



Antibiotic Removal from Wastewater via Ceramic Membrane Distillation: Toward Clean Water and Antimicrobial Resistance Mitigation Aligned with Sustainable Development Goals (SDGs)

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ABSTRACT

Antibiotic residues in wastewater represent a growing concern for environmental and public health due to their role in promoting antimicrobial resistance. This study investigates the use of ceramic membrane distillation for removing amoxicillin, ciprofloxacin, and levofloxacin under various operational conditions. Two ceramic membranes, fabricated from red clay with nanocellulose and silica additives, were tested at different pH levels, temperatures, and antibiotic concentrations. Experimental results showed consistently high rejection rates, attributed to electrostatic repulsion and membrane-liquid interaction. Zeta potential measurements supported the correlation between membrane surface charge and separation performance. The study contributes to the body of knowledge on membrane-based water purification technologies and supports goals related to clean water access and reduced pharmaceutical contamination, as reflected in the United Nations Sustainable Development Goals.

ARTICLE INFO

Article History:

Submitted/Received 10 Mar 2025

First Revised 12 Apr 2025

Accepted 24 Jun 2025

First Available Online 25 Jun 2025

Publication Date 01 Dec 2025

Keyword:

Antibiotics removal,
Ceramic membrane,
Membrane distillation,
Wastewater.

1. INTRODUCTION

Antibiotics have become a cornerstone in the treatment of microbial infections across human and veterinary medicine. Their extensive use, especially in healthcare, livestock, and aquaculture, has led to a significant increase in global antibiotic consumption over recent decades [1,2]. According to the World Health Organization, annual antibiotic usage has surpassed 100,000 tons [3]. This massive consumption has resulted in the continuous discharge of antibiotic residues into the environment via pharmaceutical manufacturing effluents, municipal wastewater, and agricultural runoff. Such discharges contribute to ecological disturbances, alter microbial communities, and pose toxicological risks to higher organisms [4,5].

A major consequence of environmental antibiotic pollution is the acceleration of antimicrobial resistance (AMR), which occurs when residual antibiotics exert selective pressure on microbial populations, fostering the emergence of resistant strains. Several countries, including Saudi Arabia, India, Canada, and the United Kingdom, have reported rising incidences of antibiotic-resistant bacteria attributed to environmental exposure [6]. These resistant microorganisms, often labeled “superbugs,” compromise the effectiveness of standard therapies and increase morbidity and mortality rates. As such, the mitigation of antibiotic pollutants in wastewater has become a global imperative aligned with public health priorities and environmental protection frameworks.

Numerous water treatment technologies have been developed to address pharmaceutical contaminants. Methods such as nanofiltration [7-9], reverse osmosis [10-12], electrodialysis [13], forward osmosis [14], and membrane distillation [14-16] have demonstrated varying degrees of efficiency in removing antibiotic compounds from aqueous matrices [17]. Among these, membrane distillation (MD) has gained attention due to its ability to operate at low pressures, tolerate high salinity, and reject non-volatile solutes.

Previous studies have primarily focused on polymeric membranes in MD applications for antibiotic removal. Although effective, these membranes face limitations in thermal stability and long-term fouling resistance [14,15]. In contrast, ceramic membranes offer enhanced thermal and chemical durability, yet their application in antibiotic separation remains limited. This study introduces a novel approach by employing two types of ceramic membranes fabricated from modified red clay with nanocellulose and silica additives, aiming to assess their separation performance across various operational conditions. The novelty of this work lies in its integrated evaluation of ceramic membrane distillation for multiple antibiotics under environmentally relevant scenarios, supported by zeta potential analysis. By addressing this gap, the study advances membrane-based water treatment strategies and aligns with Sustainable Development Goals, particularly SDG 6 (Clean Water and Sanitation) and SDG 3 (Good Health and Well-being).

2. METHODS

2.1. Membrane Fabrication.

The ceramic membranes used in this study were created following the procedure described elsewhere [18], with some modifications to the composition. Two types of membranes were produced. The first membrane (M-1) was made of 91.5% red clay, 1.5% tetraethoxysilane (TEOS) as a silica precursor ($\geq 99\%$, Aldrich), a curing agent, and 5% ammonia catalyst. This mixture was combined with 2% sodium alginate powder (LOBA Chemie Co.) in 300 mL of distilled water to make a paste suitable for extrusion.

The second membrane (M-2) was fabricated by combining 91.5% Saudi Arabian red clay with 8.5% nanocellulose powder, serving as a pore-forming agent and binder. Both membranes were shaped using a plunger-type extruder and air-dried for three days on wooden racks covered in plastic film to ensure uniform drying.

Sintering was performed in two stages using a Nabertherm electric furnace. In the first stage, the temperature was gradually increased from 25 °C to 500 °C at 1 °C/min to remove organic matter. In the second stage, the membranes were sintered from 500 to 1000 °C at a rate of 2 °C/min for three hours to achieve densification and structural stability.

2.2. Antibiotic solution preparation

Three antibiotics—amoxicillin, ciprofloxacin, and levofloxacin—were prepared in varying concentrations by dissolving pharmaceutical-grade tablets in 1 L of distilled water. Solutions were stirred for 30 minutes under controlled conditions to ensure complete dissolution. The pH was measured and adjusted using HCl and NaOH to achieve pH values of 4, 6.5, 8, 10, and 12. All membrane experiments were conducted within four hours of solution preparation to maintain antibiotic stability. The concentration ranges for each antibiotic used in the membrane feed system are listed in **Table 1**. These ranges were selected to explore the influence of feed concentration on membrane performance.

Table 1. The concentration of antibiotic solutions used to feed into the membrane vacuum system.

Antibiotic	Initial concentration (mg/L)
Amoxicillin	25, 50, 75, 100
Ciprofloxacin	18.75, 37.5, 56.25, 75
Levofloxacin	12.5, 25, 37.5, 50

2.3. Membrane and Antibiotic Characterization

2.3.1. Zeta Potential

The electrokinetic behavior of the membranes and antibiotics was measured using a SurPASS™ 3 analyzer (Anton Paar, Germany) in the presence of 1 mM KCl as background electrolyte at 25 ± 1 °C. Zeta potential measurements were conducted across a range of pH levels to evaluate surface charge properties and their implications for separation efficiency.

2.3.2. Membrane Morphology

Surface and cross-sectional morphologies of the membranes were examined using a scanning electron microscope (SEM, JSM-7100F, JEOL, USA). Membranes were vacuum-dried for 24 hours and sputter-coated with gold (SPI Inc., USA) prior to imaging.

2.4. Antibiotic Removal Test

Membranes M-1 and M-2 were tested in a vacuum membrane distillation (VMD) system operating at a feed flow rate of 55 L/h and a vacuum pressure of 3.5 mbar. The influence of feed solution temperature, pH, and antibiotic concentration on membrane performance was investigated using selected parameters. The experimental setup is illustrated in **Figures 1 and 2**.

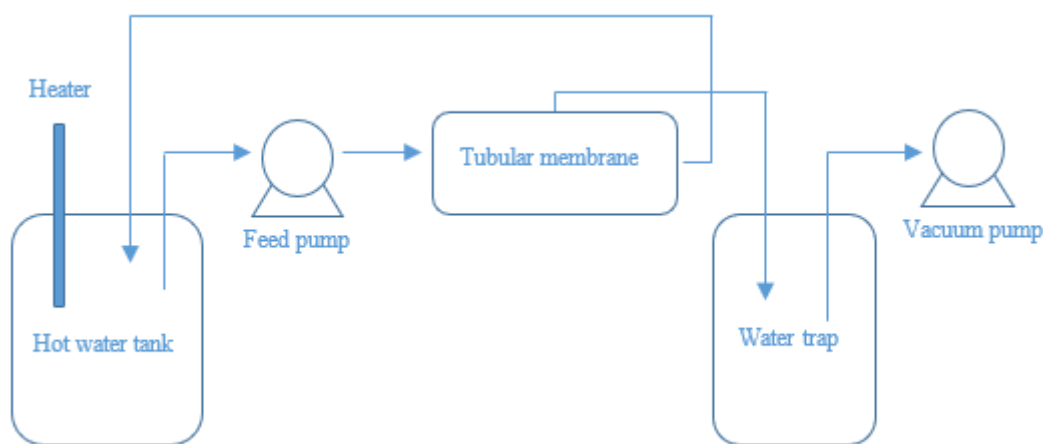


Figure 1. Schematic diagram of the Vacuum Membrane Distillation Process.



Figure 2. Vacuum membrane distillation (VMD) experimental setup

Permeate flux (J , kg/m²·h) was calculated based on the mass of condensed water (M_d), active membrane surface area (A), and time (t): $J = \frac{M_d}{A \cdot t}$.

After each cycle, membrane cleaning was performed using deionized water at 70 °C and a vacuum of 3.5 mbar. This process was repeated until flux and conductivity returned to baseline (~1 µS/cm).

Antibiotic rejection ($R\%$) was determined using $R\% = \left(1 - \frac{C_p}{C_f}\right) \cdot 100\%$, where C_p and C_f are the permeate and feed concentrations, respectively.

2.5. Liquid Chromatography–Mass Spectrometry (LC–MS/MS)

2.5.1. Chemicals and Reagents

LC–MS/MS analysis utilized methanol, acetonitrile, water, and formic acid (99%) from Merck (Germany). Analytical standards for amoxicillin, ciprofloxacin, and levofloxacin were obtained from Dr. Ehrenstorfer GmbH (Germany).

2.5.2. Analytical Conditions

Quantitative analysis was carried out using an Agilent 1290 LC system coupled with a 6500 QTrap MS (Sciex, USA), equipped with an ESI source. The mobile phases included water and acetonitrile with 0.1% formic acid. The elution protocol employed a gradient mode with a flow rate of 0.3 mL/min and a column temperature of 45 °C. Instrumental settings such as gas

flow rates, voltage, and temperatures were configured for optimal ionization and detection, as previously described (Al Tamim et al., 2022). **Table 2** lists the key mass spectrometry parameters for each antibiotic.

Table 2. LC–MS/MS antibiotics analysis parameters.

Analyte	Precursor Ion (m/z)	Product Ion (m/z)	CE	DP	EP	CXP
Amoxicillin	366.1	349.1 / 114.1	13 / 30	46	10	10
Levofloxacin	362.1	318.1 / 261.1	25 / 37	81	10	10
Ciprofloxacin	332.0	288.2 / 314.1	25 / 20	60	10	

3. RESULTS AND DISCUSSION

We successfully fabricated the ceramic membranes using two different recipes. The membrane showed a typical symmetric structure, illustrating uniform porosity from the surface to the other end, as shown in cross-sectional view in **Figure 3**.

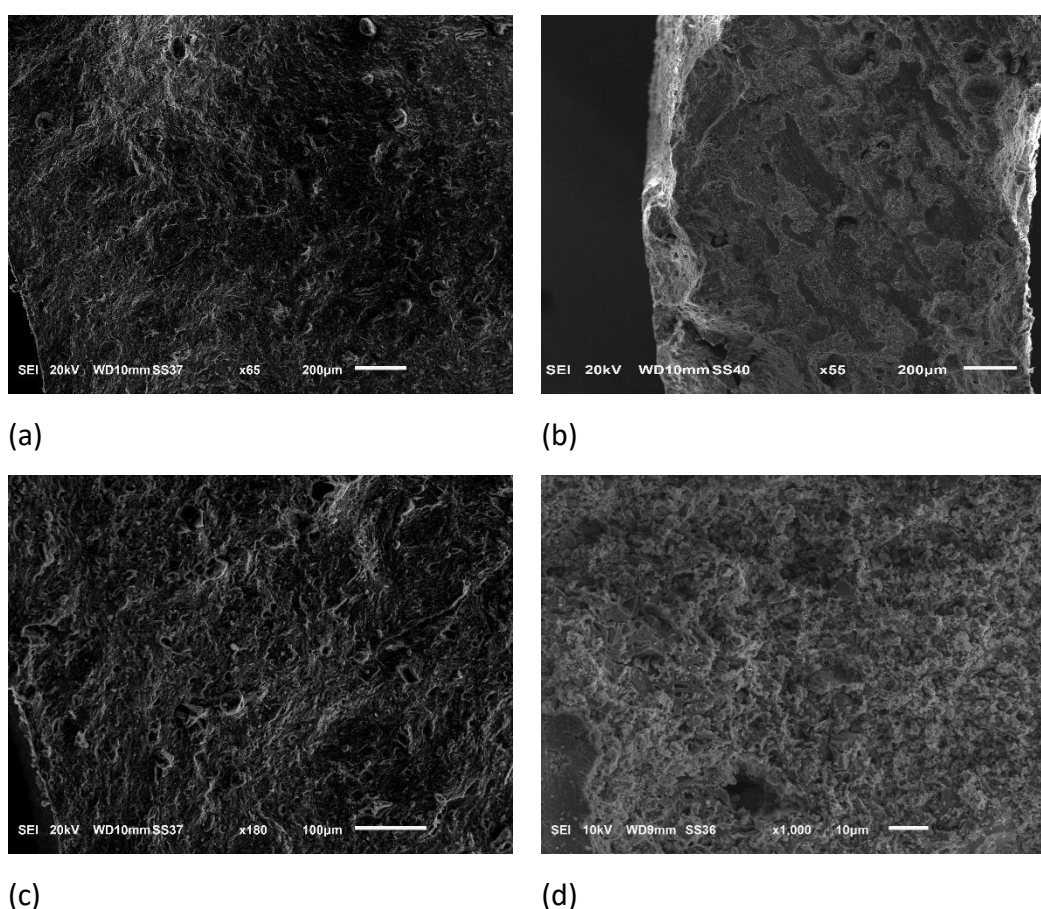


Figure 3. The tubular ceramic membrane was analyzed using scanning electron microscopy to obtain the cross-sectional view (a, b) and top view (c, d) of the membrane. Figures (a, c) and (b, d) are membranes M1 and M2, respectively.

The resulting membrane showed zeta potential as a function of pH, as presented in **Figure 4**. As shown, the membrane is slightly negatively charged across the entire pH range used in the analyses. Similarly, all proteins are negatively charged in an aqueous solution, with amoxicillin showing the highest negative charge, while levofloxacin shows almost neutral charge.

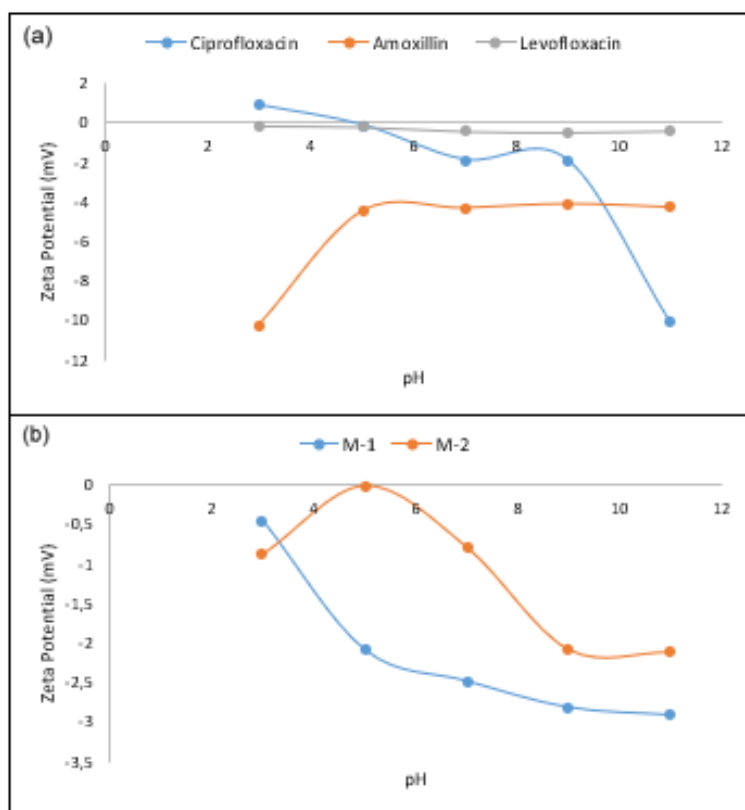


Figure 4. Zeta potentials for (a) antibiotics, (b) membranes.

Different operating parameters were then studied to investigate their impact on antibiotic removal using two ceramic membranes. These parameters were the antibiotic's concentration, feed solution temperature, and pH.

3.1 Effect of antibiotic concentrations

To investigate the impact of initial antibiotic concentration, the concentration was adjusted in each experiment run while maintaining a constant feed temperature of 50 °C and pH of 6.5. The obtained data showed that a high removal of antibiotics from water was achieved by the ceramic membranes (M1 and M2). The removal of antibiotics, including amoxicillin, ciprofloxacin, and levofloxacin, was highly significant, reaching a rate of up to 99.9%. Despite the use of various concentrations for each antibiotic in the water, it did not seem to affect the removal rate (**Figure 5**). When comparing the membranes' performance, the results showed insignificant differences in their removal rates of antibiotics ($p < 0.05$).

However, it is noticeable that the removal of amoxicillin is consistently the highest, while that of ciprofloxacin is consistently the lowest, even at a similar initial concentration. This is because the flux of ciprofloxacin is the highest across the membrane. To explain this behavior, we must examine the concept of liquid entry pressure (LEP), which is the minimum pressure required to overcome the membrane's hydrophobic forces and allow the feed liquid to penetrate the pores. LEP is proportional to the surface tension of the liquid, i.e., a mixture of water and the antibiotic—other studies, such as those reported by Vieira et al. [19] showed amoxicillin/water surface tension of above 60 mN/m while Jangde et al. [20] reported ciprofloxacin/water surface tension at a lower value of around 50 mN/m. LEP works similarly to osmotic pressure in a typical pressure-driven membrane. Therefore, the flux of ciprofloxacin is higher than that of amoxicillin at the same operating condition, resulting in lower removal of ciprofloxacin.

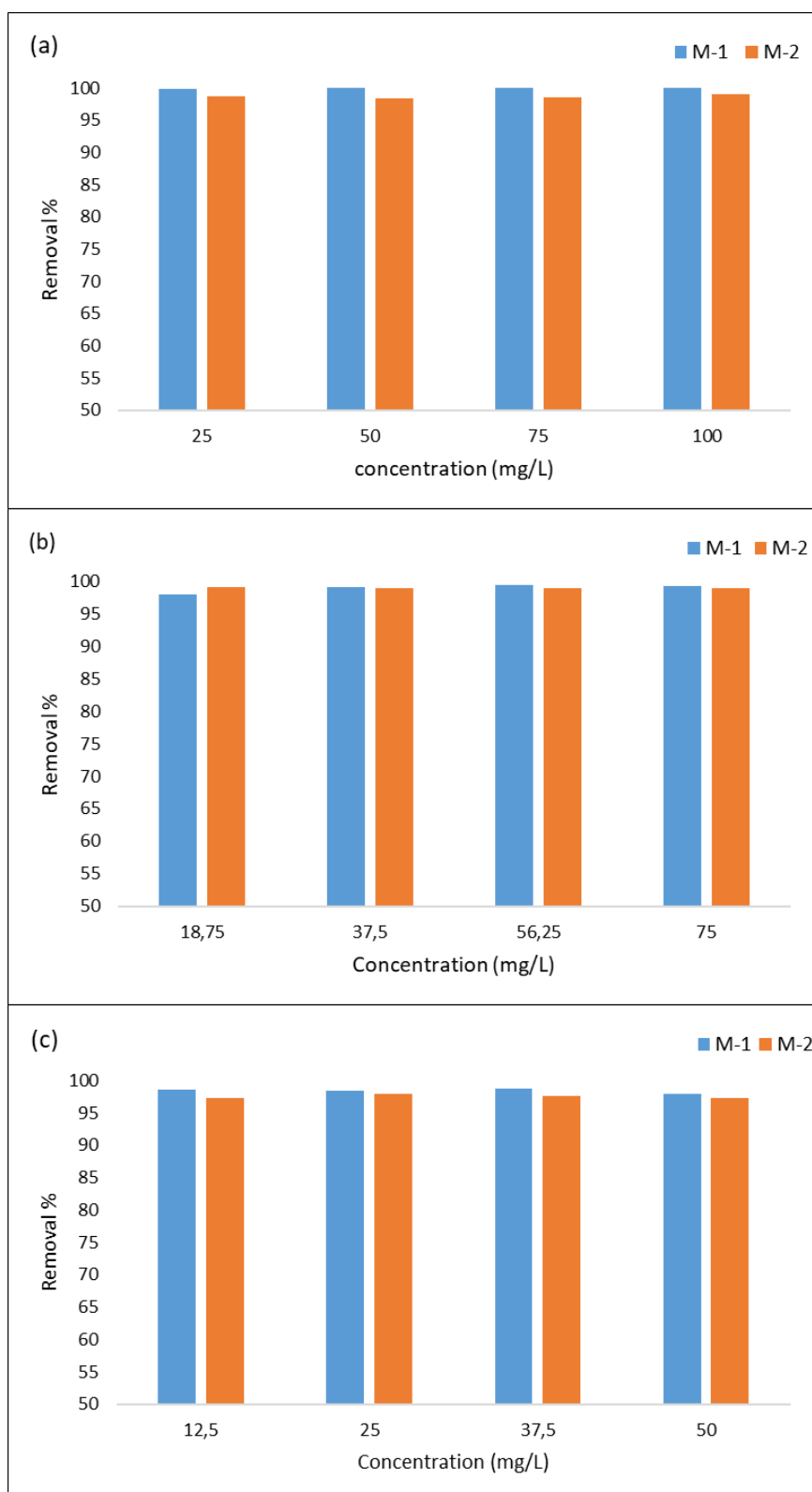


Figure 5. Effect of antibiotic concentration on the removal rate. (a) amoxicillin, (b) ciprofloxacin, (c) levofloxacin.

Additionally, **Figure 5** indicates that the M1 membrane achieves slightly higher removal than the M2 membrane in all cases. It is owing to the zeta potential of M1, which is a higher negative charge compared to that of M2, as evidenced in **Figure 4**. Hence, M1 exhibits a higher repulsive force to antibiotics than M2 does.

3.2 Feed temperature effect on the removal

Higher feed temperatures typically result in increased permeate flux due to more significant vapor pressure differences. Thus, to study the impact of the feed solution temperature on the antibiotic's removal, the feed solution temperature was adjusted to 50, 60, 70, and 80°C.

The protein removal results are shown in **Figure 6**. The results demonstrate high antibiotic removal across the studied temperatures for both membranes, with removal levels exceeding 98% for all antibiotics. It is also evident that as the temperature increases, the diffusivity of all proteins passing across the membrane increases. Thus, the removal percentage decreases as a result.

Further investigation, the **Figure 6** illustrates the relationship between the increase in feed solution temperature and the corresponding increase in flux. As mentioned earlier, this was attributed to the rise in vapor pressure differences and permeability, which results in a higher driving force for the water to diffuse across the membrane, due to the rising temperature across the membranes.

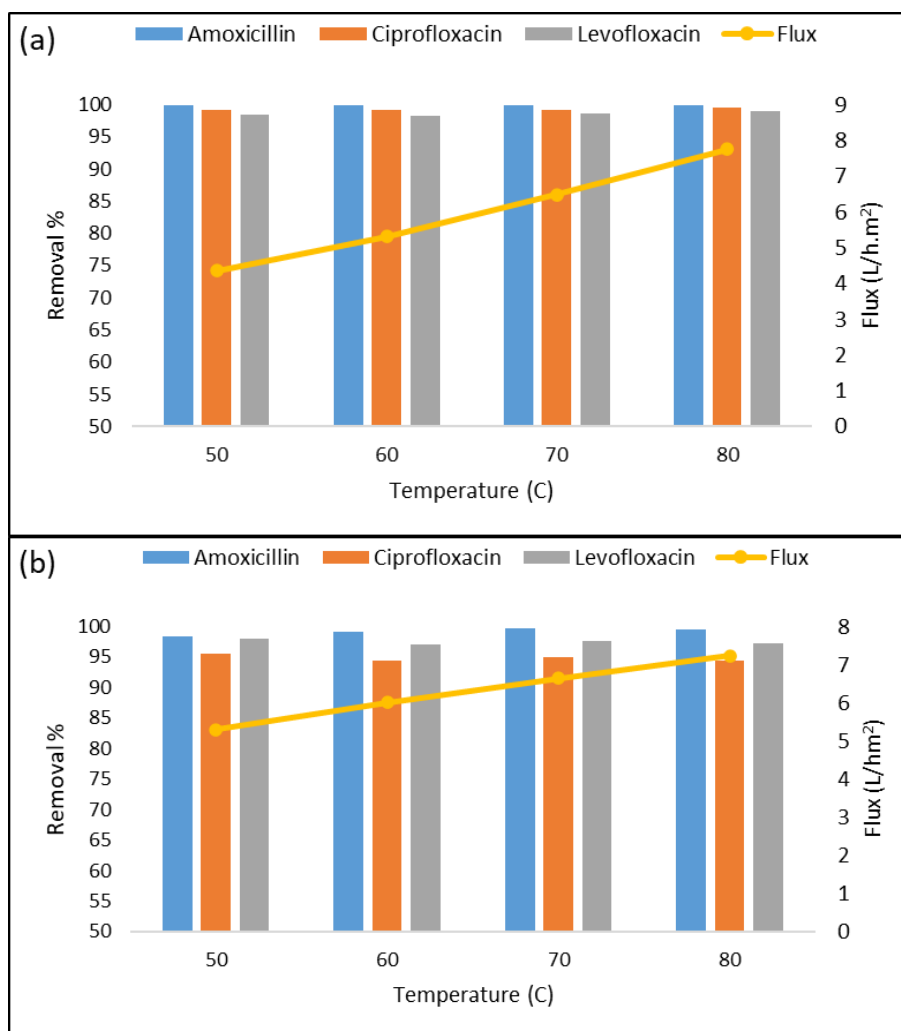


Figure 6. Effect of feed solution temperature on the antibiotic's removal. (a) M-1, (b) M-2.

3.3. Effect of feed solution pH

The influence of pH on antibiotic removal was studied by maintaining constant antibiotic concentrations at 50, 37.5, and 25 ppm for Amoxicillin, Ciprofloxacin, and Levofloxacin,

respectively, and a solution temperature of 50 °C. The solution pH was adjusted to 4, 6.5, 8, 10, and 12 by adding NaOH or HCl. The results of both membranes are shown in Error! Reference source not found.. The removal of amoxicillin and levofloxacin was over 98% in all the studied pH levels, while ciprofloxacin removal was the lowest at pH 4 and increased to almost 98% when the solution pH was increased to 6.5. These results were consistent with the measured zeta potential of the antibiotics and membranes. Similar results have also been observed in previous studies [16,20].

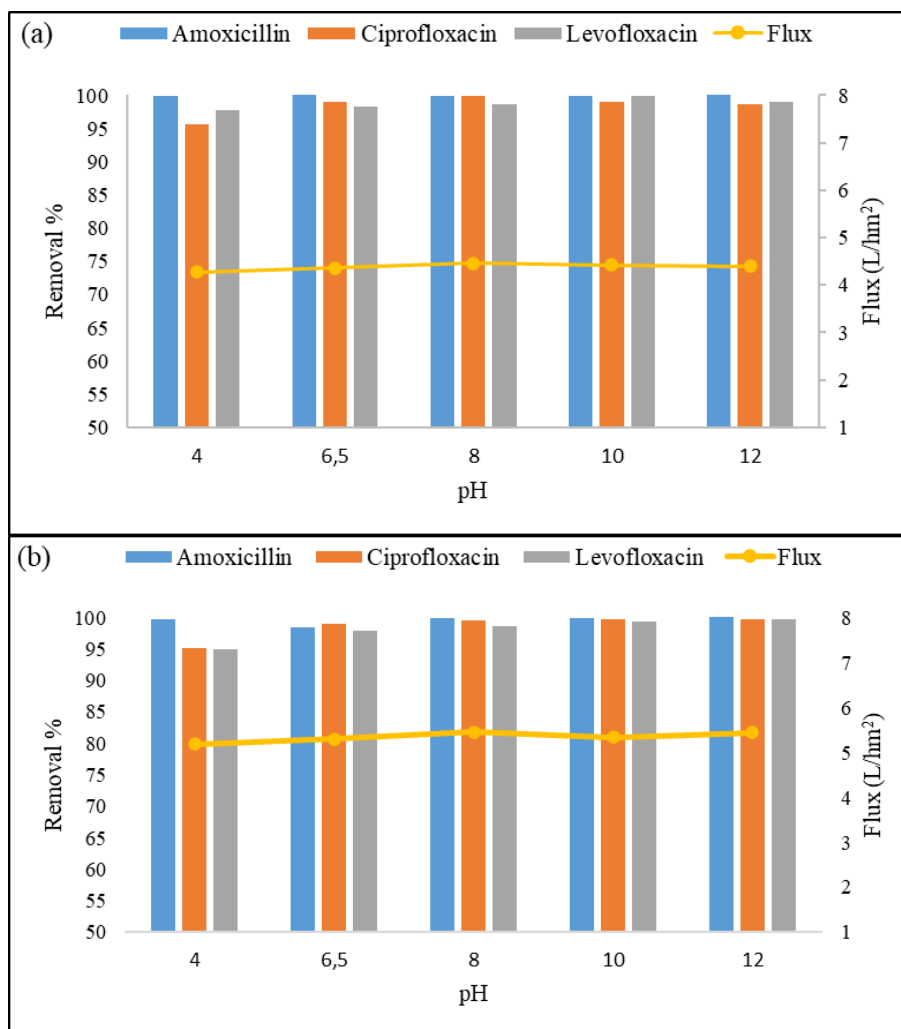


Figure 7. Effect of feed solution pH on the antibiotic's removal. (a) M-1, (b) M-2.

In general, antibiotics were rejected by both membranes at a rate greater than 95%. Even when the solution temperature, pH, and antibiotic concentration were varied, only a marginal change occurred in the removal percentage. This high rejection efficiency is achieved due to high LEP coupled with electrostatic repulsion.

3.4. Discussion

The ceramic membranes developed in this study exhibited strong and stable rejection performance across various environmental conditions, including different antibiotic concentrations, feed temperatures, and pH levels. Both membranes, particularly M-1, demonstrated effective separation mechanisms that are likely governed by a combination of surface charge interactions and liquid entry pressure. The slight variation in rejection rates among the three antibiotics can be explained by their molecular characteristics and

membrane interactions, as supported by zeta potential analysis and reference literature [19,20].

The enhanced performance of the ceramic membranes compared to polymeric counterparts previously reported [14-16] highlights the potential of ceramic-based systems for more robust and thermally stable operations. Additionally, the successful membrane regeneration using deionized water supports their practical utility for long-term applications with minimal fouling concerns. These findings confirm that ceramic membrane distillation systems can offer consistent rejection efficiency without being significantly affected by fluctuations in environmental or operational variables.

3.5. Relevance to Sustainable Development Goals (SDGs)

The removal of antibiotic residues from wastewater directly addresses key targets of the United Nations SDGs. The high rejection efficiency achieved in this study supports SDG 6 (Clean Water and Sanitation) by demonstrating an effective technology for improving water quality and treating pharmaceutical effluents. By mitigating the release of residual antibiotics into aquatic environments, ceramic membrane distillation can help reduce ecological contamination and preserve freshwater resources. Furthermore, limiting the environmental circulation of antibiotics contributes to the global effort to combat AMR, which is one of the primary concerns outlined in SDG 3 (Good Health and Well-being). By lowering the risk of resistant strain development due to environmental exposure, the application of ceramic MD supports both environmental protection and public health. As a result, the present study not only contributes technically to the field of water treatment but also aligns with broader global goals for sustainable development. This adds new information regarding SDGs as reported elsewhere [21-26].

4. CONCLUSION

This study examined the use of ceramic membrane distillation for the removal of antibiotics from wastewater. The membranes demonstrated consistent separation performance under different operating conditions. The results suggest that surface charge and membrane structure play a key role in the rejection process. This work provides a foundation for implementing ceramic-based membrane systems in wastewater treatment applications aimed at reducing antibiotic pollutants.

5. ACKNOWLEDGMENT

This work was supported by King Abdul Aziz City of Science and Technology (KACST), grant number 14-WAT3032-08.

6. AUTHORS' NOTE

The authors declare that there is no conflict of interest regarding the publication of this article. The authors confirmed that the paper was free of plagiarism.

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