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Overview of Iron-Coated Dynamic Membrane for Water Treatment

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Abstract: The increasing global demand for clean and potable water has prompted the exploration of innovative water treatment technologies. Pre-deposited dynamic membrane systems, a novel approach in wastewater treatment, have gained attention due to their versatility and effectiveness. This mini-review focuses on the application of iron oxide-based dynamic membranes in water treatment processes. It discusses the impact, formation, properties, and various water treatment applications of iron oxide dynamic membranes, highlighting their potential to revolutionize the field of sustainable water treatment.

Keywords: Coating; Iron; Porous layer; pre-deposited dynamic membrane; primary membrane.

Introduction

Water stands as one of life's most fundamental necessities, critical for drinking, food processing, and sanitation. Unfortunately, water isn't consistently accessible or affordable for everyone. Over 40% of the global population faces water shortages, and more than 700 million individuals lack access to quality drinking water (Shemer et al., 2023). The conventional method of water treatment, involving coagulation and flocculation, relies on chemical usage (Othman et al., 2021). Consequently, membrane technology emerges as a highly favored alternative due to its numerous advantages, including eco-friendliness, low capital costs, compact facility size, simplicity of operation, and energy efficiency (Gao, 2016). Additionally, the small pore sizes of the membrane facilitate the attainment of high-quality permeate. Fouling, characterized by pore blockage and/or the deposition of contaminants on membrane surfaces, poses a significant challenge to membrane applications. This issue leads to reduced flux, efficiency, and the overall longevity of the membrane system. To address this concern, various approaches are employed, including membrane patterning, surface grafting, and modification. Dynamic membrane technology is one of the membrane modification techniques used to generate a secondary filtration layer to mitigate fouling while concurrently enhancing permeate quality.

Dynamic membranes (DM) arise through the accumulation of substances on a primary membrane, resulting in improved permeate quality (Anantharaman et al., 2020). The benefits of utilizing dynamic membranes in water treatment to enhance membrane separation efficiency include cost-effectiveness, achieved through the use of economical materials for the primary membrane or porous support layer, as well as integrating feed constituents as secondary layer components. Based on the method through which the secondary layer is developed, dynamic membranes can be divided into two: self-forming and pre-deposited dynamic membranes.

The first category emanates from the deposition of feed constituents, leading to their subsequent

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agglomeration into a cake or gel layer. On the other hand, pre-deposited dynamic membranes arise from the intentional coating of particles onto the membrane surface. Iron-based pre-deposited dynamic membranes offer a versatile solution, harnessing the advantages of dynamic membrane technology and the unique properties of iron-based materials, such as simplistic application, low cost, high surface area, and the suppression of virus activities in aquatic environments, among others. By coating membranes with iron, the system effectively targets a variety of contaminants, including organic pollutants and heavy metals, thereby contributing to improved water quality (Jabbar et al., 2022). The primary objective of this review is to present the role of iron in pre-deposited dynamic membrane water treatment. This will be achieved by delving into their composition, fabrication methods, applications, and performance outcomes. Through this exploration, the review aims to present advancements and insights into iron's function while identifying its future trajectory in the realm of pre-deposited membranes.

Method

This research uses literature study methods in various international journals which discuss the potential of iron-coated dynamic membranes.

Results and Discussion

Iron in Water Treatment

Iron oxide has emerged as a promising and versatile material for water treatment, particularly in the context of wastewater treatment (Navratil, 1999). Iron oxide (FeO), in addition to its low cost (Jabbar et al., 2022), its unique properties make it well-suited for various applications in addressing the challenges of water pollution and scarcity. The adsorptive properties of iron oxide facilitate efficient wastewater treatment processes, enabling the removal of a variety of contaminants such as heavy metals (Mishal et al., 2019), (Kumari et al., 2019) and organic pollutants (Ge et al., 2011), among others. The positively charged surface of iron oxide particles makes it engage with negatively charged particles present in water, leading to the aggregation of these particles into larger masses. This characteristic renders iron oxide an effective coagulant. A noteworthy example of this capability is its implementation in surface water treatment, as demonstrated by Shabani et al. (Shabani et al., 2021), in which a remarkable decrease in turbidity was reported. When used in water treatment, iron oxide demonstrates enhanced effectiveness when its particle size is reduced to the nano-scale range for high surface area which affords high contaminants adsorption (Jabbar et al., 2022). This modification increases its surface area significantly, resulting in higher performance. This makes the use of iron oxide nanoparticles in water treatment attractive. Iron oxide nanoparticles can be synthesized through three main routes: physical, chemical, and biological methods. Additionally, these routes can be further categorized into specific techniques, as listed in Table 1. Each of the diverse techniques for producing iron oxide NPs has its own set of benefits and drawbacks (Aragaw et al., 2021).

Table 1. Synthesis Methods of Iron Nanoparticles

Physical methods	Chemical methods	Biological methods
Aerosol	Coprecipitation	Bacteria mediated
Electron beam lithography	Electrochemical decomposition	Fungi mediated
Gas phase deposition	Hydrothermal	Plant mediated
Laser-induced pyrolysis	Microemulsion	Protein mediated
Power ball milling	Sonochemical	
Pulsed laser ablation	Thermal decomposition	
Sol-gel		

(Aragaw et al., 2021**;** Ali et al., 2016)

The various approaches employed in the synthesis of iron oxide nanoparticles are intriguing. Nevertheless, embracing a green production method that utilizes plant-based extracts brings forth numerous advantages in addition to sustainability. Jabbar et al. (Jabbar et al., 2022) reported that green-synthesized iron oxide exhibits a notable tendency towards higher purity compared to counterparts produced using alternative methods. Furthermore, the process is characterized by simplicity, safety, and swift synthesis duration.

Iron oxide exhibits an astounding capacity for facile functionalization with a diverse array of additives tailored for the removal of targeted contaminants. This inherent flexibility is not only appealing but also held in high regard, as it imparts adaptability to its application in water treatment, aligning seamlessly with the dynamic nature of waterborne pollutants. For instance, Singh et al. (Singh et al., 2020) highlighted the utilization of alkali cations to functionalize iron oxide NPs, enabling the treatment of textile and pharmaceutical wastewater.

Dynamic Membrane Technology

The use of dynamic membranes (DMs) in water filtration and wastewater treatment has attracted growing interest in recent times (Malczewska, 2021), as they offer several benefits, including an alternative means of minimizing the costs associated with the application of membranes in water treatment (Li et al., 2018). In addition to cost-effectiveness, DMs are favored for water treatment for several reasons. These include high flux, low transmembrane pressure (TMP), suitability for operation under gravity, and seamless cleaning through simple jet rinsing, brushing, water backwashing, or a combination of more than one of these methods (Pollice & Vergine, 2020; Li et al., 2018).

A dynamic membrane is a semipermeable layer (secondary membrane) made of solutes on the surface of a conventional membrane, often referred to as the primary membrane (PM), which serves as an interface between the influent and the PM. The DM provides a barrier between the influent solutes and the PM, thereby permeating only the purified water. The PM is usually a porous membrane or low-cost support (such as non-woven materials or stainless-steel mesh) (Lv et al., 2022) that may otherwise be unable to effectively reject low molecular weight solutes without the presence of a secondary film. DM is known to offer resistance to flow on the membrane surface. This action hinders the movement of solutes through the membrane, promoting physical separation from the raw water in addition to low membrane fouling propensity.

DMs can be classified as pre-coated and self-forming (Ersahin et al., 2012). Pre-deposited dynamic membranes (DMs) are created by applying a deliberate layer of coating additive(s) onto the membrane surface. The pre-coated DMs are further divided into two categories: single additive membranes, created by the single-step deposition of one material, and composite (bi-layer) membranes, formed by a two-step coating of composites on the PMs. Unlike pre-coated DMs, self-formed DMs (SFDMs) do not require external materials. Instead, the rejected solutes aggregate and generate the secondary membrane, hence decreasing permeate volume but improving permeate quality (Hu et al., 2018).

Iron-Containing Dynamic Membranes

Iron-containing materials are employed either as standalone compounds or in conjunction with other substances to create a secondary layer with the desired characteristics on a primary membrane. This can enhance contaminant removal, increase flux, mitigate fouling, and induce resistance, whether thermal or chemical or a combination of two or more of these effects. The iron-containing material used for generating a secondary membrane could potentially be a compound other than iron oxides, such as iron hydroxide (Prabowo, 2015), iron chloride, and more (El Batouti et al., 2021). Various techniques as well are employed to facilitate the formation of the dynamic layer. These encompass methods such as immersion (Armendáriz-Ontiveros et al., 2020), dip-coating (Kyesmen et al., 2021; Parshetti & Doong, 2012), permeation drag (Usman et al., 2021; Raciny et al., 2011) and interfacial polymerization (IP) (Al-Hobaib et al., 2016), among others.

Dip-coating involves immersing the membrane into a solution of the iron-based compound. This process leads to the particles adhering to the membrane surface (Kyesmen et al., 2021). The excess solution is removed afterward. Permeation drags, also known as filter press or simply filtration, involves passing a solution of the iron-based compound through a membrane (Usman et al., 2021). This process causes the iron particles to be carried onto the membrane surface, where they adhere and gradually form a secondary layer.

Lakhotia et al. (Lakhotia et al., 2019) reported the incorporation of iron oxide into the creation of a secondary layer on a PES membrane through the immersion method. In essence, the PES membrane underwent a coating process involving immersion in a mixture of sodium lauryl sulfate (SLS) and m-phenylenediamine (MPD). The resulting supporting layer was subsequently coated with a solution of pre-assembled FeO NPs which facilitated interfacial polymerization at the surface. This technique had been previously reported by Al-Hobaib et al., (2016) in which a porous polyamide membrane was the supported layer.

In brief interfacial polymerization is a form of step-growth polymerization occurring at the boundary between two phases that don't mix, granting distinctive chemical and topological characteristics to the resulting polymer materials (Song et al., 2017). Among the various methods used to create a dynamic layer on the membrane surface, interfacial polymerization stands out for its exceptional success and is thus more commonly employed (Rabajczyk et al., 2021). Irrespective of the iron deposition method employed, the formation of a uniform layer significantly enhances the effectiveness of a dynamic membrane (Prabowo, 2015). Additionally, the sizes of the iron particles used in creating the dynamic membrane also play a role in the efficient removal of the targeted contaminants.

Performance and Characteristics of Iron Oxide Dynamic Membranes

The inclusion of an iron-containing layer offers membranes a range of advantages. These include improved rejection of contaminants, the ability to resist fouling, heightened strength, and other favorable attributes. These benefits have been extensively narrated by numerous researchers dedicated to achieving cleaner water and a safer environment. Their efforts focus on the efficient removal of a wide array of water contaminants from different feeds using dynamic membranes. Armendáriz-Ontiveros (2020) and other researchers conducted a study where they employed an Iron NPs layer on polysulfone, the primary membrane, to counteract biofouling during the desalination of actual seawater. The biocidal effect of the dynamic layer, formed using the immersion method, was evident in the process.

Homayoonfal et al. (2014) demonstrated that the deposition of an iron oxide layer on a polysulfone membrane led to a significant enhancement in dye rejection compared to the in situ modified polysulfone membrane. While the authors did recognize the superior strength of the blended modified membrane, it's important to note that this strength difference isn't a concern as long as the surface-coated membrane retains the necessary strength to endure operational pressures. Otherwise, it could be deemed suitable for low-pressure applications. Parshetti & Doong (2012) reported the sequential deposition of iron and nickel layers by di-coating onto three distinct membranes; PVDF, cellulose esters, and nylon-66. This process was aimed at facilitating the dechlorination of chlorinated hydrocarbons. Notably, the study revealed that the formation of a uniform layer on the nylon-66 membrane is more pronounced compared to the other two membranes and attributed it to the hydrophilic nature of the nylon-66 membrane. This uniform layer's formation manifested in the greater performance of the membrane, which is higher than that of the counterpart membranes. In their comprehensive study, Al-Hobaib et al., (2016) explored the impact of an iron oxide layer on a polyamide membrane and its consequential effects on the purification of groundwater. The authors made a noteworthy revelation as they managed to uphold an impressive 98% salt rejection rate while concurrently elevating the flux from its initial value of 26 L/m² h to a significantly improved 44 L/m² h.

Gui et al., (2015) reported the creation of an iron-containing polyacrylic acid layer on a PVDF membrane. This infusion of an iron layer onto the membrane led to its remarkable proficiency in efficiently eliminating selenium contaminants. Tian et al., (2018) reported a compelling approach involving the utilization of ferric oxide to mitigate fouling susceptibility. The deposition of the iron layer was facilitated by the humic acid attached to the PVDF membrane through permeation drag. The remarkable outcome was a significant enhancement in both the membrane's antifouling properties and its flux, marking a truly impressive advancement.

Table 2. Some Notable Iron-containing DM Application in Water Treatment

DM	PM	Method	Feed water	Cell orientation	Contaminants rejection (%)	Reference
TA-Fe^{3+}	PES	IP	Salt water	Cross-flow	98.30	(Xiao et al., 2022)
$Fe(OH)_2$	PES	Filtration	Wastewater	Dead-end	High Arsenic removal	(Usman et al., 2021)
TA–Fe^{3+}	PAN	Dip-coating	Salt water	Cross-flow	90.20	(Liu et al., 2021)
FeNPs	Psf	Immersion	Seawater	Cross-flow	Biofouling resistant	(Armendáriz-Ontiveros et al., 2020)
FeNPs	NA	Immersion	Seawater	Cross-flow	92.00	(Armendáriz-Ontiveros et al., 2019)
Fe_3O_4 NPs	Psf	IP	Dye solution	Cross-flow	97.00	(Homayoonfal et al., 2014)
Ni/Fe NPs	Nylon-66	Dip-coating	chlorinated hydrocarbons	NA	13.70	(Parshetti & Doong, 2012)
Fe^{3+} /Ag^{+}/Zn^{2+}/Ni^{2+}/Zr^{4+}	PAN	Dip-coating	Dye solution	Dead-end	95.00	(You et al., 2019)
Fe_2O_3 NPs/PA	Psf	IP	Salt water	Cross-flow	98.00	(Al-Hobaib et al., 2016)
TA–Fe3+	PES	Immersion	Salt and dye containing	Dead-end	Salt: 62.1%	(Fan et al., 2015)

Table 2 illustrates several noteworthy studies that have reported the impact of an iron-containing secondary layer on removing contaminants from water. It's worth noting that not only valenced iron is appealing for dynamic membrane generation, but Zerovalent iron (ZVI) also demonstrates significant potential as a material for forming dynamic membranes. Its remarkable effectiveness in mitigating biofouling is noteworthy. To illustrate, a study conducted by Ma et al. (2015) stands out. This study sheds light on the deposition of a nanoscale zerovalent iron (NZVI) layer onto a PVDF UF membrane, which not only enhanced water flux but also improved the removal of humic acid. Intriguingly, the resulting membrane exhibited an amplified antifouling effect attributed to the higher dosage of zerovalent iron, leading to only a marginal decline in flux.

The dynamic membrane must uphold stability on both the primary membrane and amidst the iron particles within the layer, aiming to guarantee optimal and efficient performance. Employing a primary membrane functionalized with -COOH groups, whether naturally occurring or engineered, promotes strong adhesion of the iron layer onto the supporting membrane, while also facilitating the coordination of particles within the deposited layer. Enhanced: Tannic acid serves as an additive abundant in -COOH groups, facilitating the swift and smooth formation of an ironriched layer on membranes. This is attributed to its robust electrostatic and coordinative interactions (Kinfu & Rahman, 2023). Wu et al. (2019) reported the deposition of a steadfast iron layer with the assistance of TA. Furthermore, the resulting layer demonstrates hydrophilic and antifouling properties. Various polyphenols are utilized to bolster the adhesion of iron-based dynamic membranes across a range of water treatment applications that involve pre-deposited dynamic membranes, potentially extending the lifespan of these membranes. Therefore, it is essential to introduce anionic additives prior to depositing iron-based materials, especially when dealing with cationic ferric compounds like Fe^{2+} and Fe^{3+}.

The assessment of the effectiveness of dynamic membranes, including those made from iron-based materials, primarily revolves around two key factors: the uniform distribution across the membrane surface and its interaction with the pores within. These aspects directly govern the membrane's capacity to reject contaminants. Moreover, the robustness of the membrane can be evaluated by its responsiveness to backwashing procedures. Among the various techniques available, scanning electron microscope (SEM) provides unparalleled insights into the uniformity of the membrane on the PM surface, as well as its intricate interactions with the pores of the PM (Anantharaman et al., 2020). With transmission electron microscope (TEM), the composition of DM could be obtained (Sehasree Mohanta, 2021). Atomic Force Microscopy (AFM) makes the surface roughness of the membrane easier while the molecular bonds and the functional group of DMs determined using Fourier-Transform Infrared Spectroscopy (FTIR) (Bai et al., 2015). Acquiring information pertaining to the pore size distribution of dynamic membranes (DMs) holds crucial significance (Deng et al., 2008). This essential data can be effectively obtained through porosimetry analysis, a technique that plays a pivotal role in this characterization process. Employing X-ray spectroscopy on dynamic membranes (Zhang & Zhao, 2017) facilitates the examination of atomic interactions within the dynamic layer and their interplay with the PM.

Conclusion

The core focus of this review lies in exploring the utilization of iron-based compounds within pre-deposited dynamic membranes for water treatment. The surge in interest towards employing pre-deposited dynamic membranes for water treatment, with the goal of removing a wide array of contaminants, is evident. Leveraging the cost-effectiveness and availability of iron, its application within this domain continues to surge. This study not only illuminates the diverse forms of iron utilized in dynamic membranes but also highlights the various methodologies of deposition and coating. Moreover, it underscores the practical applications and performance benchmarks of membranes embellished with iron compounds. Upon dissecting a range of literary sources, it becomes apparent that for the successful deposition and optimal performance of iron-coated pre-deposited dynamic membranes, the primary membrane must be imbued with hydrophilicity through the application of hydrophilic additives. This strategic measure paves the way for the creation of a uniform layer, thus ensuring the attainment of high-quality permeate, increased antifouling propensity with little flux decline. Furthermore, it suggests that the size of the iron compounds is a crucial determinant of the membrane's capacity for contaminant removal. It is crucial to recognize that research into iron-coated dynamic membranes holds a promising future, especially when delving deeper into the stability of the iron-containing layer's coating. This involves considering factors such as the size of the deposited iron particles and the dependence of the layer's stability on the number of valency present within different iron particles, among other considerations.

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