org/10.1007/978-94-007-4470-7\\_2](https://doi.org/10.1007/978-94-007-4470-7_2).
- [21] McLaughlin MJ, Smolders E, Zhao FJ, Grant C, Montalvo D. Chapter One - Managing cadmium in agricultural systems, Editor(s): Donald L. Sparks, *Advances in Agronomy*. Academic Press. 2021; 166: p.1-129. <https://doi.org/10.1016/bs.agron.2020.10.004>.
- [22] Taka M, Sillanpää N, Niemi T, Warsta L, Kokkonen T, Setälä H. Heavy metals from heavy land use? Spatio-temporal patterns of urban runoff metal loads. *Sci. Total Environ.* 2022; 817:152855. <https://doi.org/10.1016/j.scitotenv.2021.152855>.
- [23] Houessionon MGK, Ouendo ED, Bouland C, Takyi SA, Kedote NM, Fayomi B, Fobil JN, Basu N. Environmental Heavy Metal Contamination from Electronic Waste (E-Waste) Recycling Activities Worldwide: A Systematic Review from 2005 to 2017. *Int. J. Environ. Res. Public Health.* 2021; 18(7): 3517. <https://doi.org/10.3390/ijerph18073517>.
- [24] Bradl HB. Chapter 1 - Sources and origins of heavy metals. Editor(s): H.B. Bradl, *Interface Sci. Technol.* Elsevier. 2005; 6: p.1-27. [https://doi.org/10.1016/S1573-4285\(05\)80020-1](https://doi.org/10.1016/S1573-4285(05)80020-1).
- [25] Shetty SS, Deepthi D, Harshitha S, Sonkusare S, Naik PB, Kumari S, Madhyastha H. Environmental pollutants and their effects on human health. *Heliyon.* 2023; 9(9): e19496. <https://doi.org/10.1016/j.heliyon.2023.e19496>.
- [26] Järup L. Hazards of heavy metal contamination. *Br. Med. Bull.* 2003; 68(1): 167-182. <https://doi.org/10.1093/bmb/ldg032>.
- [27] United States Environmental Protection Agency (EPA). (2021). *Drinking Water Standards*.
- [28] United States Environmental Protection Agency (EPA). (2020). *National Pollutant Discharge Elimination System (NPDES)*.
- [29] Fouda-Mbanga BG, Velempini T, Pillay K, Tywabi-Ngeva Z. Heavy metals removals from wastewater and reuse of the metal loaded adsorbents in various applications: A review. *Hybrid Advances.* 2024; 6:100193. <https://doi.org/10.1016/j.hybadv.2024.100193>.
- [30] Qasem NAA, Mohammed RH, Lawal DU. Removal of heavy metal ions from wastewater: a comprehensive and critical review. *npj Clean Water.* 2021; 4(36). <https://doi.org/10.1038/s41545-021-00127-0>.
- [31] Kampalanonwat P, Supaphol P. The Study of Competitive Adsorption of Heavy Metal Ions from Aqueous Solution by Aminated Polyacrylonitrile Nanofiber Mats. *Energy Procedia.* 2014; 56:142-151. <https://doi.org/10.1016/j.egypro.2014.07.142>.
- [32] Zamora-Ledezma C, Negrete-Bolagay D, Figueroa F, Zamora-Ledezma E, Ni M, Alexis F, Guerrero VH. Heavy metal water pollution: A fresh look about hazards, novel and conventional remediation methods. *Environ. Technol. Innov.* 2021; 22: 101504. <https://doi.org/10.1016/j.eti.2021.101504>.
- [33] Zaimie MZA, Sarjadi MS, Rahman ML. Heavy Metals Removal from Water by Efficient Adsorbents. *Water.* 2021; 13(19):2659. <https://doi.org/10.3390/w13192659>.
- [34] Vardhan KH, Kumar PS, Panda RC. A review on heavy metal pollution, toxicity and remedial measures: Current trends and future perspectives. *J. Mol. Liq.* 2019; 290: 111197. <https://doi.org/10.1016/j.molliq.2019.111197>.
- [35] Carolin CF, Kumar PS, Saravanan A, Joshiba GJ, Naushad M. Efficient techniques for the removal of toxic heavy metals from aquatic environment: A review. *J. Environ. Chem. Eng.* 2017; 5(3):2782-2799. <https://doi.org/10.1016/j.jece.2017.05.029>.
- [36] Fu F, Wang Q. Removal of heavy metal ions from wastewaters: a review. *J Environ Manage.* 2011;92(3):407-418. <https://doi.org/10.1016/j.jenvman.2010.11.011>.
- [37] Ayach J, El Malti W, Duma L, Lalevée J, Al Ajami M, Hamad H, Hijazi A. Comparing Conventional and Advanced Approaches for Heavy Metal Removal in Wastewater Treatment: An In-Depth Review Emphasizing Filter-Based Strategies. *Polymers.* 2024; 16(14):1959. <https://doi.org/10.3390/polym16141959>.
- [38] Barman MK, Bhattarai A, Saha B. Applications of ion exchange resins in environmental remediation. *Vietnam J. Chem.*, 2023, 61(5), 533-550. <https://doi.org/10.1002/vjch.202300027>.
- [39] Rath BS, Kumar PS. Application of adsorption process for effective removal of emerging contaminants from water and wastewater. *Environ. Pollut.* 2021; 280: 116995. <https://doi.org/10.1016/j.envpol.2021.116995>.

- [40] Younas M, Rezakazemi M. Chapter 1 - Introduction to Membrane Technology. Book Editor(s): Mashallah Rezakazemi, Mohammad Younas. Wiley-VCH GmbH. 2022; p.1-16. <https://doi.org/10.1002/9783527831036.ch1>.
- [41] Qasem NAA, Mohammed RH, Lawa DU. Removal of heavy metal ions from wastewater: a comprehensive and critical review. *npj Clean Water* (2021) 4: 36; <https://doi.org/10.1038/s41545-021-00127-0>.
- [42] Ahmad A, Ghazi ZA, Saeed M, Ilyas M, Ahmad R, Khattak AM, Iqbal A. A comparative study of the removal of Cr(VI) from synthetic solution using natural biosorbents. *New J. Chem.* 2017; 41:10799-10807. DOI: 10.1039/c7nj02026k.
- [43] Moradi G, Zinadini S, Rajabi L, Derakhshan AA. Removal of heavy metal ions using a new high performance nanofiltration membrane modified with curcumin boehmite nanoparticles. *Chem. Eng. J.* 2020; 390. <https://doi.org/10.1016/j.cej.2020.124546>.
- [44] Hemdan B, Garlapati VK, Sharma S, Bhadra S, Maddirala S, Varsha KM, Motru V, Goswami P, Sevda S, Aminabhavi TM. Bioelectrochemical systems-based metal recovery: resource, conservation and recycling of metallic industrial effluents. *Environ. Res.* 2022; 204(Part D):112346. <https://doi.org/10.1016/j.envres.2021.112346>.
- [45] Kikuchi T, Tanaka S. Biological removal and recovery of toxic heavy metals in water environment. *Crit. Rev. Environ. Sci. Technol.* 2012; 42(10):1007-1057. <http://dx.doi.org/10.1080/10643389.2011.651343>.
- [46] Vidu R, Matei E, Predescu AM, Alhalaili B, Pantilimon C, Tarcea C, Predescu C. Removal of Heavy Metals from Wastewaters: A Challenge from Current Treatment Methods to Nanotechnology Applications. *Toxics.* 2020; 8(4):101. <https://doi.org/10.3390/toxics8040101>.
- [47] Benalia, M.C., Youcef, L., Bouaziz, M.G. et al. Removal of Heavy Metals from Industrial Wastewater by Chemical Precipitation: Mechanisms and Sludge Characterization. *Arab J Sci Eng* 47, 5587–5599 (2022). <https://doi.org/10.1007/s13369-021-05525-7>.
- [48] Bashir, A., Malik, L.A., Ahad, S. et al. Removal of heavy metal ions from aqueous system by ion-exchange and biosorption methods. *Environ Chem Lett* 17, 729–754 (2019). <https://doi.org/10.1007/s10311-018-00828-y>.
- [49] Rajendran S, Priya AK, Kumar PS, Hoang TKA, Sekar K, Chong KY, Khoo KS, Ng HS, Show PL. A critical and recent developments on adsorption technique for removal of heavy metals from wastewater-A review. *Chemosphere.* 2022; 303(2): 135146. <https://doi.org/10.1016/j.chemosphere.2022.135146>.
- [50] Malaeb L, Ayoub GM. Reverse osmosis technology for water treatment: State of the art review. *Desalination.* 2011; 267(1):1-8. <https://doi.org/10.1016/j.desal.2010.09.001>.
- [51] Mahmoud AED, Mostafa E. Nanofiltration Membranes for the Removal of Heavy Metals from Aqueous Solutions: Preparations and Applications. *Membranes.* 2023; 13(9):789. <https://doi.org/10.3390/membranes13090789>.
- [52] Yaqub M, Lee SH. Heavy metals removal from aqueous solution through micellar enhanced ultrafiltration: A review [Internet]. Vol. 24, Environmental Engineering Research. Korean Society of Environmental Engineering; 2018. p. 363–75. Available from: <http://dx.doi.org/10.4491/eer.2018.249>.
- [53] Hinchliffe A, Porter K. A Comparison of Membrane Separation and Distillation. *Chem. Eng. Res. Des.* 2000; 78(2): 255-268. Doi:10.1205/026387600527121.
- [54] Zhang C. Absorption principle and techno-economic analysis of CO<sub>2</sub> absorption technologies: A review. *IOP Conf. Ser. Earth Environ. Sci.* 2021; 657(1): 012045. DOI:10.1088/1755-1315/657/1/012045.
- [55] Soniya M, Muthuraman G. Comparative study between liquid–liquid extraction and bulk liquid membrane for the removal and recovery of methylene blue from wastewater. *J. Ind. Eng. Chem.* 2015; 30. DOI: 10.1016/j.jiec.2015.05.032.
- [56] Mpongwana N, Rathilal S. A Review of the Techno-Economic Feasibility of Nanoparticle Application for Wastewater Treatment. *Water.* 2022; 14: 1550. DOI: 10.3390/w14101550.
- [57] Baker, R.W. Membrane Technology and Applications. 2<sup>nd</sup> Edition, John Wiley and Sons, Ltd., Hoboken. 2004. <https://doi.org/10.1002/0470020393>.
- [58] da Silva Biron D, dos Santos V, Zeni M. Overview Membranes Separations. In: Ceramic Membranes Applied in Separation Processes. Topics in Mining, Metallurgy and Materials Engineering. Springer, Cham. 2018; p. 7-29. [https://doi.org/10.1007/978-3-319-58604-5\\_2](https://doi.org/10.1007/978-3-319-58604-5_2).
- [59] Krishnan S, Zulkapli NS, Kamyab H, Taib SM, Din MFBM, Abd Majid Z, Chaiprapat S, Kenzo I, Ichikawa Y, Nasrullah M, Chelliapan S, Othman N. Current technologies for recovery of metals from industrial wastes: An overview. *Environ. Technol. Inno.* 2021; 22:101525. <https://doi.org/10.1016/j.eti.2021.101525>.
- [60] Issaoui M, Limousy L. Low-cost ceramic membranes: Synthesis, classifications, and applications. *C. R. Chimie.* 2019; 22:175-187. <https://doi.org/10.1016/j.crci.2018.09.014>.
- [61] El Batouti M, Al-Harby NF, Elewa MM. A Review on Promising Membrane Technology Approaches for Heavy Metal Removal from Water and Wastewater to Solve Water Crisis. *Water.* 2021; 13(22):3241. <https://doi.org/10.3390/w13223241>.
- [62] Sandu T, Sârbu A, Căprărescu S, Stoica E-B, Iordache T-V, Chiriac A-L. Polymer Membranes as Innovative Means of Quality Restoring for Wastewater Bearing Heavy Metals. *Membranes.* 2022; 12(12):1179. <https://doi.org/10.3390/membranes12121179>.
- [63] Xiang H, Min X, Tang C, Sillanpää M, Zhao F. Recent advances in membrane filtration for heavy metal removal from wastewater: A mini review. *J. Water Process Eng.* 2022; 49: 103023. <https://doi.org/10.1016/j.jwpe.2022.103023>.



- [64] Qu F, Liang H, Zhou J, Nan J, Shao S, Zhang J, Li G. Ultrafiltration membrane fouling caused by extracellular organic matter (EOM) from *Microcystis aeruginosa*: Effects of membrane pore size and surface hydrophobicity. *J. Membr. Sci.* 2014; 449:58-66. <http://dx.doi.org/10.1016/j.memsci.2013.07.070>.
- [65] Banerjee P, Dey TK, Sarkar S, Swarnakar S, Mukhopadhyay A, Ghosh S. Treatment of cosmetic effluent in different configurations of ceramic UF membrane-based bioreactor: toxicity evaluation of the untreated and treated wastewater using catfish (*Heteropneustes fossilis*). *Chemosphere.* 2016; 146:133-144. <https://doi.org/10.1016/j.chemosphere.2015.12.004>.
- [66] Krüger R, Vial D, Arifin D, Weber M, Heijnen M. Novel ultrafiltration membranes from low-fouling copolymers for RO pretreatment applications. *Desalination. Water Treat.* 2016; 57:23185–23195. <https://doi.org/10.1080/19443994.2016.1153906>.
- [67] Hu Z, Wen X, Si X. Pre-ultrafiltration or pre-ozonation for EDCs removal in a combined ultrafiltration and ozonation process. *J. Chem. Technol. Biotechnol.* 2016; 91(12):2929-2934. DOI 10.1002/jctb.4907.
- [68] Xu YC, Wang ZX, Cheng XQ, Xiao YC, Shao L. Positively charged nanofiltration membranes via economically mussel-substance-simulated co-deposition for textile wastewater treatment. *Chem. Eng. J.* 2016; 303:555-564. <http://dx.doi.org/10.1016/j.cej.2016.06.024>.
- [69] Speth TF, Summers RS, Gusses AM. Nanofiltration foulants from a treated surface water. *Environ. Sci. Technol.* 1998; 32:3612-3617. <https://doi.org/10.1021/es9800434>.
- [70] Van de Lisdonk CAC, Van Paassen JAM, Schippers JC. Monitoring scaling in nanofiltration and reverse osmosis membrane systems. *Desalination.* 2000; 132:101-108. [https://doi.org/10.1016/S0011-9164\(00\)00139-9](https://doi.org/10.1016/S0011-9164(00)00139-9).
- [71] Mo H, Ng HY. An experimental study on the effect of spacer on concentration polarization in a long channel reverse osmosis membrane cell. *Water Sci. Technol.* 2010; 61(8):2035-2041. doi:10.2166/wst.2010.116.
- [72] Torki M, Nazari N. Evaluation of biological fouling of RO/MF membrane and methods to prevent it. *Eur. J. Adv. Eng. Technol.* 2017; 4(9):707-710.
- [73] Lee KP, Arnot D, Mattia D. A review of reverse osmosis membrane materials for desalination-development to date and future potential. *J. Membr. Sci.* 2011; 370(1-2):1-22. <https://doi.org/10.1016/j.memsci.2010.12.036>.
- [74] Su X, Li W, Palazzolo A, Ahmed S. Concentration polarization and permeate flux variation in a vibration enhanced reverse osmosis membrane module. *Desalination.* 2018; 433:75-88. <https://doi.org/10.1016/j.desal.2018.01.001>.
- [75] Kumar RV, Pugazhenth G. Removal of chromium from synthetic wastewater using MFI zeolite membrane supported on inexpensive tubular ceramic substrate. *J. Water Reuse Desalination.* 2017; 7(3):365-377. doi: 10.2166/wrd.2016.096.
- [76] Choudhury PR, Majumdar S, Sahoo GC, Saha S, Mondal P. High pressure ultrafiltration CuO/hydroxyethyl cellulose composite ceramic membrane for separation of Cr(VI) and Pb(II) from contaminated water. *Chem. Eng. J.* 2018a; 336:570-578. <https://doi.org/10.1016/j.cej.2017.12.062>.
- [77] Choudhury P, Mondal P, Majumdar S, Saha S, Sahoo GC. Preparation of ceramic ultrafiltration membrane using green synthesized CuO nanoparticles for chromium (VI) removal and optimization by response surface methodology. *J. Clean. Prod.* 2018b; 203:511-520. <https://doi.org/10.1016/j.jclepro.2018.08.289>.
- [78] Adam MR, Salleh NM, Othman MHD, Matsuura T, Ali MH, Puteh MH, Ismail AF, Rahman MA, Jaafar J. The adsorptive removal of chromium (VI) in aqueous solution by novel natural zeolite based hollow fibre ceramic membrane. *J. Environ. Manage.* 2018; 224:252-262. <https://doi.org/10.1016/j.jenvman.2018.07.043>.
- [79] Hubadillah SK, Othman MHD, Tai ZS, Jamalludin MR, Yusuf NK, Ahmad A, Rahman MA, Jaafar J, Abdul Kadir SHS, Harun Z. Novel hydroxyapatite-based bio-ceramic hollow fiber membrane derived from waste cow bone for textile wastewater treatment. *Chem. Eng. J.* 2020; 379. <https://doi.org/10.1016/j.cej.2019.122396>.
- [80] Chaukura N, Katengeza G, Gwenzi W, Mbiriri CI, Nkambule TTI, Moyo M, Kuvarega AT. Development and evaluation of a low-cost ceramic filter for the removal of methyl orange, hexavalent chromium, and *Escherichia coli* from water. *Mater. Chem. Phys.* 2020; 249:122965. <https://doi.org/10.1016/j.matchemphys.2020.122965>.
- [81] Aloulou W, Aloulou H, Khemakhem M, Duplay J, Daramola MO, Ben Amar R. Synthesis and characterization of clay-based ultrafiltration membranes supported on natural zeolite for removal of heavy metals from wastewater. *Environ. Technol. Innov.* 2020; 18: 100794. <https://doi.org/10.1016/j.eti.2020.100794>.
- [82] Roy S, Majumdar S, Sahoo GC, Bhowmick S, Kundu AK, Mondal P. Removal of As(V), Cr(VI) and Cu(II) using novel amine functionalized composite nanofiltration membranes fabricated on ceramic tubular substrate. *J. Hazard. Mater.* 2020; 399:122841. <https://doi.org/10.1016/j.jhazmat.2020.122841>.
- [83] Ganji R, Peyravi M, Khalili S, Jahanshahi M. Bilayer adsorptive ceramic membranes supported cage-like mesoporous silica for hexavalent chromium removal: Experimental and DFT studies. *Process Saf. Environ. Prot.* 2020; 143:45-54. <https://doi.org/10.1016/j.psep.2020.06.004>.
- [84] Zavala MAL, Romero-Santana H, Mon´arrez-Cordero BE. Removal of Cr(VI) from water by adsorption using low-cost clay-perlite-iron membranes. *J. Water Process. Eng.* 2020; 38:101672. <https://doi.org/10.1016/j.jwpe.2020.101672>.
- [85] Dizge N, Ozay Y, Boussemghoune M, Chikhi M. Preparation of catalytic polyether sulfone coated ceramic membrane for reduction of hexavalent chromium. *J. Water Process. Eng.* 2021; 40:101946. <https://doi.org/10.1016/j.jwpe.2021.101946>.
- [86] Zhang J, Yan M, Sun G, Liu K. Simultaneous removal of Cu(II), Cd(II), Cr(VI), and Rhodamine B in wastewater using TiO<sub>2</sub> nanofibers membrane loaded on porous fly ash ceramic support. *Sep. Purif. Technol.* 2021; 272:118888. <https://doi.org/10.1016/j.seppur.2021.118888>.

- [87] Roy S, Bhowmick K, Singh P, Bhowmick S, Mukherjee M, Majumdar S, Sahoo GC, Mondal P. Removal of heavy metals by surface tailored copper ion enhanced ceramic-supported-polymeric composite nanofiltration membrane. *J. Environ. Chem. Eng.* 2021; 9:106368. <https://doi.org/10.1016/j.jece.2021.106368>.
- [88] Bat-Amgalan M, Miyamoto N, Kano N, Yunden G, Kim HJ. Preparation and Characterization of Low-Cost Ceramic Membrane Coated with Chitosan: Application to the Ultrafine Filtration of Cr(VI). *Membranes*. 2022; 12:835. <https://doi.org/10.3390/membranes12090835>.
- [89] Pranoto, Inayati, Firmansyah F. Effectiveness Study of Drinking Water Treatment Using Clays/Andisol Adsorbent in Lariat Heavy Metal Cadmium (Cd) and Bacterial Pathogens. *IOP Conf. Ser.: Mater. Sci. Eng.* 2018; 349. doi:10.1088/1757-899X/349/1/012047.
- [90] Yusof MSM, Othman MHD, Mustafa A, Abdul Rahman M, Jaafar J, Ismail AF, Feasibility study of cadmium adsorption by palm oil fuel ash (POFA)-based low-cost hollow fibre zeolitic membrane. *Environ. Sci. Pollut. Res.* 2018; 25:21644-21655. <https://doi.org/10.1007/s11356-018-2286-6>.
- [91] Shahzad HK, Hussein MA, Patel F, Al-Aqeeli N, Atieh MA, Laoui T. Synthesis and characterization of alumina-CNT membrane for cadmium removal from aqueous solution. *Ceram. Int.* 2018; 44:7189-7198. <https://doi.org/10.1016/j.ceramint.2018.06.175>.
- [92] Hussein MA, Shahzad HK, Patel F, Atieh MA, Al-Aqeeli N, Baroud TN, Laoui T. Porous Al<sub>2</sub>O<sub>3</sub>-CNT Nanocomposite Membrane Produced by Spark Plasma Sintering with Tailored Microstructure and Properties for Water Treatment. *Nanomaterials*. 2020; 10:845. doi:10.3390/nano10050845.
- [93] Bhowmick K, Roy S, Mukherjee M, Sahoo GC, Majumdar S, Mondal P. Removal of cadmium by in-situ Cu nanoparticle enhanced ceramic supported-polymeric composite NF membrane. *Mater. Today: Proc.* 2021; 47:1496-1499. <https://doi.org/10.1016/j.matpr.2021.04.398>.
- [94] Lahnaifi A, Elgamouz A, Tijani N, Jaber L, Kawde A. Hydrothermal synthesis and electrochemical characterization of novel zeolite membranes supported on flat porous clay-based microfiltration system and its application of heavy metals removal of synthetic wastewaters. *Microporous Mesoporous Mater.* 2022; 334. <https://doi.org/10.1016/j.micromeso.2022.111778>.
- [95] Fadila CR, Othman MHD, Adam MR, Takagi R, Yoshioka T, Khongnakorn W, Rahman MA, Jaafar J, Ismail AF. Adsorptive membrane for heavy metal removal: Material, fabrication, and performance. *Mater. Today: Proc.* 2022; 65(7):3037-3045. <https://doi.org/10.1016/j.matpr.2022.03.582>.
- [96] Sisnayati R, Komala R, Suryani. The Effect of Rice Husk Addition as Additive Materials on the Characterization of Ceramic Membrane and Their Application on Water River Treatment Process. *Sci. Technol. Indones.* 2019; 4(1). <https://doi.org/10.26554/sti.2019.4.1.11-17>.
- [97] Jiménez-Castañeda ME, Medina DI. Use of Surfactant-Modified Zeolites and Clays for the Removal of Heavy Metals from Water. *Water*. 2017; 9(4):235. <https://doi.org/10.3390/w9040235>.
- [98] Hubadillah SK, Othman MHD, Harun Z, Ismail AF, Rahman MA, Jaafar J. A novel green ceramic hollow fiber membrane (CHFM) derived from rice husk ash as combined adsorbent-separator for efficient heavy metals removal. *Ceram. Int.* 2017; 43:4716-4720. <http://dx.doi.org/10.1016/j.ceramint.2016.12.122>.
- [99] Innocenzi V, Prisciandaro M, Tortora F, Di Celso GM, Vegliò F. Treatment of WEEE industrial wastewaters: Removal of yttrium and zinc by means of micellar enhanced ultrafiltration. *Waste Manage.* 2018; 74:393-403. <https://doi.org/10.1016/j.wasman.2017.12.018>.
- [100] Li P, Li YX, Wu YZ, Xu ZL, Zhang HZ, Gao P, Xu SJ. Thin-film nanocomposite NF membrane with GO on macroporous hollow fiber ceramic substrate for efficient heavy metals removal. *Environ. Res.* 2021; 197:111040. <https://doi.org/10.1016/j.envres.2021.111040>.
- [101] Foorginezhad S, Zerafat MM, Mohammadi Y, Asadnia M. Fabrication of tubular ceramic membranes as low-cost adsorbent using natural clay for heavy metals removal. *Clean. Eng. Technol.* 2022; 10:100550. <https://doi.org/10.1016/j.clet.2022.100550>.
- [102] Hubadillah SK, Othman MHD, Abdul Kadir SHS, Jamalludin MR, Harun Z, Abd Aziz MH, Rahman MA, Jaafar J, Nomura M, Honda S, Iwamoto Y, Fansuri H. Removal of As(III) and As(V) from water using green, silica-based ceramic hollow fibre membranes via direct contact membrane distillation. *RSC Adv.* 2019; 9:3367-3376. <https://doi.org/10.1039/C8RA08143C>.
- [103] Zavala MAL, Bouchez BF. Montmorillonite-perlite-iron ceramic membranes for the adsorption/ removal of As(III) and other constituents from surface water. *Ceram. Int.* 2022; 48(21):31695-31704. <https://doi.org/10.1016/j.ceramint.2022.07.091>.
- [104] Muhamad N, Abdullah N, Rahman MA, Abas KH, Aziz AA, Othman MHD, Jaafar J, Ismail AF. Removal of nickel from aqueous solution using supported Zeolite-Y hollow fiber membranes. *Environ. Sci. Pollut. Res.* 2018; 25:19054-19064. <https://doi.org/10.1007/s11356-018-2074-3>.
- [105] Zaidan NM, Ahmad N, Nakamura Y, Abubakar M. Sayong Ball Clay Membrane for Copper and Nickel Removal from Effluent. *J. Adv. Res. Fluid Mech. Therm. Sci.* 2021; 79(2):131-138. <https://doi.org/10.37934/arfm.79.2.131138>.
- [106] Chaghomi B, Lotfollahi MN. Hybrid ceramic membrane microfiltration and electrolysis process for Cu-removal from aqueous solutions. *Ceram. Int.* 2022; 48(20):29967-29976. <https://doi.org/10.1016/j.ceramint.2022.06.264>.
- [107] Choudhury PR, Majumdar S, Sarkar S, Kundu B, Sahoo GC. Performance investigation of Pb(II) removal by synthesized hydroxyapatite based ceramic ultrafiltration membrane: Bench scale study. *Chem. Eng. J.* 2019; 355:510-519. <https://doi.org/10.1016/j.cej.2018.07.155>.

- 
- [108] Masturi, Siqid AL, Susilawati, Edie SS, Widodo RD, Alighiri D. The effect of thermal activation for natural zeolite on the performance of ceramic membrane as  $Pb^{2+}$  ion adsorption. *J. Phys.: Conf. Ser.* 2019; 1170. doi:10.1088/1742-6596/1170/1/012055.
- [109] AlSawaftah N, Abuwatfa W, Darwish N, Hussein G. A Comprehensive Review on Membrane Fouling: Mathematical Modelling, Prediction, Diagnosis, and Mitigation. *Water*. 2021; 13(9):1327. <https://doi.org/10.3390/w13091327>.
- [110] Bokhary, A., Tikka, A., Leitch, M., Liao, B. Membrane Fouling Prevention and Control Strategies in Pulp and Paper Industry Applications: A Review. *Journal of Membrane Science and Research*, 2018; 4(4): 181-197. <https://doi.org/10.22079/jmsr.2018.83337.1185>.
- [111] Morales N, Mery-Araya C, Guerra P, Poblete R, Chacana-Olivares J. Mitigation of Membrane Fouling in Membrane Bioreactors Using Granular and Powdered Activated Carbon: An Experimental Study. *Water*. 2024; 16(17):2556. <https://doi.org/10.3390/w16172556>.
- [112] Du X, Shi Y, Jegatheesan V, Haq IU. A Review on the Mechanism, Impacts and Control Methods of Membrane Fouling in MBR System. *Membranes*. 2020; 10(2):24. <https://doi.org/10.3390/membranes10020024>.