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Comparison between the performance efficiencies of reverse osmosis and nanofiltration membrane systems in removing heavy metal ions from industrial wastewater

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Abstract

Toxic contaminants that impact the health of humans and other animals include industrial wastewater, including cadmium, cobalt, chromium, and lead ions. To protect the environment, technologies such as "Reverse osmosis" (RO) and "Nanofiltration" (NF) membrane systems efficiently remove these ions from industrial effluent. Industrial wastewater samples were generated at room temperature and treated with both membrane systems in the laboratory. The samples contained Cd, Pb, Cr, and Co ions at concentrations ranging from 10 to 500 ppm, pressures ranging from 3 to 11 bar, and pH levels ranging from 4±0.2 to 7±0.2. Based on the findings, the RO system was able to remove Pb, Cd, Co, and Cr ions with efficiency of 98.55%, 97.97%, 97.308%, and 97.106%, respectively, when operated under the following conditions: $pH = 6\pm0.2$, pressure = 11 bar, pollutants concentrations = 500 ppm, time = 90 min at 25±2 °C. Operating parameters included a pH of 6±0.2, a pressure of 11 bar, a concentration of pollutants of 500 ppm, a duration of 90 minutes at 25±2 °C, and removal efficiencies of 96.37 percent for Pb, 95.44 percent for Cd, 94.478 percent for Co, and 93.965 percent for Cr in the (NF) system.

Key words and phrases: Membrane technology; reduce heavy metal HM ions; reverse osmosis RO; nanofiltration NF system.

Mathematics Subject Classification: 16W70

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1. Introduction

A possible global water shortage is an enormous problem that humanity is now trying to solve. An increase in the global population causes a chain reaction: more people need more manufactured goods, which means more factories need more water. Water contaminating water sources endangers human life and technological progress [1].

Even minute concentrations of heavy metal ions in water can devastate humans. Organ failure, behavioral issues, impaired learning capacity, and memory loss are among the many negative consequences of HM poisoning. Research by Singh et al. and Jamil et al. [2, 3] suggests that the symptoms and effects of metal poisoning can differ from one individual to another, depending on the specific type and concentration of metal in their blood. Professionals in water treatment still confront the important and difficult task of effectively filtering out undesirable minerals from water systems. The most effective methods for treating water polluted with heavy metal ions are electrochemical processes, ion exchange, adsorption, membrane filtration, and chemical precipitation [4–7].

Conventional secondary treatment processes cannot successfully treat many industrial effluents due to the high requirements to eliminate certain hazardous chemicals and resistant organic compounds. Therefore, state-of-the-art techniques for treating wastewater are required [8].

Some metals can accumulate in living things and eventually end up somewhere else. Zinc is necessary for plant and human growth, but too much of it can cause problems, including hemolysis and low hemoglobin levels, as Alnasrawi et al. [9] pointed out. The UN aspires to have accomplished its sustainability and environmental preservation objectives by 2030, according to Mengting et al. [10]. It is impossible to achieve the Sustainable Development Goal (SDG) of a clean water environment because of water contamination from industrial effluents that contain harmful HMs, according to Kurniawan et al. [11]. The main reason hazardous materials (HMs) pollute water to an unacceptable degree is that proper water treatment technologies cannot reduce pollution levels. So, even after treatment, the water is still unsafe to drink, and over 2 billion people worldwide still don't have enough of it [12].

Natural disasters may occur due to the discharge of harmful substances into our water system, including heavy metals and pharmaceuticals. Heavy metals, such as cadmium Cd (II), are a threat to aquatic life and, in turn, to human health because they can contaminate food sources. Dangerous amounts of antibiotics like moxifloxacin (MFX) in water treatment effluent threaten humans and promote the growth of drug-resistant microbes [13].

Membranes must conform to the precise criteria established by the World Health Organization (WHO) for chemical concentrations permitted in potable water—including water from industrial and urban areas—to isolate the target metals from actual, diluted water solutions. The water quality index for human consumption was determined by a series of fifteen critical physical and chemical characteristics. Sodium, potassium, chloride, nitrite, nitrate, and sulfate ions are among these characteristics, along with electrical conductivity, turbidity, pH, biochemical oxygen demand, dissolved oxygen, total alkalinity, and total hardness [14].

Due to its efficiency, cost-effectiveness, and absence of environmental harm, membrane separation technologies have replaced the previous, more polluting methods as the favored way for treating various industrial wastewater. Multiple membrane separation methods were employed to extract heavy metals from wastewater [15]. Included in this group of techniques were microfiltration (MF), "ultra-filtration" (UF), "reverse osmosis" (RO), and nanofiltration (NF).

Although there are limitations to the current technologies used to treat pharmaceutical effluents, membrane technology does provide an alternative. Physical characteristics and chemical composition of pollutants are the primary determinants of a membrane's efficacy. In contrast with the high pressures RO membranes utilize, microfiltration and UF utilize moderate pressures. To eliminate larger and more visibly sized airborne particles is the primary function of particulate matter filters [16].

Things with specific weight characteristics that allow them to be classified are shortened to HM. Several chemicals can be detected in the air in industrialized cultures. The human body cannot function properly without trace levels of several metals, including zinc, cadmium, copper, lead, and nickel. Acute or chronic poisoning can result from an overdose of any of these medications [17].

The membrane technology has been proven to be the most effective method for treating industrial wastewater. This method removes many contaminants at little cost and with little effort. Because membrane technology is so good at eliminating chemical and physical impurities and reaching 100% purity, it has increasingly relied on water treatment systems [18]. According to Gherasim and Mikulášek [19], various membrane systems can remove heavy metals from wastewater. These systems include reverse osmosis (RO), nanofiltration (NF), hybrids of the two, and others. Reverse osmosis (RO) is a separation technique that uses pressure to force a feed solution through a membrane. Solute and solvent are separated by a membrane pushed across by pressure. Now that it's permeable, the membrane may let solvents through while blocking minerals. The polymer layer, responsible for the actual separation process, is a crucial component of RO membranes. The ability of RO to remove particles, ions, and bacteria from liquids makes it an integral part of many industrial processes. Three factors: water flow rate, solute concentration, and pressure, influence the RO diffusion process [20].

Substances with molecular weights between 200 and 1000 Daltons can flow through a nanomembrane in nanofiltration (NF), which operates under pressure. A method for purifying water, nanofiltration filters out organic molecules with low molecular weight and dissolved ions that carry charges. Nanofiltration is a method that separates molecules by utilizing fragile and porous membranes with pores as tiny as a single nanometer. The operating pressure range of nanofiltration (NF) devices is from 0.3 to 1.4 MPa. A procedure that falls somewhere in the middle of ultrafiltration (UF) and reverse osmosis (RO), nanofiltration (NF) is also known as "bulk reverse osmosis" on occasion. Anions, or ions with a negative charge, can pass through nanofiltration membranes due to their electrical properties. Nanofiltration is one of the most well-known and cost-effective methods for desalinating salt water, also known for its remarkable energy efficiency. According to Dach and Madlool [21, 22], nanofiltration, nanofiltration offers a clear benefit over RO and electrolysis regarding operational and maintenance expenses.

This research aimed to determine whether it would be possible to use RO and NF in a pilot plant setting to extract Pb, Cd, Co, and Cr ions. The controlled variables were the pressure (in bar), pH, and the concentration of HMs ions (in ppm). Once the practical results were available, we compared them to RO and NF of other comparable projects.

2. Materials, Equipment, and Apparatus

Heavy Metals (HMs):

For this study, four different HMs solutions were chosen: Pb(II), Cd(II), Co(II), and Cr(III)—these ions are polluting in their nitrate form and become water-polluting ions when dissolved in water. This work's contaminants are shown in Table 1.

Table 1: HMs chemicals			
HMs ions	Nitrates	M.wt g/mol	Company
Pb^{+2}	$Pb(NO_3)_2$	331.2	HIMEDIA
$\mathrm{Cd}^{\scriptscriptstyle+2}$	$Cd(NO_3)_2$	236.42	CDH
Co^{+2}	$Co(NO_3)_2.6H_2O$	291.03	THOMAS
			BAKER
Cr^{+3}	$Cr(NO_3)_3.9H_2O$	400.15	CDH

Auxiliary Chemicals:

The chemicals used in modifying the pH and cleaning membranes are listed below in Table 2.

Table 2: Auxiliary chemicals			
Chemicals	Purity	M.wt g/mol	
HCI	37%	36.46	
HNO_3	68%	63.01	
NaOH	99.9	39.997	

Preparation of heavy metal solutions:

A lead ion solution is prepared by dissolving lead nitrate $Pb(NO_3)_2$ in deionized water, and depending on the required lead ion concentration, 1.6 g of lead nitrate is dissolved in tap water to make a 1000 ppm (mg/l) solution of lead. It is based on stoichiometry chemical calculations and in the same way as the rest of the ions.

Experimental Work:

Industrial wastewater containing the Pb⁺², Cd⁺², Co⁺², and Cr⁺³ concentrations were prepared by dissolving the required amount of Pb(NO₃)₂, Cd(NO₃)₂ · Co(NO₃)₂.6H₂O, and Cr(NO₃)₃ in tap water. Determine the pH of the solution to the desired value by adding 0.5 M nitric acid (HNO₃) or 1 M NaOH. The experimental RO and NF Pilot Plant is illustrated schematically and photographically in Figure 1. The feed tank was used to prepare the feed solution. The feed line pressure switch can operate within a 0 to 25 bar pressure range. A low-pressure pump draws the feed solution from the feed vessel to the micro filters, which remove pollutants from the water. Then, a high-pressure pump introduces the solution to the RO or NF membrane pipe.

Following treatment, the pure water was released from a pipe, as shown in Figure 1 and Table 3, with the feed flow rate regulated by a flow meter (ranging from 2 to 150 l/min). The feed hopper is used to recycle the concentrate waste stream, combined with the feed tank. The NF membrane's properties are shown in Table 4 by The Dow Chemical Company, whereas the RO membranes are shown in Table 3 by Vontron Membrane Technology Co., Ltd.

The concentration of heavy metal ions can be measured using the Atomic Absorption Spectrophotometer (AA-7000) manufactured by the Shimadzu Company in Japan. The drain valve was opened after the findings were obtained to remove the solution and rinse the system with deionized water.





Figure 1: Pilot plant system schematic and photographic involving RO and NF Starting with the feedtank, moving on to the low-pressure pump, microfilter, and high-pressure pumps NF or 5-RO, 6-Indicators of pressure, 7-Meters for flow, 8-Glob valve, and 9-Get valve.

	eristics of equipment in the system.
Equipment	Characteristics
Pump of low-pressure	• Type of pump: Pentax/INOX100/50Italy
(Feed pump)	• $Q = (5-45) l/min$
	• $H(m) = 25-46$
	• HP = 1
	• $P = 0.74 \text{ kW}$
	• $V = 230 V$
	• A=4.6 A
	• Ip = 44
	• $Tmax = 50 $ °C
	• $H_{Z} = 50$
	Continuous duty of One Phase
Pump of high-pressure	• Type of pump: PentaxUS-200/7Italy
	• $Q = (30-140)$ l/min
	• H (m) = 12.5-73.6
	• HP = 2
	• $V = 230$
	• A = 9
	• $T \max = 50 \circ C$
	• Ip = 44
	• $H_Z = 50$
	• $P = 1.65 \text{ kW}$
	• Continuous duty of One Phase
Holding Tank	Type: polyethylene
	Capacity: 200 litter
Flow meters	2–1501/min, Germany
Pressure Gauges	0–25 bar, Germany

Type of membrane	RO	NF
Model	ESPA1-4040	NF-4040
Material	Composite polyamide	Composite polyamide
Module	Spiral wound	Spiral wound
Size (ID, length) (inch)	(4x40) inch	(4x40) inch
Effective area (m ²)	7.9	7.6
Max operating temp (°C)	45	45
Max pressure (bar)	41.4	41
pH range of water	2-10	3-10

Table 4:	Characteristics	of RO	membrane	and NF	membranes
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One can ascertain the removal efficiency by contrasting the solute concentrations in the feed input (CF) and permeate (CP) solutions. The feed solution concentration in milligrams per liter (mg/L) and the solute removal efficiency are the two variables that need to be considered in this calculation [23].

$$\%R = \frac{C_F - C_P}{C_F} \times 100\tag{1}$$

The pH is adjusted, and the membrane surface is cleaned from metal scales using hydrochloric acid or another low-pH solution. The cleaning solution can be recycled by pumping it to the RO or NF membranes multiple times. The next step is a thorough rinsing with tap water in the system. The process is carried out until the conductivity of the product stream is comparable to that of the feed water [24].

A low-pH solution, like hydrochloric acid, can be adjusted to a specific pH to eliminate the metal scales from the membrane surface. Pumping the cleaning solution to the RO or NF membranes often allows it to be recycled. Xu et al. [24] state that once the product stream's conductivity matches the feed water's, the system is flushed many times with tap water.

3. Results and Discussion

RO System

Effect of HMs Concentrations on Removal Percentage:

Figures 2a, b, c, and d illustrate the outcomes of treating various Pb^{+2} , Cd^{+2} , Co^{+2} , and Cr^{+3} concentrations in the RO system. Since the ion removal %R increases as the concentration of lead ions does, it is evident that the treatment duration and concentrations of lead ions played a role.

Lead ion concentrations between 10 and 500 ppm improve the elimination and water purification rate. With a concentration of 500 ppm and a duration of 90 minutes, the most effective lead removal rate (%R) is achieved, with a rate of 98.55%. When all other factors remain constant, the optimal removal %R and purification for cadmium, cobalt, and chromium are 97.97%, 95.97%, and 95.12%, respectively. The following parameters were used: temperature = 25 °C, pressure = 11 bar, pH = 6, and 10–500 ppm concentrations. In light of these findings [25–28], after 80 to 90 minutes, there was less of a change, indicating that the influence of time started to diminish. As a result, the best performance for ion removal happens after 80 minutes, and 90 minutes is the most perfect. Notably, the removal order is affected by the mass of the dissolved ions; for example, a high %R is observed for Pb⁺² and all other ions when the atoms have a large mass. It was found that as the feed concentration (CF) is increased, the removal rate (%R) likewise increases, as a result of which It is in agreement with high removal and Cp falls at high concentrations greater than 300 ppm, leading to an increase in the percentage of removal [29], which are as follows: "Pb⁺²>Cd⁺²>Co⁺²>Cr⁺³" After 80–90 minutes, after 80 minutes, the rejection shift became less pronounced because the RO's positive charge across the membrane became more muscular, making it more challenging to block ions. Ion flux across the membrane is thus not significantly enhanced. Therefore, the removal rate change becomes economically infeasible after 90 minutes, and the optimal time is 90 minutes; this aligns with [30,31].

Effect of Pressure on Removal Percentage:

The effect of pressure on the efficiency of the RO system is significant because increasing the efficiency of RO requires increasing the pressure to the permissible limit. Accordingly, it was noted that the effect of pressure is positive, such that increasing the pressure from (3 to 11) bar leads to an increase in the removal percentage (%R) for the best %R: 97.39% for Pb⁺², 97.12 % for Cd⁺², 96.74 for Co⁺², and 96.57 % for Cr⁺³, as shown in Fig. 3, (Operation conditions: T = 25 °C, P = 11 bar, pH = 6, operation time = 90 min), the best pressure is 11 bar. After the pressure increased from 9 bar to 11 bar, there was a considerable increase in the removal of HMs ions. The reason is that pressure has a positive effect, but it can't be raised too high because it would cause the RO membrane to get saturated and the effect to turn negative. Consequently, 11 bar gave the best performance under pressure. As the operating pressure rises, the removal %R falls, reducing the resistance across the RO membrane and controlling the thickness of the boundary layer, which ultimately causes the membranes to become compacted. In Fig. 3, removal efficiency hits 97.4% at 11 bars. This result agrees with the findings [32,33,35].

Effect of pH on Removal Percentage:

An analysis of the effects of pH on the removal of heavy metal ions from industrial wastewater using the RO system reveals that the removal rate and efficiency of RO are enhanced with an increase in pH. The working area served as the basic medium as HMs ions did not precipitate. Consequently, RO works more efficiently when the acidic pH is increased. Find out what RO's best performance is. At optimal operating conditions (T = 25 °C, P = 11 bar, pH = 6±0.2, operation time = 90 min), Fig. 4 indicates that %R increases as pH increases from 4 to 7. The best values for Pb⁺², Cd⁺², Co⁺², and Cr⁺³ were 97.738%, 97.552%, and 97.308%, respectively. The substance's acidity or basicity is defined by the hydrogen ion, which is positive. This is the explanation behind this.



Figure 2: The RO process's HMs rejection rate as a function of time and concentration (a) "for Pb⁺², (b) for Cd⁺²", (c) for Co⁺², and (d) for Cr⁺³, under optimal operating parameters (T=25 °C, P=11 bar, time=90min, and pH=6±0.2)

When the pH of a material is low, hydrogen ions, which are positive, and counter ions, which are harmful, cover its surface. A positive charge on metal ions and a positive charge on hydrogen ions form an attractive force. The membrane's capacity to remove HMs was greatly diminished and membrane fouling was accelerated due to this repulsive force. To counter this, the repulsion force diminishes as pH increases, increasing the HMs adsorption force between the membrane surface and HMs. Hence, it reduces the fouling qualities of membranes. The H+ impact vanishes, and the effect of HM

precipitation becomes more probable in a neutral media with a pH of 7. It follows that the change removal rate reduces with a pH of 7 or above. The findings of [7,28,32,34,36] agree with this outcome.



Figure 3: The influence of pressure on the rate of HM rejection in the RO process was studied under the following conditions: T=25 °C, P=3-11 bar, pH=6±0.2, 500 ppm, and operating time=90 min.



Figure 4: Operational conditions: T=25 °C, P=11 bar, 500 ppm, operating time=90 min) The effect of pH on the rejection rate of HMs for the RO process.

Nanofiltration NF System

Effect of HMs Concentrations on Removal Percentage:

At a concentration of 500 and a time of 90 minutes, the removal rate is 96.37% Pb⁺², which is the best lead ions removal rate %R. With the same concentration and time, the best removal %R and purification for HMs Cd⁺² is 95.44%, Co⁺² is 92.79%, and Cr⁺³ is 90.73%, respectively. Under the conditions T = 25 °C, P = 11 bar, operation time = 90 min, and pH = 6, as shown in Fig. 5. a, b, c, and d. From these results, the change was less after 80 to 90 min, which means that the effect of time began to decrease. Therefore, the optimal performance for removing ions is after 80 min, and the most appropriate is 90 min. It was noted in Fig. 5 that the removal sequence depends on the dissolved ions, which are as follows: Pb⁺²>Cd⁺²>Co⁺²>Cr⁺³, but less than RO. As illustrated in the section *Effect of HMs Concentrations on Removal Percentage*.

Effect of Pressure on Removal Percentage:

Increasing the NF system's efficiency necessitates raising the pressure to the allowable limit, so the impact of pressure on efficiency is substantial. Consequently, it was observed that pressure has a positive effect, meaning that HMs removal percentages increase as the pressure increases from 3 to 11 bars. As shown in Figure 6, the optimal pressure yields removal percentages of 94.75% for Pb⁺², 94.476% for Cd⁺², 94.098% for Co⁺², and 93.948% for Cr⁺³. The NF membrane saturates at a certain point, and after that, the effect turns negative, even though pressure initially has a positive impact. Consequently, 11 bar was the ideal pressure performance. But not as much as RO. The section on the effects of pressure on removal percentage illustrates this.

Effect of pH on Removal Percentage:

Impact of the pH level on removing HMs from industrial water using a negative NF system. It was also found that the removal rate increases when pH increases, thus increasing the efficiency of the NF system. The observation from Figure 7 was that with the increase in pH ($4\pm0.2 - 7\pm0.2$) of the solution, there was a corresponding rise in %R of HMs, the best percentage of removal reached 95.534% for Pb⁺², 94.926% for Cd⁺², 94.478% for Co⁺², and 93.965% for Cr⁺³, at pH=6±0.2. The Best operations conditions: Pressure 11 bar, pH =6±0.2, 90 min, 500 ppm, and 25 °C, as in Fig. 7. Further to the explanation of the *effect of pH on Removal Percentage*, also raises the pH causes the precipitation of HMs

ions which enhances the efficiency of the nanofiltration membrane and thus increases the %R removal rate at pH=6. Nevertheless, in the medium of pH = 7±0.2, there is no influence of H+, and the impact of precipitation of HMs is more probable. Thus, the change removal rate declines when the pH is \geq 7±0.2. This result is consistent with the results of [28,32,36].



Figure 5: The heavy metal rejection rate for the NF process is ffected by the following variables: (a) duration, (b) concentration of HMs ions (Cd⁺², Co⁺², and Cr⁺³), (T=25 °C, P=11 bar, pH=6±0.2, and 10-500 ppm).



Figure 6: The NF process's rejection rate of HMs as a function of pressure (T=25 °C, P=3-11 bar, pH=6±0.2, 500 ppm, operating time=90 min) is studied.



Figure 7: Operating conditions for the NF process: temperature=25 °C, pressure=11 bar, pH=4-7, 500 ppm, and 90 minutes of operation to determine the effect of pH on the rejection rate of HMs.

Comparison of the Performance Efficiencies of RO and NF systems:

In Table 5. The Comparison of the performance efficiency of RO and NF systems, and the best condition for them.

Parameters	RO	NF	Ref.
Temperature, °C	25	25	Our work
Pressure, bar	11	11	Our work
pН	6±0.2	6±0.2	Our work
operation time, min	90	90	Our work
Heavy metals ppm	500	500	Our work
% R Pb ⁺²	98.55	96.37	Our work
$\% R Cd^{+2}$	97.97	95.44	Our work
% R Co+2	97.308	94.478	Our work
% R Cr ⁺³	97.106	93.965	Our work
% R Pb $^{+2}$	98.8	76.9	Lumami Kapepula et al., 2022
$\% R Cd^{+2}$	98.6	92.3	Lumami Kapepula et al., 2022
% R Co ⁺²	_	_	
% R Cr ⁺³	99.2	98.2	Lumami Kapepula et al., 2022
% R Pb ⁺²	100	_	Al-Jlil & Alharbi,2010; Lumami Kapepula, & Luis, 2024
% R Cd ⁺²	99.86	_	Al-Jlil & Alharbi, 2010; Lumami Kapepula, & Luis, 2024
% R Co ⁺²	100	_	Al-Jlil & Alharbi, 2010; Lumami Kapepula, & Luis, 2024
% R Cr ⁺³	87.92	_	Al-Jlil & Alharbi, 2010; Lumami Kapepula, & Luis, 2024
% R Cd ⁺²	99	90	Abu Qdais et al., 2004

Table 5: Efficiency comparison of RO and NF systems under ideal conditions

Comparison table between RO and NF with some references with similar practical conditions to this work. It was found that the optimal performance of RO is the best for various pollutants and better than NF.

4. Conclusion

HMs are substances that accumulate in industrial and sanitation facilities' water environments and sewage. These substances may exist in the environment. These HMs include Lead, Cadmium, Cobalt, and Chromium, naturally occurring metals. These elements exist in small quantities in the environment. They are beneficial to health when taken in moderate quantities, but when taken in large amounts, any of the elements can cause acute symptoms or chronic toxicity. From the results obtained, it was observed that employing the RO and NF systems to eliminate the ions of some HMs dissolved in industrial wastewater provided high efficiency, especially in the RO system, and could eradicate high concentrations of more than 500 ppm. The highest removal %R of the Pb⁺² ion reached 97.106%. While in the NF system, the highest removal %R of the Pb⁺² ion reached 97.106%. While in the NF system, the highest removal %R of the Pb⁺² ion reached 94.478%, and the Cr⁺³ ion reached 93.965%. These results were under operational conditions: pH = 6±0.2, pressure = 11 bar, concentration = 500 ppm, time = 90 min, and

T = 25 °C. The efficiency of the reverse osmosis RO system was higher than that of the nanofiltration NF system in the given conditions of this work. The treated water was achieved with a concentration below the permissible concentration of HMs in the case of the industrial wastewater mixture. The treated water was obtained below the permissible concentration of HMs in the case of an industrial wastewater mixture containing heavy metals. However, changing the removal rate decreases after 90 minutes and becomes uneconomical, and the most appropriate time is 90 minutes.

5. Abbreviations

- % R Removal percentage.
- Cd^{+2} Cadmium ion.
- CF The concentration of the inlet solution pm.
- Co^{+2} Cobalt ion.
- CP The concentration in the permeate ppm.
- Cr⁺³ Chromium ion.
- g Gram.
- HMs Heavy metals.
- M Molar concentration.
- M.wt Molecular weight.
- NF Nanofiltration.
- Pb⁺² Lead ion.
- ppm Parts per million.
- RO Reverse Osmosis.

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