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Evaluation of Commercial Reverse Osmosis and Nanofiltration Membranes for the Removal of Heavy Metals from Surface Water in the Democratic Republic of Congo

Vercus Lumami Kapepula ^{1,2,3,4,*}, Mar García Alvarez ^{3,4}, Vida Sang Sefidi ^{3,4},
Estella Buleng Njoyim Tamungang ^{5,6}, Théophile Ndikumana ², Dieu-Donné Musibono ⁷,
Bart Van Der Bruggen ^{8,9} and Patricia Luis ^{3,4,*}

- ¹ Hydrobiology Research Center, Department of Hydrology, Uvira 73, Democratic Republic of the Congo
 - ² Department of Chemistry, University of Burundi/Bujumbura, Bujumbura 1550, Burundi
 - ³ Materials & Process Engineering (iMMC-IMAP), UCLouvain, Place Sainte Barbe 2, B-1348 Louvain-la-Neuve, Belgium
 - ⁴ Research & Innovation Center for Process Engineering (ReCIPE), Place Sainte Barbe 2, B-1348 Louvain-la-Neuve, Belgium
 - ⁵ Department of Chemistry, Faculty of Science, University of Dschang, Dschang 96, Cameroon
 - ⁶ Department of Chemistry, Higher Teacher Training College, The University of Bamenda, Bamenda 39, Cameroon
 - ⁷ Department of the Environment, University of Kinshasa, Kinshasa 127, Democratic Republic of the Congo
 - ⁸ Process Engineering for Sustainable Systems (ProcESS), Katholieke Universiteit Leuven, Celestijnenlaan 200F, B-3001 Leuven, Belgium
 - ⁹ Faculty of Engineering and the Built Environment, Tshwane University of Technology, Private Bag X680, Pretoria 0001, South Africa
- * Correspondence: lumamikapepula@gmail.com (V.L.K.); patricia.luis@uclouvain.be (P.L.)



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Abstract: This study evaluates the performance of commercial reverse osmosis (RO) and nanofiltration (NF) membranes for the removal of metal ions from synthetic water and surface water carried from the north-west of Lake Tanganyika in the city of Uvira, in the east of the Democratic Republic of Congo. Metal ion analyses were performed by the standardized ICP-MS and ICP-OES methods. The RO membrane showed higher metal ion rejection in high-concentration solutions (synthetic samples) prepared in the laboratory as well as in low-concentration samples from real raw water collected near Lake Tanganyika. Rejection levels were higher than 98% for Cr³⁺, Pb²⁺, Cd²⁺, As³⁺, Ni²⁺, and Sb⁺³ ions in the synthetic solutions, and 99.2, 98.8, 98.6, 99.2, 98.4, and 98.8%, respectively, in the real samples. The concentrations of metals in the permeate varied depending on the feed concentration and were 0.15 to 1.02 mg/L, 0.33 to 22 mg/L, and 0.11 to 22 mg/L in RO, NF90, and NF270 membranes, respectively. Regarding the NF membranes, the rejection of Cr, Ni, and Cd ions was interesting: 98.2, 97.8, and 92.3%, respectively. However, it was lower for Pb, As, and Sb ions: 76.9, 52.5 and 64.1%, respectively. The flux of NF was 329 to 375 L/m².h, much higher than for RO membranes, which had a flux of 98 to 132 L/m².h. The studied membranes are thus a feasible solution to remove the studied metals from real water sources at low concentrations since they meet the standards of the World Health Organization on specific values assigned to chemicals from industrial sources and human habitation areas where these ions are present in drinking water.

Keywords: reverse osmosis; nanofiltration; wastewater; heavy metals; Lake Tanganyika

1. Introduction

Pollution of water by heavy metals is a critical global environmental problem [1–6]. Millions of tons of toxic metals such as As, Cd, Cr, Cu, Hg, Ni, Pb, Se, V, and Zn, delivered by manufacturing and mining activities, are annually released or transported to the environment and may undergo transformations that have great environmental, public health,

and economic impacts [7–10]. In addition, drinking water scarcity is one of the problems caused by population growth, global industrialization [11–15], mining, and agricultural shortages. The application of environmentally unfriendly techniques in industry and waste management, and natural forces such as volcanoes, earthquakes, or storms [16–18], have caused heavy metals to reach alarmingly toxic levels in the environment [19–21].

Due to their high solubility in aquatic environments, toxic metal ions can be easily adsorbed and concentrated in the tissues of living beings as their progress through the trophic chain can cause considerable health problems [22]. The removal of heavy metals is of particular concern because of their recalcitrant nature and persistence in the environment [23]; moreover, the removal of heavy metals from wastewater is an active area of research with many gaps [24].

Unlike many other pollutants, removing heavy metals from the environment is difficult because they cannot be chemically or biologically degraded and are ultimately indestructible [25]. Heavy metals released into water bodies by waste have an incorrigible impact on the aquatic system and destroy the self-purification capacity of an aquatic body [26], as well as posing a carcinogenic risk to public health [27]. This situation affects freshwater bodies, such as the case of Lake Tanganyika, given its lower altitude compared to that of the agglomerations erected on its banks. Unfortunately, it constitutes the main receiving environment for the pollution generated in Bujumbura (on the Burundian side), Uvira, Kalemie, and Moba (on the DR Congo side), Kigoma and Kipili (on the Tanzanian side), and Mpulungu (on the Zambian side). These agglomerations are also home to a diversity of chemical and agrifood industries and activities (markets for example) that generate enormous amounts of wastes in both liquid and solid forms [28], which if not treated will constitute a potential danger to the lake.

The World Health Organization (WHO) produces a series of water quality guidelines covering drinking water, the safe use of wastewater, and safe bathing water [29]. Table 1 presents the latest version, updated in 2017, of the surface and drinking water quality guidelines, which are the benchmarks for the safety of drinking water.

Table 1. World Health Organization’s quality guidelines for drinking water [29].

Element	Concentration Normally Found in Surface Water [28]	WHO Guidelines
Sb	<4 µg/L	0.02 mg/L
As	-	0.01 mg/L
Cd	<1 µg/L	0.003 mg/L
Mn	-	0.1–0.2 mg/L
Cr ³⁺ , Cr ⁶⁺	<2 µg/L	Chrome total: 0.05 mg/L
Cu ⁺²	-	2 mg/L
Pb	-	0.01 mg/L
Hg	<0.5 µg/L	0.01 mg/L
Ni	<0.02 µg/L	0.07 mg/L
Zn	-	3 mg/L

Several processes can be used to remove metal ions from wastewater, in particular precipitation, adsorption on activated carbon, ion exchange, membrane filtration, and hybrid systems [30–35]. Membrane separation technology is the most cost-effective and widely applied technology for water purification. Generally, the membrane acts as a selective barrier that allows for the passage of certain constituents while inhibiting the passage of other constituents based on different mechanisms depending on the type of membrane (e.g., size-exclusion, diffusion). Filtration-based membrane processes have been classified into microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), reverse osmosis (RO), and forward osmosis (FO), in which the first two often use a microporous membrane while the last three use a thin composite membrane with a selective dense layer [36]. In order to remove ions, NF and RO membranes have demonstrated good retention capacities. Boussouga et al. [37] showed that the variations in water salinity had no impact on the high

rejection of As (V) in nanofiltration, while the variation of pH significantly affected the rejection of As (V) and the presence of humic acid in the water increased the overall release of As (V). In the presence of humic acids, an improvement in As (V) rejection occurred with the NF270 membrane by 82–94%. At different salinity conditions (0.58–20 g L⁻¹), rejection of As (V) with both NF270 and NF90 membranes showed a consistently high removal of 82–88% and 93–98%, respectively. The elimination of Sb compounds did not depend on the pH of the solution since the oxidation state of antimony changes from Sb (III) to Sb (V) in a very short time. The elimination of Sb (III) and Sb(V) is nearly constant over a pH range of 3–10, unless Sb (III) removed with the NTR-729HF membrane greatly decreases by 60.2 to 45.7% with a reduction in pH from 7 to 10 [38]. Experiments conducted by Qdais et al. [39], show that RO and NF membranes can remove synthetic wastewater containing 250 ppm of Cu²⁺ and Cd²⁺ ions at 98% and 99% and the permeate concentration would be reduced to 3 ppm. Using NF, more than 90% of the copper ions were removed. Mohsen-Nia et al. [40] examined the effect of increasing ion size on membrane rejection; Cu²⁺, and Ni²⁺ ions were chelated by the disodium salt of ethylenediaminetetraacetic acid (Na₂EDTA), a commonly used chelating agent. The results obtained indicate that Na₂EDTA can increase the size of the chelated Cu²⁺ and Ni²⁺ ions, and therefore, their rejection in the RO removal process was 99.5%.

Ipek et al. [41] showed that treatment of the feed solution with EDTA did not influence the removal of Zn²⁺ in NF. The removal of Ni²⁺ and Zn²⁺ ions from the untreated solution was 99.3 and 98.9%, and after treatment, 99.7 and 99.6%, respectively. The recovery rate, rejection, and saturation factor all increased with increasing applied pressure. Increasing the pH improved the removal of Pb²⁺ from 86 to 99%, but did not affect the removal of Ni²⁺ from 93–99%; this is due to the differences in interaction between the surface of the membrane and the anion present in the solution [42]. The rejection of Ni²⁺ and Cr⁶⁺ ions increased with increasing pH but did not change significantly by the transmembrane pressure difference (10, 20, and 30 bar) for NF90 and NF270 membranes. The optimum conditions were found at 30 bars with a pH of 10 for the NF90 and NF270 membranes. Under optimal conditions for the NF90 membrane, the Ni²⁺ and Cr⁶⁺ rejection values were 99.2 and 96.5%, respectively. For the NF270 membrane, the Ni²⁺ and Cr⁶⁺ rejection values were 98.7 and 95.7%, respectively [43]. Lead ion rejection was greater than 98%, and increased slightly with increasing crossflow velocity and operating pressure and decreased slightly with increasing feed metal concentration. The rejection and permeate flux decreased slightly and increased sharply, respectively, with increasing feed pH [44]. The elimination of Ni²⁺, Pb²⁺, and Cu²⁺ ions from synthetic water using nanofiltration was 85%, 78%, and 66%, respectively, under specific conditions of concentration, pH, and temperature [45]. The pH of the solution and feed concentration has a significant impact on the separation of metals, and the pressure plays a different role in the rejection and permeability of metals, depending on the kind of membrane. According to the solution diffusion theory, water and solute molecules diffuse through each dense layer [46]. The rapid diffusion of water molecules and slow diffusion of solute molecules result in an increased rejection; meanwhile, the slow diffusion of water molecules and rapid diffusion of solute molecules result in a decreased rejection. The water flux also has a common dilution effect.

Given the fact that much work has been carried out in RO and NF on the rejection of three ions at maximal in solution by adjusting the pH of the feed solution and a chemical pretreatment before filtration, the present study presents a two-fold objective: first, the evaluation of the performance of several commercial membranes in reverse osmosis and nanofiltration in terms of retention, flow rate, and permeability, applied to synthetic water containing six different ions; and second, the assessment of the retention of several ions in real waters without prior pretreatment, which is a source of pollution to Lake Tanganyika.

2. Materials and Methods

2.1. Sampling Site of Real Samples

Real water samples were taken at the discharge collector located at the point indicated in Figure 1. This is the last point after the households and before the real water enters the lake, and where we sampled the maximum amount of real water emitted with the coordinates 029°08'39.0'' of longitude N and 03°24'26.3'' of latitude E in the city of Uvira, in the Democratic Republic of Congo, flowing northwest into Lake Tanganyika. These samples were first filtered on Whatman MN 615. ϕ 125 mm REF 431 012 filter paper model to retain large particles in solution, before being filtered through RO and NF membranes in the laboratory. Figure 1 shows the study area, as located by Google Earth and Arc GIS version 9.3 software. GPS was used to determine the geographic coordinates of the sampling sites.

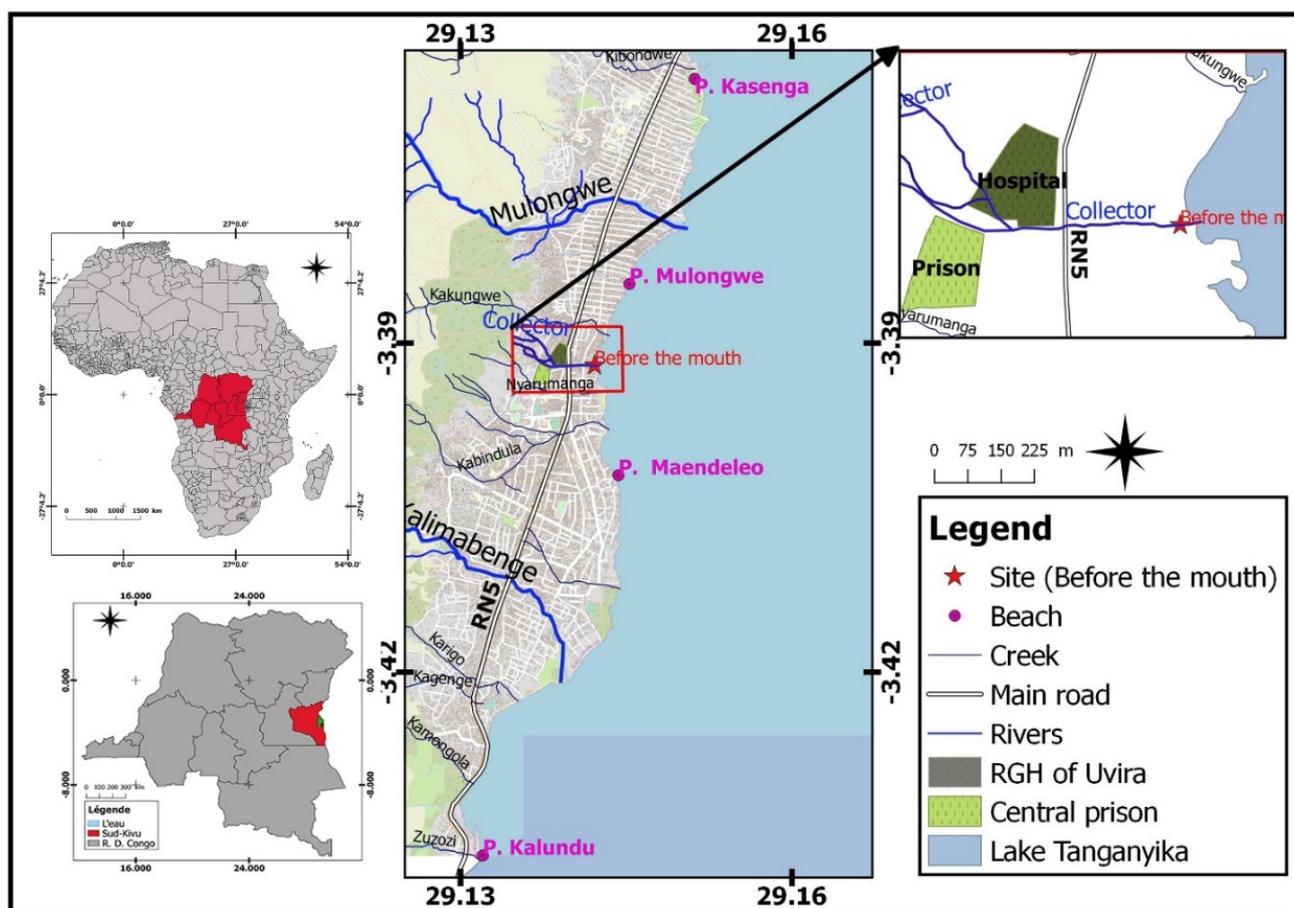


Figure 1. Location of sampling sites.

A chemical analysis was made to determine the concentration of each element in the actual wastewater (shown in Table 2). The initial concentrations were determined by IPC-MS and OES, measured in ppb and ppm. ICP-MS was used to detect low-concentration metals in the permeate and real waters and ICP-OES was used to determine toxic metal ions in high concentrations in synthetic waters. Real samples were filtered before being introduced in the membrane crossflow device. Surprisingly, the initial concentration before filtration was higher than after filtration, which may be due to the adsorption of metals on the removed particles. In Table 3, the measured concentrations before and after filtration are shown.

Table 2. Ion concentration in the real water sample.

Elements	Concentration before Filtration (ppm) *	Concentration after Filtration (ppb) *
Co	<0.05	28.13
Al	0.155	<1
As	<0.05	6.36
Ba	0.085	16.96
Cd	<0.05	<0.05
Cr	0.06	<0.05
Cu	<0.05	5.66
Fe	0.815	23
Mn	0.47	337.27
Ni	0.03	6.25
Pb	<0.05	<0.05
Sb	<0.05	1.57
Sn	<0.05	<0.05
Ti	<0.05	26.33
W	<0.05	5.27
Zn	<0.05	184.26
Ca	40	47,390
K	66.5	53,920
Mg	15	16,900
Na	78	930
Si	16	18,700

* ICP-OES was used to determine the concentrations before filtration (in ppm) and ICP-MS was used to measure the concentration after filtration (in ppb).

Table 3. Specifications of the membranes used in the experiments (information provided by the supplier).

Characteristics	RO X-20™ Membrane	NF90 Membrane	NF270 Membrane
Allowable operating pH range	4–11	NA	NA
Maximum operating temperature, °C	Continuous	45	45
Maximum feed turbidity, NTU	2	NA	NA
Maximum feed SDI, 15 min	5	NA	NA
Average salt discharge (%)	99.5	97	NA
Minimum salt discharge (%)	98.5	NA	NA
Filtration pressure (bar)	NA	41	41

NA: not available.

2.2. Synthetic Samples

In order to determine the membrane performance under different metal concentrations in water, synthetic solutions were prepared by adding specific amounts of metals, simulating high-concentration effluents (i.e., industrial effluents) and low-concentration effluents (i.e., water in natural water bodies) of heavy metals (chromium, arsenic, nickel, antimony, lead and cadmium). The low-concentration solutions had 15 ppm, and the high-concentration solutions had 56 ppm; the solutions were freshly prepared from an analytical-grade standard solution (Sigma Aldrich, Merck KGaA 64,271 Darmstadt, Germany) containing 1000 mg L⁻¹ of As³⁺, Cr³⁺, Pb²⁺, Cd²⁺, Ni²⁺, and Sb³⁺. A standard solution (Certipur®) was used to prepare the stock solutions.

2.3. Membrane Crossflow System Setup

The Armfield FT17 lab-scale device was used for evaluating the performance of membranes in a crossflow filtration application. This device enables rapid determination of crossflow filtration performance using a range of membrane types with relatively small sample volumes (1 L). Figure 2 shows the schema of the experimental setup. Both pre-

filtrated real samples (Section 2.1) and synthetic solutions (Section 2.2) were used as the feed solution in the crossflow setup.

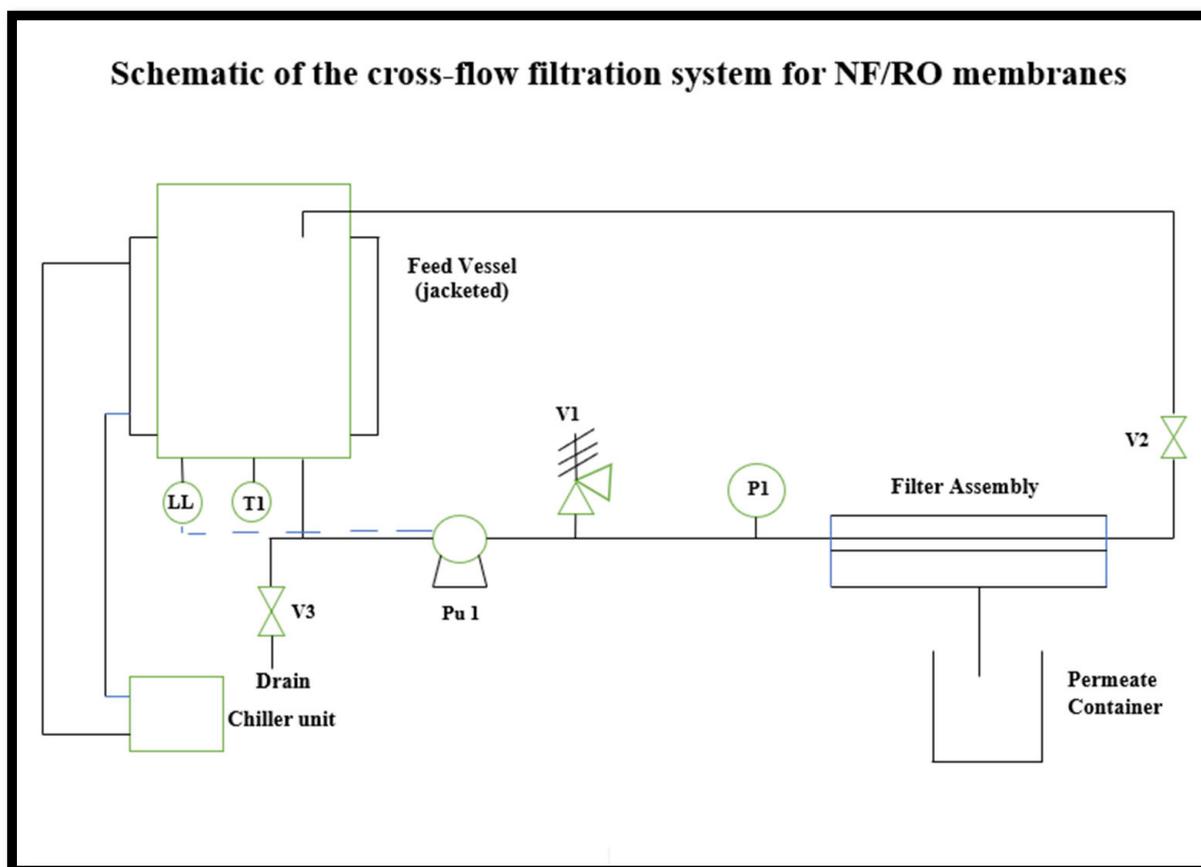


Figure 2. Legend: PI: pressure indicator (left); LL: low-level sensor (pump protection); LH: high level (vessel overfilling protection); P1: pressure sensor; T1: temperature sensor; V3, V2, V1: valves.

2.4. Commercial Membranes

The membranes used in this work are the polyamide urea RO X-20™, NF90, and NF270, supplied by Lennotech Water Treatment Solutions (Distributieweg 3, 2645 EG, Delfgauw, The Netherlands). The selection of these membranes for this study was based on average salt removal. The specifications of these membranes are presented in Table 3.

2.5. Experimental Procedure

Before the experiment, the membranes were soaked in ultrapure water for 30 min. The membranes were then inserted into the crossflow RO setup to filter the samples at different pressures, notably 50 bar for RO and 38 bar for NF. The choice of this pressure is justified by the characteristics of the nanofiltration membrane, which can be operated up to a maximum pressure of 41 bar. Referring to the literature [43], the pressure of 10, 20, and 30 bars did not significantly change the rejection at nanofiltration.

The prefiltered real water (see Table 2) and synthetic solutions were filtered in the RO unit. The feed capacity was 1/2 L at a constant maximum pressure of 50 bar in RO and 38 bar in NF. The temperature varied from 299.14 K for ions concentrated at 15–26 ppm and 303.14 K for ions concentrated at 56 ppm in RO, and in nanofiltration was 296.14 K for ions 15–26 ppm at 297.14 K for 56 ppm concentration. After filtration, the permeate and retentate of the real waters, because of their low feed concentrations, were analyzed by the standardized method [47,48] with an inductively coupled plasma mass spectrometer (ICP-MS), model NexION 5000 Multi-Quadrupole ICP Mass Spectrometer, provided by

PerkinElmer[®], and inductively coupled plasma optical emission spectroscopy (ICP-OES) provided by Agilent Technology was used for the synthetic solutions because of their higher concentrations. This analytical technique allows for the identification of the atomic composition of a particular sample. It uses the unique photophysical signals of each element to successfully detect the type and relative amount of each element within the complexity of a compound. From values of composition in the retentate and permeate (C_r and C_p , respectively), the retention was calculated as indicated in Table 4. In addition, the permeation flux, J , and the permeability, L_p , were obtained using the equations also listed in Table 4. In this work, the osmotic pressure was 0.026 bar, leading to a driving pressure (transmembrane pressure) of 48.96 bar.

Table 4. Main parameters used to evaluate the performance of RO and NF membranes.

Parameter	Formula	Equation	Ref
Retention (%) where C_p and C_r are the permeate and retentate Mass balance calculation	$R = 1 - \frac{C_p}{C_r} \cdot 100$ $(C.V)_f = (C.V)_r + (C.V)_p$	(1)	Luis et al. (2018) [49]
Permeation flux flux (J) ($L \cdot m^{-2} \cdot h^{-1}$)	$J = \frac{Q_p}{S}$ where Q_p is the permeate flow rate ($L \cdot h^{-1}$), S (0.00664 m^2) is the membrane area	(2)	Luis et al. (2018) [49]
Driving pressure DP $\Delta\pi$ is the osmotic pressure gradient (bar) C is the total concentration of ions (mol L^{-1}) $R = 0.082 \text{ L atm K}^{-1} \text{ mol}^{-1}$ T (K)	$DP = \Delta P - \Delta\pi$ $\pi = RTC$ $C = \frac{n}{V}$ with n as the total number of moles of ions	(3)	Van der Bruggen et al. (2018) [50]
Membrane Permeability L_p is the solvent (water) permeability ($L \cdot m^{-2} \cdot h^{-1} \cdot \text{bar}^{-1}$)	$L_p = \frac{J}{DP}$	(4)	Luis et al. (2018) [49]

3. Results and Discussion

3.1. Effect of the Metals' Feed Concentration on the Membrane Performance

The removal of ions by the studied membranes is shown in Figure 3 using synthetic solutions with 15, 26, and 56 ppm of metals. The synthetic solutions were filtered under a pressure of 50 bar at 303.14 K for the reverse osmosis membrane, and 38 bar at 297.14 K when using the nanofiltration membranes.

The results showed that the retention of Cr^{3+} , Cd^{2+} , Ni^{2+} , Pb^{2+} , Sb^{3+} , and As^{3+} ions was >98% (99.2, 98.6, 98.8, 99.2, 98.4, and 98.8%, respectively) independently of the concentration for the reverse osmosis membrane.

It was observed that the rejection of Cr^{3+} is similar to that of Ni^{2+} , although the size of Cr^{3+} is larger than that of Ni^{2+} . These two ions are larger than Cd^{2+} , Pb^{2+} , and Sb^{3+} , of which the sizes are the same. Removal of antimony was found to be <65% and arsenic <55% in nanofiltration.

The RO membrane gave much higher ion rejection than the nanofiltration membranes. The rejection of ions by the NF 90 membrane was almost at the same level as that at NF270, except for the 56 ppm feed solution, which was higher. Therefore, it was considered that size exclusion is not the only key factor in ion rejection. However, NF270 gave a lower ion rejection than NF 90 due to its larger pore size and also due to the interaction with the membrane. The *t*-test on single samples shows that the difference is not significant between NF90 and NF270 (*p*-value: 0.6 to 0.8) for all concentrations, but it is highly significant between RO and NF (*p*-value: 0.001 to 0.0001). The structure of the RO and NF membranes

would be the factor to influence the p-value. Very few studies have been conducted on the removal of antimony by membrane processes, unlike arsenic. Kang et al. [37] achieved a rejection of 60% Sb (III) and 95% Sb (V) by reverse osmosis using the ES-10 membrane, and 45–60% Sb (III) and 95% Sb (V) rejection using the NTR-729HF membrane. These values are lower than our research results using the reverse osmosis membrane.

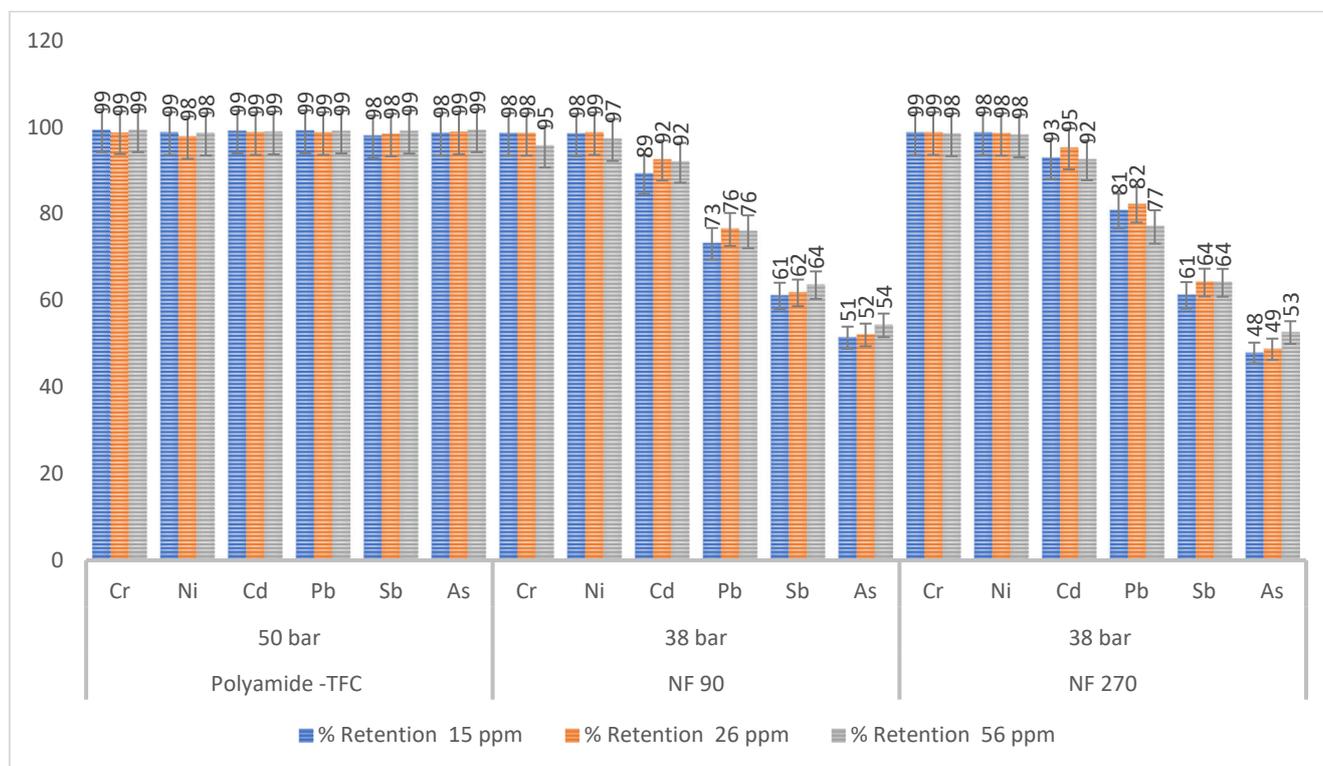


Figure 3. Removal of heavy metals as a function of feed concentration.

Previous studies showed that 99% rejection As (V) could easily be achieved with NF and RO membranes. The Pb retention is low and ranges from 73 to 82% using nanofiltration. However, despite this difference in sizes between the ions, the retention is very high and is almost similar in reverse osmosis, due to the interaction between the ions. Similar results have been reported by [51,52], who mentioned that Cd^{2+} can be removed at a slightly higher efficiency compared to Cu^{2+} .

In recent years [53] many works have been devoted to the removal of heavy metal ions using nanofiltration. It was reported the application of a thin-film composite nanofiltration polyamide membrane for the rejection of nickel ions from aqueous wastewater [54]. It was observed a nickel rejection of 98% and 92% for a feed concentration of 5 and 250 mg/L respectively, and they studied the binary separation capacity of heavy metals (cadmium and nickel) of a commercial NF membrane starting from solutions [55]. The maximum solute rejection observed for nickel and cadmium ions was 98.94% and 82.69%, respectively, for an initial feed concentration of 5 mg/L. The nickel rejection is similar to our results, unlike cadmium, which varies between 89 to 95%, and it is greater than their result. This difference would be due to the initial concentration and the pressure applied. Household and industrial wastewaters contain other heavy metal ions at various concentrations.

Several research studies have been conducted on the removal of As (III) and (V) by NF [53], and the removal of As (V) and As (III) was comparable. The low rejection of As (III) suggests that size exclusion governed their separation behavior and not charge interaction [53]. Boussouga et al. [37] confirmed the efficiency of NF to remove As (V), unlike As (III), which is much more difficult to remove, except when tight-pore NF membranes are used. Furthermore, NF membranes can only reject <50% of As (III) at $\text{pH} \leq 8$ [56,57].

Sato et al., Sen et al., and Tay et al. [58,59] recommended pretreatment by oxidation prior to filtration to improve As (III) rejection. These results highlight the role of charge interactions between the membrane and ions in determining the rejection performance of NF membranes, particularly for As species [60]. The X-20TM type TFC PA RO membrane was efficient in removing heavy metals (at low- or high-feed solution concentration), but the permeate flux was low and the applied pressure was very high. The nanofiltration membranes showed good ion rejection at low concentrations and the permeate flux was very high, but the retention was low for high-concentration feed solutions. Many low-income countries have very limited energy resources; consequently, the application of nanofiltration becomes relevant to lift this heavy metal challenge. On the other hand, despite the excessive energy consumption in RO, it still has many advantages to removing inorganic, organic, and microplastic micropollutants [61].

3.2. Ion Removal in Real Waters

It was observed that the retention of ions in the real wastewater, filtered by reverse osmosis, at 50 bar pressure at 303.14 K, for Co, Al, As, Ba, Cr, Cu, Ni, Sb, Sn, Ti, and Zn ions was 100% and for Fe, Mn, W ions, was 94.5, 99.2, and 88.7, respectively.

The retention of Co, Al, Ba, Cr, Cu, and Sn ions was also 100% by NF90 and NF270 membranes, while for As, Fe, Mn, Ni, Ti, and Zn ions, it was 99.8, 82.2, 88, 94.9, 99.2, 96.6% and 98.3, 90, 92.5, 98.6, 99.1, 95.9%, respectively. Tungsten was not retained by NF90 but NF270 had retained it by 27.2%. Cd, Pb, and Sn were below the ICP-MS detection limit.

The complete elimination of toxic ions and low level of heavy metal concentration in the permeate, below the WHO standard, means that good-quality water can be obtained. Figure 4 below, shows the ion retention of real waters.

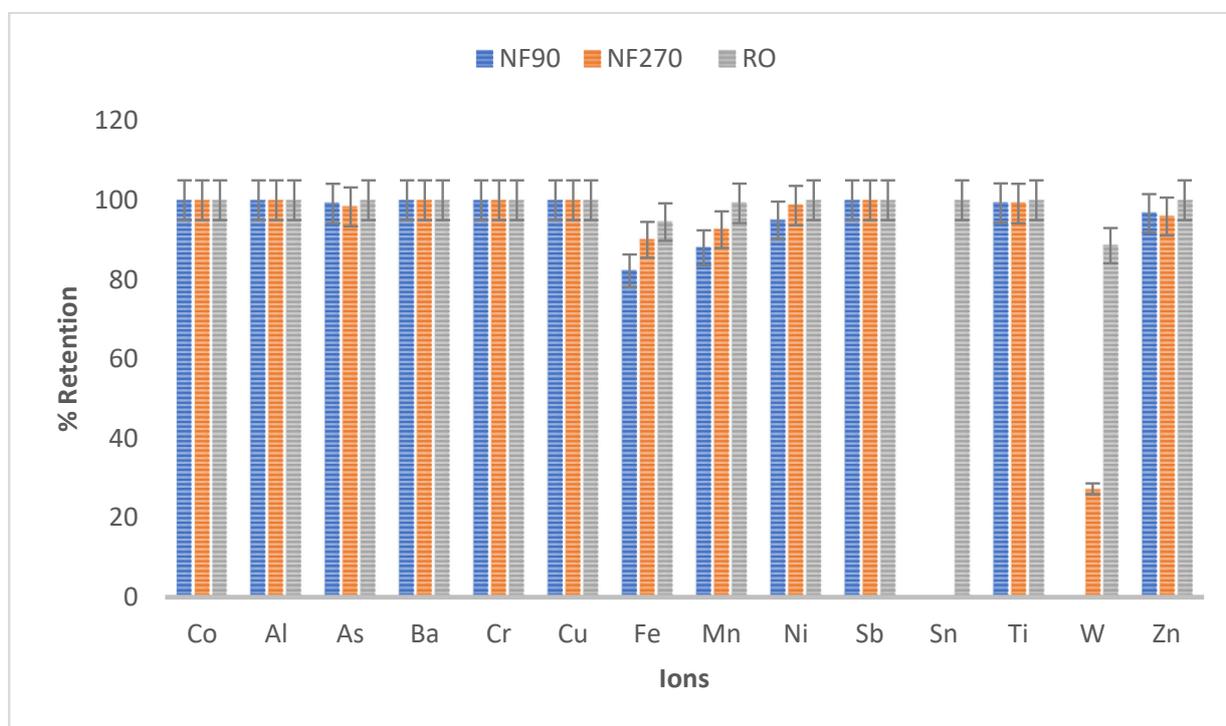


Figure 4. Retention of charged ions in real waters; analysis by ICP-MS.

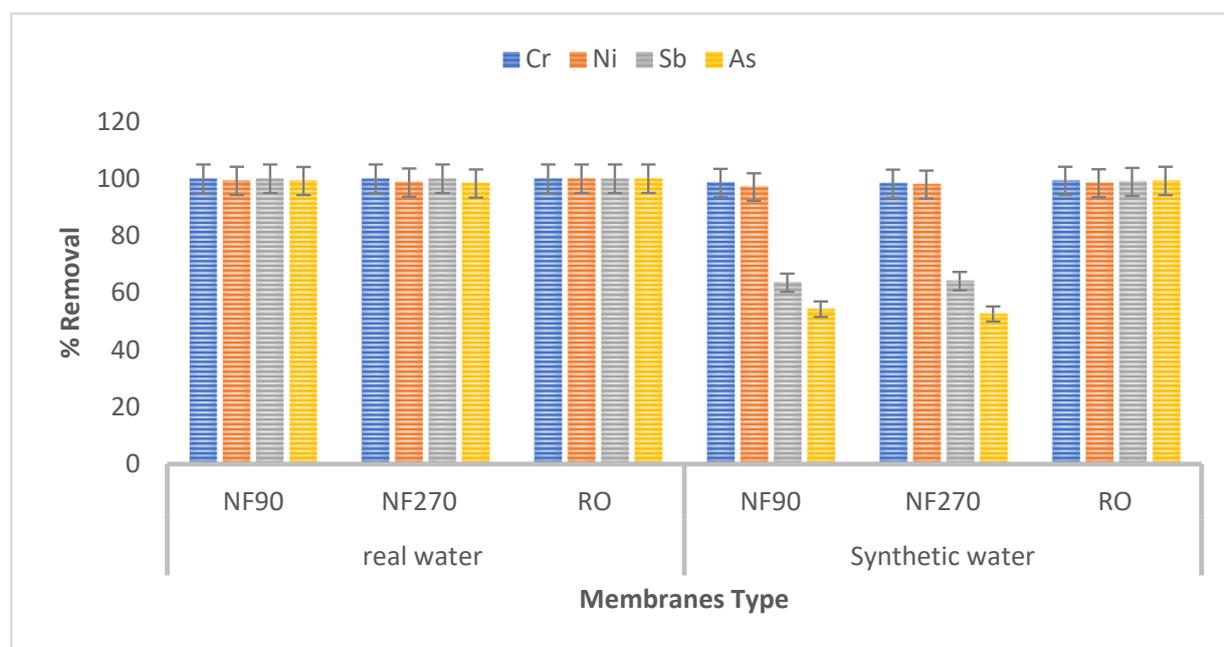
The filtration of real wastewater with RO and NF eliminated the ion concentration to the WHO standard, as shown in Table 5.

Table 5. Ion concentrations in permeate.

Elements	Ions Conc (ppb)		
	NF90	NF270	RO
Co	0	0	0
Al	0	0	0.00
As	0.30	0.36	0.00
Ba	0	0	0
Cd	<0.05	<0.05	<0.05
Cr	0	0	0
Cu	0	0	0
Fe	64.66	17.96	6.54
Mn	78.10	90.70	0
Ni	1.30	0.91	0
Pb	<0.05	<0.05	0
Sb	0	0	0
Sn	<0.05	<0.05	0
Ti	0.33	0.41	0
W	5.06	3.80	1.51
Zn	12.90	17.85	0.00

3.3. Comparison of Real and Synthetic Water Retention

Figure 5 shows the results of retention of the RO and NF membranes for real and mimicked surface water. It is observed that the removal of metals from the real water was higher compared to the synthetic solution. In real water, under the same feed pressures of 50 bars at 303.14 K in reverse osmosis and 38 bars at 297.14 K in nanofiltration, the Cr and Sb ions were completely eliminated from the permeate filtered by the three membranes. The rejection of Ni^{2+} and As^{3+} ions for NF90 was 99.20% and 99.18%, and for NF270, it was 98.68% and 98.3%, respectively. The Cd^{2+} and Pb^{2+} values in the permeate were below the detection limit by ICP-MS. No trace of these ions had been identified in the permeate after filtration by reverse osmosis. The retention of Ni^{2+} by NF90 (99.2%) agreed with the results obtained by Basaran et al. [44], for the same membrane.

**Figure 5.** Comparison of % removal between synthetic waters and real water.

On the other hand, for the synthetic solution, the rejection of Cr^{3+} , Ni^{2+} , Sb^{3+} , and As^{3+} ions were 98.49, 97.06, 63.53%, and 54.25%, respectively, for NF90, followed by 98.25, 97.97, 64.11%, and 52.59% for NF270 and finally 99.2, 98.41, 98.87%, and 99.21% for reverse osmosis. Those values are the same as the ones obtained for real wastewater, except for As^{3+} and Sb^{3+} .

The high rejection of the real waters can be explained by the low concentration of the load in ppb, unlike the concentration of the synthetic solution, which was 56 ppm. Lower concentrations in the feed thus involve higher rejection. This is in agreement with the results obtained by Shigidi et al. [62], whereby the removal of ions at 99.8% was possible in a 5 ppm feed solution and decreased to 94.3% in a 30 ppm feed solution and finally to 77.2% for the 100 ppm feed solution. Boussaga et al. [37] found that the presence of humic acids in wastewater can influence the removal of ions. [37].

Table 6 reviews the developments and potential applications related to nanofiltration and reverse osmosis, with a particular focus on heavy metal removal. Results from the present study show that the application of RO without chemical pretreatment achieved a more superior removal of toxic metal ions than that obtained by Chan et al. [52], Algreiri et al. [45], and Shigidi et al. [62], and was similar to other works, such as Ipek et al. [41] and Mohsen-Nia et al. [40], whose feed solutions were chemically treated. Removal of Cr, Ni, and Cd ions was excellent in nanofiltration and like previously treated feed solutions.

Table 6. List of key studies on heavy metal removal by using reverse osmosis and nanofiltration.

Type	Membrane	Element	Initial Conc	Type of Water	Removal Efficiency (%)	Conditions	Conc Permeate	Ref
NF	NF270	As (V)	250 µg/L	Synthetic	82–88	Feed pressure of 20 bars	ND	Youssef et al., 2021 [37]
	NF90				93–98			
RO	ES-10	As III	50 µg/L	Drinking water	75	0.75 MPa	ND	Kang et al., 2000 [38]
		Sb III			60			
RO	NTR-729Hf	As V	50 µg/L	Drinking water	95	0.75 MPa	ND	Kang et al., 2000 [38]
		Sb V			95			
RO	Polyamide	As III	250 ppm	Synthetic	20–43	ND	3.2 ppm	Qdais et al., 2004 [39]
		Sb III			45.7–60.2			
NF	Polyamide	As V	250 ppm	Synthetic	80–95	ND	13.16 ppm	Mohsen-Nia et al., 2007 [40]
		Sb V			95			
RO	Polyamide	Cd^{2+}	500 mg/l	Synthetic	98.5	Operation pressure 5 atm 3 atm	2.01 ppm	Chan et al., 2008 [52]
		Ni^{2+}	500 mg/l	Synthetic	82–97			
RO	Polyamide	Cu II	<500 µg/L	Drinking water	99.5	Operational pressure 1100 kPa	ND	Ipek et al., 2005 [41]
		As III			20–55			
RO	Polyamide	As V	<500 µg/L	Drinking water	91–99	Operational pressure 1100 kPa	ND	Ipek et al., 2005 [41]
		Ni ²⁺			44–169 mg/L			
NF	NF70	As III	ND	Drinking water	99	0.75 MPa	ND	Jekel et al., 2006 [42]
		As V			99			
NF	Polyamide	As III	1 ppm	Drinking water	<30	0.75 MPa	ND	Maher et al., 2014 [43]
		As V			60–90			
NF	NF90	Pb II	1 ppm	Drinking water	86	0.75 MPa	ND	Maher et al., 2014 [43]
		Ni II			93			
NF	NF90	Cu II	1 ppm	Drinking water	98.72	0.75 MPa	ND	Maher et al., 2014 [43]
		Pb II			99.61			
NF	NF270	Mn II	1 ppm	Drinking water	99.31	0.75 MPa	ND	Maher et al., 2014 [43]
		Ni II			99.11			
NF	NF270	Zn II	1 ppm	Drinking water	99.51	0.75 MPa	ND	Maher et al., 2014 [43]
		Ni II			99.2			
NF	NF270	Cr VI	Wastewater	Wastewater	96.5	10, 20, and 30 bars	ND	Basaran et al., 2015 [44]
		Ni II			98.7			
NF	NF270	Cr VI	Wastewater	Wastewater	95.7	10, 20, and 30 bars	ND	Basaran et al., 2015 [44]
		Ni II			95.7			

Table 6. Cont.

Type	Membrane	Element	Initial Conc	Type of Water	Removal Efficiency (%)	Conditions	Conc Permeate	Ref
NF	AFC 80	Pb II	5–250 mg/L	Synthetic	>98	25 bars	<0.05 ppm	Gherasim et al., 2014 [45]
		Pb II	15 mg/L	Wastewater	>99			
		Cd	5 mg/L		>99			
RO		Ni II			98.5	1 and 4 bars	ND	Algureiri et al., 2016 [46]
		Cu II			97.5			
		Pb II		Synthetic	96			
NF		Ni II	50 ppm		85	1, 2, 3 and 4 bars		
		Cu II	100 ppm		66			
		Pb II	150–200 ppm		78			
RO NF		Cu II	2 g/L	Synthetic	>95	Operating pressures 35 bar	ND	Cséfalvay et al., 2009 [51]
RO	TFC PA	Co II	39.4	Synthetic	98.6	41 bars	ND	Ricci et al., 2017 [63]
		Ni II	214.9		98.1			
		Mg II	242.9		98.6			
		Pb II	0.034		<1 ppb		<1 ppb	
		Zn II	0.153		<0.002 ppb		<0.002	
RO		Cd II	0.025	Real water	<0.1 ppb	1.76 MPa	<0.1 ppb	Thaçi, et al., 2019 [64]
		Co II	0.018		<0.2 ppb		<0.2 ppb	
		Mn II	1.146		99.48		0.006 ppb	
		Ni II	0.004		<0.5 ppb		<0.5 ppb	
RO	BW30XFR	Cr VI	5 30 100	Synthetic	99.8 94.3 77.2	10, 30, and 45 bars	ND	Shigidi et al., 2022 [62]
		Cr III			99			
		Ni II			98–99			
RO	X-20TM	Cd II	15, 26, 56 ppm	Synthetic	99	50 bars	0.15–1.02	
		Pb II			99			
		Sb III			98–99			
		As III			98–99			
		Cr III			95–98			
		Ni II			97–99			
	NF90	Cd II	15, 26, 56 ppm	Synthetic	89–92	38 bars	0.33–22	This study
		Pb II			73–76			
		Sb III			61–64			
NF		As III			51–54			
		Cr III			98–99			
		Ni II			98			
	NF270	Cd II	15, 26, 56 ppm	Synthetic	92–95	38 bars	0.11–22	
		Pb II			77–82			
		Sb III			61–64			
		As III			48–53			

ND: No data.

3.4. Transmembrane Flux

The results in Sections 3.1–3.3 show that the performance of membranes is evaluated based on the concentrations of the feed solutions. Permeate flux is one of the key factors demonstrating the effectiveness of membranes in resisting fouling [49].

The permeate flux of synthetic solutions and real waters was determined on the three membranes. The experimental results are presented in Figure 6 below, under operating pressures of 48.9 bar in reverse osmosis and 36.9 bar in nanofiltration. The nanofiltration membranes exhibited a high flux and a more severe flux drop in reverse osmosis.

The water permeabilities of the nanofiltration membranes were much higher than those of the osmosis membranes and are, respectively, 2.7, 2.5, and 1.9 L/m².h/bar for RO, 10.1, 8.9, and 8.3 L/m².h/bar for NF90, and finally 9.9, 8.8, and 8.4 L/m².h/bar for NF270.

A study [65] showed that the water permeability of NF90 and NF270 membranes were 6.05 and 14.86 L/m².h/bar and that of seawater reverse osmosis (SWRO) was 5.91 L/m².h/bar. Another study [66] showed that pure water permeability and salt rejection of both NF90 and BW30 membranes were measured by crossflow permeation tests with pure water and an aqueous solution of 1000 mg/L of NaCl, and were found to be 9.4 and 4 L/m².h/bar, respectively. The difference in permeability is explained by the crossflow filtration system, the applied operating pressure, and the solute concentrations.

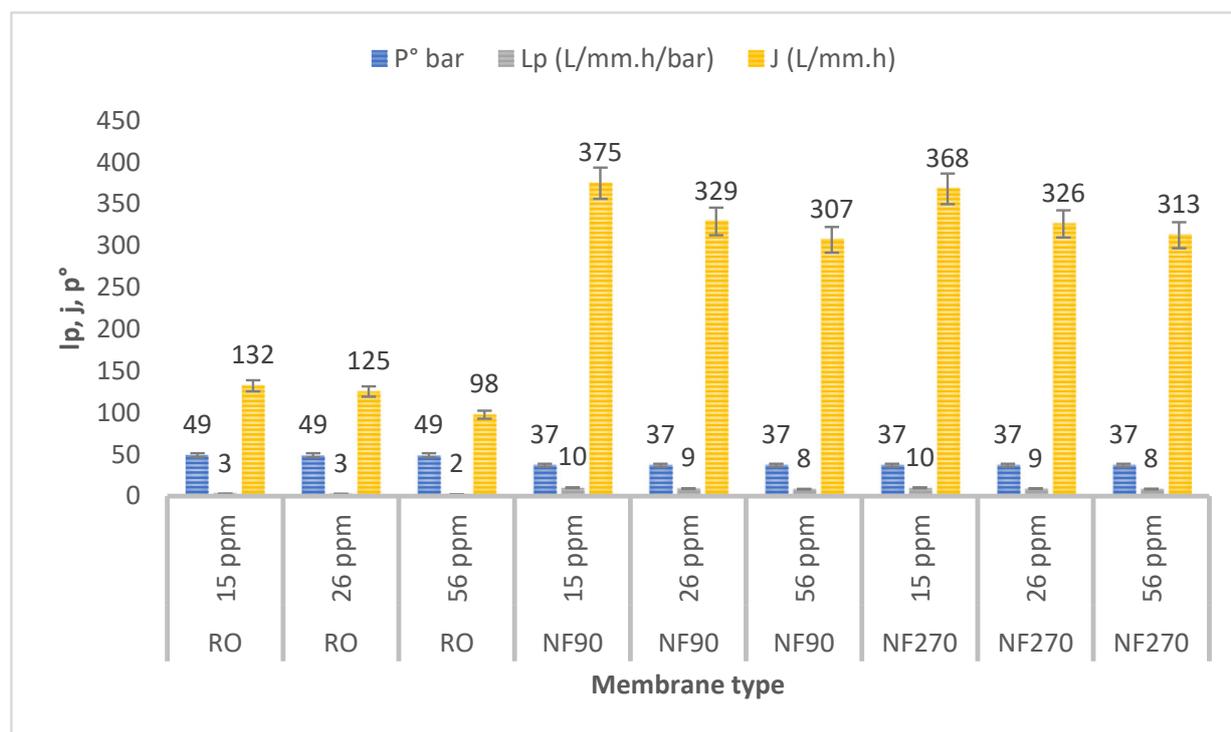


Figure 6. Permeability and permeate flow at constant pressure.

The increase in the concentration of solute slightly decreased the membrane permeability, and the temperature was raised from 299.14 to 303.14 K for RO, and from 296.14 to 297.14 K for NF. Low permeability due to solute concentration was observed at 15 ppm nanofiltration and 56 ppm reverse osmosis. Nanofiltration membranes had the highest water permeability and the rejection was lower than for reverse osmosis.

4. Conclusions

Three commercial nanofiltration and reverse osmosis membranes were successfully tested using synthetic solutions prepared in the laboratory and real water. The results showed that among the studied membranes, the reverse osmosis membranes had a lower permeability compared to nanofiltration membranes and the ion rejection was higher for the synthetic solutions. The ion release rate in the real water was higher and met the WHO standard.

The elimination of heavy metals at low concentrations in the permeate implies that good-quality water can be transported to Lake Tanganyika without disturbing the ecosystem, as well as perhaps the possibility of reusing it by the local population.

In conclusion, nanofiltration is recommended for the removal of heavy metals from low-concentration wastewater, and reverse osmosis would be the best choice for a very high concentration of heavy metals in wastewater. However, commercial membranes are very expensive and present some challenges in terms of excessive energy consumption, are sometimes inefficient for removing some heavy metals, and a low flux is observed. It would therefore be interesting to develop specific low-cost membranes such as mixed-matrix membranes (MMM) based on a biodegradable polymer and nanoparticle, which combine several advantages in terms of high permeate flux, good selectivity, low manufacturing cost, and low energy consumption.

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Abbreviations

Conc	Concentration
DRC	Democratic Republic of Congo
FO	Forward osmosis
ICP-MS	Inductively coupled plasma mass spectrometer
ICP-OES	inductively coupled plasma optical emission spectroscopy
Init	Initial
MF	Microfiltration
MMM	Mixed-matrix membranes
Na ₂ EDTA	Disodium salt of ethylenediaminetetraacetic acid
NA	not available
NF	Nanofiltration
ND	No data
PA	Polyamide
RO	Reverse osmosis
SWRO	Seawater reverse osmosis
TFC	Thin-film composite
UF	Ultrafiltration
WHO	World Health Organization

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