

Recent Advances in Iron, Manganese and Ammonia Removal from Brackish Groundwater Using Membranes

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DOI:10.21608/PSERJ.2024.253459.1295

Received xx xxxx 202x

Revised xx xxxx 202x

Accepted xx xxxx 202x

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ABSTRACT

In the coming decades, there will be a significant issue with the availability of drinkable water as a result of population growth, present consumption patterns and climate change. This issue will have a similar social impact to rising energy prices. By far the most prevalent and easily accessible source of freshwater is groundwater, which is then followed by lakes, reservoirs, rivers, and wetlands. But it contains minimal amounts of microbiological or chemical pollutants. Simultaneously; There is always ammonia (NH_4^+), manganese (Mn^{2+}), and ferrous iron (Fe^{2+}) at the same time in the majority of anaerobic groundwater sources as a result of human activity and natural processes, presenting a severe risk to the security of supplies of drinking water. Problems including an unwanted taste, a brown color, pipeline obstructions, and a danger to public health because of the possibility of nervous system harm brought on by high concentrations of Fe^{2+} and Mn^{2+} in water sources. Consuming high levels of NH_4^+ is also linked to major health hazards for people, including ionic balance disruption in cells that could cause convulsions. Additionally, the transformation of NH_4^+ into cancer-causing trihalomethanes and organochlorines is possible. Various techniques have been developed to remove Fe^{2+} , Mn^{2+} and NH_4^+ from sources of drinking water. The use of an ultrafiltration membrane and its features were believed to have much potential to overcome many of the problems related to ammonia, manganese and iron contamination. Any kind of adsorptive, filtration, membrane bioreactor (MBR) process, and so forth, can be used in conjunction with ultrafiltration to remove the aforementioned contaminants.

Keywords : Brackish water; UF pretreatment; Iron-manganese removal; Ammonia removal

1 INTRODUCTION

One of the major issues facing the world today is water scarcity [1,2], which has led to the classification of many Middle Eastern, South East Asian, and North African nations as areas with a lack of water [3, 4]. Population increase and climate change put more strain on the supply of fresh water for human use [5]. The World Health Organization (WHO) estimates that 2.1 billion people worldwide lack access to clean drinking water, and through 2025, half of all people will live under water-stressed regions [6,7]. Techniques such as reusing water

and saltwater desalination has already been adopted to address the issue of the growing need for fresh water [8,9]. Fresh water from saline water is supplied by more than 15,000 desalination plants worldwide, and this number will only increase as researchers look for ways to improve the treatment procedures energy and cost-effectiveness [10]. Approximately 58% of the desalination capacity in the globe is currently located in the Middle East and North Africa, combined, and every day, 90 million m^3 of water are desalinated worldwide, according to the International Energy Agency (IEA) [11]. Over the past century, industrial water treatment has developed from a voluntary and elective procedure to an essential

multistage process that significantly improves the quality of drinking water. Following numerous experiments and setbacks, the technology has produced a series of clear treatment procedures that are widely used on a variety of raw waters [12]. Membrane technology is among the most advanced techniques for producing high-quality water [13, 14]. Reverse osmosis membrane technology, one of the most frequently utilized, is widely employed in brackish water treatment, drinking water production, wastewater treatment, and seawater desalination [15–17].

Brackish water, which is frequently used as a water source, may have trace amounts of microbiological or chemical pollutants [18,19]. Re phrase this paragraph Because of both natural and man-made processes, In the majority of anaerobic groundwater sources, NH_4^+ , Mn^{2+} , and Fe^{2+} ions are often found together, which effect a serious risk to the security sources of potable water [20]. Excessive levels of Mn^{2+} and Fe^{2+} in water sources causes bitterness, brown discoloration, obstructions in pipelines, and possible nervous system damage that could be harmful to the public health [20–22]. Overconsumption of NH_4^+ is linked to major health hazards for people as well, including upset of the cells ionic balance, which may cause convulsions. Furthermore, ammonia can be transformed into organochlorines and trihalomethanes, both of which are carcinogenic [20]. According to recommendations made by the World Health Organization (WHO), Drinking water should have no more than $300 \mu\text{g}\cdot\text{L}^{-1}$ of iron, $100\mu\text{g}\cdot\text{L}^{-1}$ of manganese, and $500 \mu\text{g}\cdot\text{L}^{-1}$ of ammonia. [21]. Natural groundwater often includes Mn and Fe in the most soluble forms possible, as divalent ions called Fe^{2+} and Mn^{2+} . When exposed to air, they oxidize and change from being colorless in soluble form to insoluble forms of Fe^{3+} and Mn^{4+} , respectively, leaving the water with a brown-red tint. Different methods for removing Fe^{2+} , Mn^{2+} , and NH_4^+ from groundwater sources have been developed to address these issues [20,22].

The majority of water treatment facilities used the traditional technique to remove ammonia, iron, and magnesium from groundwater. Aeration, flocculation, sedimentation, filtering, and disinfection are the steps in this traditional process. These techniques need a big plant area, expensive maintenance, and a large labour. This article focus was on using a thorough approach to cover all notable research completed in this area up till now. A final assessment was then conducted to determine the best effective method for removing iron, manganese, and ammonia ions. [23].

Ultrafiltration (UF) and its hybrid process are examples of membrane technology that has seen a sharp growth in use recently in the water treatment industry to produce resources for drinking water. Operational issues that were previously associated with conventional techniques can be resolved with UF technology. Superior divalent ion retention at reduced operating pressure and increased flow at reduced energy usage are further benefits of UF membranes [24]. This research aims to critically analyze

a unique treatment system that removes iron, manganese, and ammonium simultaneously and simultaneously from groundwater. Any kind of adsorptive, filtration, membrane bioreactor (MBR) process, and so forth, can be used in conjunction with ultrafiltration to remove the aforementioned contaminants.

2 IRON, MANGANESE AND AMMONIA SOURCES

In water supplies; ammonia, iron, and manganese are the most common inorganic compounds. In addition to human activity, a variety of natural mechanisms connected to the geological foundation have an impact on water quality. The Communities in Europe set lawful guidelines for the maximum amounts of pollutants that could be present in products in 1998.

2.1. Iron

After aluminum, iron (Fe) is the second most common metal on Earth. It makes up roughly 5% of the crust of the earth. in nature pure iron is rare because of its ions of iron (Fe^{2+} & Fe^{3+}) easily reacted with oxygen and molecules of sulfur to produce oxides, carbonates, sulfides and hydroxides. The highest prevalent form of iron in nature is found in its oxides (World Health Organization, 2003). Soils and rocks naturally contain iron that dissolves in water. It could also come from water's corrosive effect on exposed iron or steel tanks and pipelines. Periodically, significant levels of iron from acid runoff from mining operations or industrial wastes may also be present in surface waters. Iron contamination of water sources results in a number of aesthetic and functional issues, including poor color and taste, deposition in the water distribution system that raises turbidity, and staining. Water soluble Fe^{2+} compounds and water insoluble Fe^{3+} complexes are usually distinct from one another. The latter can only dissolve in extremely acidic solutions of water; nevertheless, in some situations, they become more soluble when reduced to Fe^{2+} [25]. The content of iron ions in the rivers have been found to be $700 \mu\text{g}/\text{liter}$ (mg/L). Anaerobic brackish groundwater generally has ferrous concentrations ranging from 0.5 to 10 mg/L, however in rare occasions they may surpass 50 mg/L. Typically, 0.3 mg/L or less of iron is present in drinking water. Countries that distribute water using galvanized iron, steel, and cast iron pipelines, or that employ different iron salts as coagulating agents in facilities that treat water, may have higher iron contents [26].

2.2. Manganese

Manganese causes issues with water supplies that are comparable to those brought on by iron. It is mainly identified as manganese dioxide in soils, specifically in the forms of Mn^{2+} and Mn^{4+} . Because manganese is much less common than iron in nature, it can be found in water supplies less frequently and in smaller amounts.

Additionally, due to the fact that ferrous solutions are less stable than manganous solutions, they are less likely to oxidize or precipitate than iron. As a result, natural surface waters may contain quantities of 1 mg/L of manganese, but is frequently detected at 0.2 mg/L or lower values.[27].

2.3. Ammonia

Ammonia comes in two forms: nonionized (NH_3) and ionized (NH_4^+). Both types of ammonia are known to be the main contaminants in sources of drinking water. Because the nonionised form is soluble in lipid and uncharged, it is the most toxic. This substance is more easily absorbed through biological membranes than the hydrated and charged NH_4^+ ions [28]. Numerous studies have been published, however they only linked NH_3 effects to overall ammonia toxicity [29, 30]. It has also been documented that ionized NH_4^+ has negative effects, particularly when it manifests in sufficiently high a concentration [31, 32]. Due to the biological breakdown of nitrogenous organic matter, ammonium may be found in most waterways, while discharges of industrial waste can also find their way into surface and ground waters. Additionally, runoff from agricultural regions where ammonium is utilized as fertilizer causes it to pass through surface waterways. Through water disinfection process with chlorine, they found that ammonia reacts with Cl_2 and produce chloramine which is a carcinogenic material. this contaminant needs to be removed. Additionally, fish poisoning, surface water eutrophication, and oxygen depletion are caused by ammonium in water systems.[25].

3 GROUNDWATER SOURCES

Groundwater resources make up an additional one-third of freshwater withdrawals. 50% of people on the planet use this resource for travel. Consequently, the phrase "hidden sea" is occasionally used to refer to groundwater. Different regions of the world have varying levels of groundwater usage, based on technological and cultural patterns as well as geographic location. An initial issue could be the mining of unsustainable wells and a negligible rate of recharge. The degree of contamination has increased due to the groundwater resources' proximity to mineral resources. While this is happening, municipal leachates have been noted as another source of contamination for underground resources. [33].

Groundwater typically contains dissolved forms of iron, manganese, and ammonia that don't change over time. Although most groundwater is free of microorganisms, the presence of manganese and iron in some aquifers encourages the growth of bacteria known as crenofoms, which reduce iron. Scientific names for these include Gallionella, Leptothrix, Sphaerotilus, and Crenothrix. Crenofoms collect in pipes to create dense, stringy, jelly-like masses that can reduce a system's ability to transport water. [25]. Total dissolved solid (TDS) concentrations in groundwater resources that fall into the brackish category

typically range from 1000 to 10,000 mg/L. Wells and aquifers are examples of both surface water (rivers and lakes) and groundwater (wells and aquifers) resources. Generally speaking, brackish water is easier to treat because it usually contains less pollution than saline sea water. On the other hand, every subclass has special characteristics. The salinity of brackish groundwater resources is higher than that of surface water [34]. Moreover, when the groundwater table drops, the salinity profile rises. Naturally, subterranean resources with a greater mineral element concentration. Because subterranean resources are limited, surface water has a significantly higher biological profile and pathogenic organism content [35]. Figure 1 provides a schematic diagram showing the salinity of various water resources.



Figure 1 – Different water resources salinity.

4 OVERVIEW OF TECHNOLOGIES

Ammonia, manganese, and iron are treated using a variety of unit processes. As previously mentioned, Iron and manganese in dissolved form are often found in water sources. These dissolved forms are transformed using conventional technologies into a particulate form that can be filtered and clarified. An oxidant is needed to change dissolved forms into particulate forms. Conventional oxidation methods are well-understood and have practical applications. Particulate forms of iron and manganese can be effectively removed utilizing traditional sedimentation and filtration techniques. Generally speaking, traditional technologies don't use proprietary treatment technologies and are easy to use. New technologies could be advantageous in scenarios where there are several contaminants or small sites. For systems that meet the following requirements, nontraditional technologies might be worth taking into account: • Elevated levels of radioactive elements •the Surface waters-impacted wells; wells with excessive levels of manganese, iron and limited

area; problematic residual disposal sites; and sites where extremely high quantities of backwash could affect water production. The following are examples of non-traditional treatment techniques for manganese, iron and ammonia removal at one step: Ballasted flocculation and Membrane filtration biological filtration [36].

SEQUESTRATION

A chemical added to groundwater is referred to as a sequestrant in the process of sequestration. Iron, manganese, and to enable them to remain in solution, ammonia ions form a connection with the chemical. Generally, sources with concentrations of iron and manganese below $600\mu\text{g.L}^{-1}$ and less than $100\mu\text{g.L}^{-1}$, respectively, are the only ones that can be applied for manganese and iron sequestration for drinking water treatment. Sequestration of source water concentrations above these limits is typically prohibited by regulators and may cause aesthetic problems in the distribution system [36].

OXIDATION, CLARIFICATION, FILTRATION

The most methods of removal treatment for iron and manganese include oxidation to transform the dissolved metals forms to solids, which is subsequently followed by filtration. Before filtering, a clarifying step is often required if the concentrations in the source water are above 8 to 10 mg.L^{-1} combined iron and manganese. For both greensand-type filters A and sand/anthracite combined iron and manganese concentration of 8 mg.L^{-1} will often lead to a filter running time of less than 24 hrs. Numerous technologies, including conventional sedimentation, solids-contact clarifiers, plate or tube settlers, ballasted flocculation and dissolved air flotation can be used to achieve clarification. anthracite filter, manganese dioxide ore, Manganese dioxide-coated sand media or a mono-medium sand are some examples of the filtration types that can be employed in this treatment approach [36].

MANGANESE DIOXIDE-COATED MEDIA FILTRATION AND OXIDATION

Manganese greensand and other ore-type catalytic media coated with manganese dioxide have long been used in filtration processes to remove manganese and iron. Technological developments in manganese dioxide media have resulted in increased loading rates and decreased backwash requirements in recent times. To eliminate iron and manganese, a variety of specialized systems with manganese dioxide coating are offered. The basic treatment involves oxidizing iron and manganese in addition to potassium permanganate or chlorine, and then filtering the precipitates, though the mechanics involved may differ. Manganese and iron oxidation and reduction are catalyzed by the manganese dioxide coating on the filtering materials. These media may be separated

into two categories: manganese dioxide-coated sand and manganese dioxide ore. All of these media are the same in that they remove iron and manganese from water by combining chemical oxidation with catalytic media [36].

OXIDATION, MEMBRANE FILTRATION

Pathogens and turbidity from groundwater under the direct influence of surface water (GUDI) and surface water are commonly removed from groundwater using membranes. Manganese and iron removal is another application for membranes. For GUDI wells that need to be treated for manganese and iron, membrane treatment is frequently a practical choice. The way membrane systems work is by filtering out particles bigger than the membrane's pore size. The standard classification for microfilters is between 0.05 and 1.0 microns, while ultrafilters are typically classified between 0.005 and 0.05 microns. Following conventional oxidation, dissolved manganese and iron that have been reduced to particle form is able to be separated on the membrane. Numerous producers of micro/ultrafilters have expertise iron and manganese removal from water. Every system has its own membrane and operates quite differently when it comes to cleaning and backwashing. There are systems for immersed and pressurized membranes [36].

BIOLOGICAL FILTRATION

The process of biological filtration functions hydraulically similarly to a pressure filter, involving the pumping of raw water using a high-pressure vessel that holds granular media. However, the biological processes don't need any type of chemical oxidants, in contrast to the majority of other pressure filtration methods, which rely on the formation of a chemical precipitate and subsequent filtering. Rather, the pressure vessel is made into an environment that is conducive to bacterial growth. The iron, manganese, and ammonia in the unfiltered water are oxidized by these bacteria and are subsequently trapped in the filter as dense precipitates. Compared to the amorphous precipitates created by chemical oxidizing processes, these precipitates are more compact. As a result, the biological filter can retain iron and manganese up to five times better. Longer filter run times are made possible by the system's enhanced metal retention capacity. The raw water is continuously infused with air to give the bacteria the right conditions for growth. It is significant to remember that the environmental requirements for biological manganese elimination and biological iron removal are not the same. Consequently, in situations containing manganese and iron, Biological filtration requires two stages: one of them for the biological removal of manganese and the other for the biological removal of iron. of manganese. The exclusive patented systems called Ferazur® and Mangazur® are used in biological filtering processes to eliminate iron and manganese, and they are made by InfilcoDegremont, Inc [36]. Researchers have made an attempt to look into a few

instances of biological filtering. An alternative to conventional water purification plants is using trickling filters to remove biological ammonia, iron, and manganese elimination. For treating groundwater (about 2.2 mg/L of NH_4^+ , 15.0 mg/L of Fe^{2+} , and 1.2 mg/L of Mn^{2+}), A membrane bioreactor modified with powdered activated carbon (PAC-MBR) was developed. It is necessary to purify three influents with varying degrees of pollution, such as raw water and the typical bio-sand filter's post-filtration effluent, which has varying quantities of dissolved oxygen (e.g., 9 mg.L⁻¹ and 6 mg.L⁻¹), the two lab-scale PAC-MBR systems are put into place and allowed to run for 220 days. The outcomes demonstrated that, regardless of the quantity of pollutants, 200µg/L of iron, 100µg/L of manganese, and 50µg/L of ammonia were found in a good MBR effluent produced; however, the initial phase of operation varied (20–49d) System I (PVDF) demonstrated good performance with the development of transmembrane pressure (TMP), showing a minor TMP increase between 7.0 and 17.0 kPa, in contrast, system II (PVC) showed a significant increase between 20.0 and 60.0 kPa. In short, the primary cause of membrane fouling was a cake layer made of oxides and PAC. High-throughput sequencing study revealed that the PAC-MBR systems depend on the ammoniaoxidizing bacteria (AOB) Nitrosomonas and Nitrospira, and the mnoxidizing bacteria (MnOB) Leptothrix, Pseudomonas, Hyphomicrobium, and Planctomyces[37,38].

For the purpose of treating groundwater that contains Fe^{2+} , Mn^{2+} , and NH_4^+ , In remote locations, decentralized water supplies can be provided by low-pressure ultrafiltration membrane (LPM) systems. It is difficult to enhance the LPM systems' functionality, such as their removal capacity and steady flux. In this work, the performance of a new opposite-flow low-pressure ultrafiltration membrane (O-LPM) technology was evaluated. According to experimental findings, 1.87 and 1.74 times more steady flux was produced by the O-LPM systems than by the conventional. A mixed pollutant system including 0.5 mg.L⁻¹ of Fe^{2+} , 0.3 mg.L⁻¹ of Mn^{2+} , and 1.0 mg.L⁻¹ of NH_4^+ reduced the 16-day O-LPM ripening period for Mn^{2+} removal to 8 days. Additionally, the ammonia removal efficiency reached to 80.97% from 61.46%. The O-LPM systems have a greater steady flux range due to the bio-cake layer being thinner and more uniform than in the D-LPM systems. In general, O-LPM system had higher concentrations of functional bacteria (IOB, NOB, and MnOB) than D-LPM systems did. When everything is said and done, these results are very important for treating groundwater in remote spaces and provide guidance for widespread application of the O-LPM system in decentralized water systems. [21]. (The membrane filtration of iron and manganese investigated the impact of changing the artificial groundwater's pH on the membrane performances and permeate quality. To determine how well two commercially available polyamide nanofiltration and ultrafiltration membranes (PA-NF, PA-UF) may treat groundwater for drinking

water sources, tests were conducted on them.) The artificial groundwater's permeate quality is deemed satisfactory for meeting the WHO drinking water standard when manganese and iron concentrations approach 0.1 and 0.3 mg/L, respectively. According to previous studies, membrane performance has greatly enhanced at pH values between 3 and 11. For all studied membranes, rejection of iron increased as the pH of the feed solution rose at a 100 mg/L feed concentration. Using a 50 mg/L feed concentration, the manganese rejection, however, displayed different performance patterns for every membrane. Both the solute's characteristics and the membrane surface properties were significantly impacted by the feed solution's pH. This indicates that the tested membranes' performance has increased due to the solute membrane contact mechanism.[39].

For the provision of ground water, low-pressure ultrafiltration membranes (LPM) are the perfect technology. The applicability and associated methods for eliminating ammonia (NH_4^+), manganese (Mn^{2+}), and ferrous iron (Fe^{2+}) from the source water, however, were not known. To fill this important information gap we carried out a thorough comparison of the MnO_2 -preloaded membrane (Mn-LPM) and the pristine membrane (pr-LPM). Mn-LPM increased the removal of NH_4^+ , Mn^{2+} , and Fe^{2+} while allowing for a higher water flow. On the first day, the efficiency of removal Mn^{2+} immediately had reached 99.6%. MnO_2 preloading improved NH_4^+ removal capability as well. The increase of NH_4^+ concentrations have produced a thicker bio-cake layer, which is consistent with increased pollutant removal efficiency and a drop in stable flux, according to a number of instrument characterizations. Anaerobic Ammonia Oxidation probably helps the removal of high NH_4^+ concentrations. These results significantly expand our understanding and support the creation of low-pressure monitoring (LPM) systems for accessible groundwater supplies in remote locations[40].

RESIDUALS

All processes used to remove ammonia, iron, and manganese produce residuals. Each site will have different requirements for residuals treatment based on the amount of available land, whether it makes sense to dispose of waste in a sanitary sewer, whether recycling to the plant's head is feasible, and other considerations. When choosing an iron, manganese and ammonia removal process, it is important to take into account the process of treatment produced residuals as well as the available treatment alternatives. The following are a few residuals treatment procedures that could be taken into account: • Batch settling with solids discharged to the sewer and decant recycling; • Equalization followed by treatment using plate settlers with decant recycle; • Direct sewer discharge; • Lagunitas • Dewatering by machine.

CONCLUSION

Globally, the three main contaminants of water reservoirs are manganese, iron, and ammonia. There are numerous physicochemical methods for treating water. However, the research shows that these methods are unable to efficiently remove all the contaminants at once since each technique shows a varied removal for each contaminant. However, there appear to be some benefits to using UF membrane and biological processes, such as reduced expenses and greater rates of simultaneous and coordinated elimination. This review includes a few published papers on the simultaneous and combination removal of manganese, iron, and ammonia. The majority of methods, according to analysis, require many steps to ensure that all contaminants are removed at high rates. The main findings of a detailed examination of the factors influencing simultaneous pollutant removal are as follows:

- Ammonium is oxidized (primarily) by microbes, whereas manganese and iron removed from brackish groundwater using a variety of chemical-physical, microbiological, and/or combinations of these techniques.
- Simultaneous elimination of iron, manganese, and ammonia is compared using pr-LPM and Mn-LPM systems. Fe^{2+} , Mn^{2+} , and NH_4^+ were removed through both systems at steady fluxes. The incremental NH_4^+ concentration reduced steady flow while improving clearance of pollutants. The highest concentration of NH_4^+ which might be treated was raised from 1.0 $\text{mg}\cdot\text{L}^{-1}$ to 1.5 $\text{mg}\cdot\text{L}^{-1}$ and nearly all of the within the first 10 days, Mn^{2+} was eliminated. The preloaded MnO_2 enhanced the elimination of pollutants and steady flow. The bio-cake layer became thicker as a result of the increasing NH_4^+ concentrations.
- Iron, ammonia, and manganese can all be eliminated in one step at the same time, even in the presence of high pollution levels and hydraulic loading. In particular, it was discovered that in every instance, PA-NF rejected both metals at a higher rate than PA-UF. Furthermore, the outcomes demonstrated that the PA-NF membrane effectively filtered out Fe^{2+} and Mn^{2+} ions to levels that were safe for drinking water according to WHO guidelines. The effective separation layer of the PA-NF membrane's structure was primarily responsible for its excellent separation performance.

Declaration of competing Interest

The authors state that none of their known financial conflicts or interpersonal connections might have had an impact on the work presented in this publication.

Acknowledgments

The Egyptian National Research Center Foundation supported the study. [Grant No. 6/3/24].

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