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# Removal of Antibiotics from Wastewater by Comparison of Coagulation, Membrane and Adsorption Methods

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## ABSTRACT

In the present study, oxytetracycline (OTC) was selected as a medical contaminant with an organic structure to increase the efficiency and feasibility of comparison with other research studies. Therefore, the removal of OTC from medicinal effluents using different methods; namely, activated carbon, bentonite adsorption, iron(III) chloride coagulation, membrane filtration and reverse osmosis (RO), was investigated. Results indicated that the removal of soluble antibiotics in the factory wastewater by adsorption method was between 35% and 77%, at a pH of 6.5 and the total dissolved solids (TDS) and OTC concentration were reduced by 55% and 77%, respectively. In the coagulation method, where the suitable pH ranges from 5.5 to and 7.5, the optimal dosage of iron (III) chloride coagulation was between 40 and 100 mg/l, which resulted in 98.7% of OTC removal. It was concluded that the highest OTC adsorption of 98.7% in the RO membrane and TDS removal were obtained at a pressure of 9 bars. All the experiments on these membranes were performed within 4 months and the membranes still performed well. Therefore, it is concluded that long-term operation has little effect on the performance of the membrane and this membrane is a suitable membrane for the concentration of OTC. Finally, the RO is defined as the suitable method for OTC removal, after passing once through the membrane, while the other methods are proper to be used in the pre-treatment of medicinal wastewater.

**KEYWORDS:** Antibiotics, Absorption, Coagulation, Membrane filtration, Reverse osmosis.

## INTRODUCTION

Antibiotics are among the most non-biodegradable contaminants. They are considered important in the environment (Koch et al., 2021; Yang et al., 2021). If these drugs are not removed from wastewater, soil and other environmental divisions, they can threaten the quality of surface, ground and drinking waters. Antibiotics, which are not metabolized, often enter the aquatic environment (Salari et al., 2022; Kümmerer, 2001; Al-Ghazawi, 2021).

Most antibiotics are partially degraded in aqueous environments. Quinolones, for example, are adsorbed to a considerable amount of sewage sludge, soil and sediment and do not decompose. These antibiotics in different soils with a long half-life have fascinated a lot of attention (Hassanzadeh et al., 2014; Seifrtova et al., 2009; Brown and Balkwill, 2009; Jaradat et al., 2019). Previous studies indicated that the biodegradation of antibiotics in wastewater is not a sufficient and complete process for the removal of antibiotics (Atugoda et al., 2021; Oluwole et al., 2020).

Concentrations below the therapeutic level play an

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important role in the selection of resistant bacteria and genetic transmission in identified bacteria. Exposure of bacteria to concentrations below the therapeutic level of antimicrobials is the cause of the resistance-incidence increase. Besides, resistance can be transmitted to other bacteria in other environments, such as groundwater or drinking water (Hassanzadeh et al., 2015; Brown and Balkwill, 2009). It is important to protect groundwater, one of the most important natural resources, from such pollutants (Ostad-Ali-Askari et al., 2021; Ostad-Ali-Askari and Shayannejad, 2021; Ostad-Ali-Askari, 2022).

Oxytetracycline (OTC) belongs to the family of tetracyclines and was first identified in 1949 by Woodward (Avisar et al., 2010; Rivas et al., 2020; Hassanzadeh et al., 2014). This substance inhibits bacterial growth and reproduction by restraining the synthesis of bacterial proteins (Uddin et al., 2021; Seifrtova et al., 2009). Water-soluble OTC has a molecular formula of $C_{22}H_{24}N_2O_9$, a molecular weight of 496 gr/mol, a pH of 6.5 and two absorption wavelengths of 290 and 348 nm, belonging to carbonyl functional groups (-C=O-) and hydroxyl (Hassanzadeh et al., 2014; Brown and Balkwill, 2009).

Due to the necessity of eliminating antibiotics from the environment, different methods of removal have been performed by researchers, where each method has its own merits and demerits (Teymouri et al., 2023a; Teymouri et al., 2023b; Pavia, 1987; Li et al., 2008; Lee et al., 2020; Patidar and Sirvastava, 2021; Cuerda-Correa et al., 2021; Elkacmi and Bennaiah, 2019). These methods include physical operations, such as liquid-phase extraction, adsorption, membranes, biological processes, as well as chemical processes, such as electro-chemistry, sound waves, chemical oxidation and advanced oxidation (Salari et al., 2018; Shoorangiz et al., 2019; Rakhshandehroo et al., 2018; Salari et al., 2018; Salari et al., 2021; Teymouri et al., 2023a; Teymouri et al., 2023c; Teymouri et al., 2020; Salari et al., 2019).

The adsorption process is one of the most common methods for removing organic pollutants from the aquatic environment. In this method, temperature, contaminant concentration, type of adsorbent, contact time, pH, contact surface and mixing speed are important parameters in optimizing removal efficiency (De Gisi et al., 219; Khader et al., 2021). Activated carbon is a well-known adsorbent in treating biologically resistant contaminants and has been used to remove many antibiotics. However, there is a main problem in this method, which is carbon regeneration and non-destruction of the antibiotic molecules (Zadaka et al., 2007). Membrane processes are also used to remove biodegradable hard pollutants and ultrafiltration, nanofiltration and reverse osmosis are the membrane processes useful in removing antibiotics with 90-99% efficiency (Derakhshan et al., 2016).

Notwithstanding, operating these systems is very costly and the membranes are easily clogged and destroyed. In addition, the destruction of the molecular structure has not been completely accomplished and the antibiotics remained in the resulting sludge (Koyuncu et al., 2008). OTC removal has been done from physical operations, such as adsorption and photolysis, as well as biological and chemical processes, including ozonation, $UV/TiO_2$ photocatalysis, advanced oxidation ($UV/H_2O_2$) and Fenton (Hasanzadeh et al., 2014).

Biological and chemical removal methods alone cannot be used to effectively remove antibiotics. Sometimes, when using some methods to remove antibiotics, the toxicity of treated wastewater remains unchanged or it becomes even more toxic than the original compounds. In addition, all these methods are not used due to complexity and high operating cost, although combined processes can be introduced as the best solution for the treatment of wastewater containing antibiotics (Derakhshan et al., 2016).

Unfortunately, in spite of extensive investigations, there is still a considerable lack of integration for the removal of OTC antibiotics. Therefore, in the current study, the removal of these compounds from aqueous solutions to determine the effect of adsorption, coagulation and membrane-filtration method was studied. Hence, in these methods, three goals; namely, the removal of TDS, electric conductivity (EC) and OTC were considered. Different percentages of activated carbon and bentonite for the adsorption method, different ranges of pH and coagulants were chosen for assessing the performance of the coagulation method and finally, pH and pressure were taken into account for membrane filtration.

## MATERIALS AND METHODS

The optimization of three methods of absorption, coagulation and reverse osmosis is explained below.

Adsorption processes are usually optimized by the following isotherms. These isotherms are simply the relationship between moles adsorbed per unit mass of adsorbent and the adsorbed concentration remaining in the solution in equilibrium at a constant temperature (isothermal). Advanced coagulation is considered in the removal of organic carbon materials using a two-step method. The first step is to reduce a certain percentage of incoming organic carbonaceous materials based on organic materials and raw-water alkalinity, while the second step is to remove organic carbonaceous materials when, according to the quality characteristics of raw water, the desired percentages of organic carbonaceous materials (TOC) are removed in this stage. In the case of these methods, it is necessary to perform the jar test to reach the criteria of the second stage of TOC removal and to determine the type and amount of coagulant. Optimizing the RO process was achieved by changing pressure and pH.

**Measurement of OTC**

The effluent was collected from Aras Bazaar pharmaceutical company in Iran and OTC production centrifuges were provided and transported in the laboratory experiment possible time .95% pure OTC powder is also provided by the same company. Other required materials are potassium permanganate ($KMnO_4$), sodium hydroxide (NaOH) and distilled water. All of these materials are made on a laboratory scale by the German company named Merck.

Two high-performance methods, high-performance liquid chromatography (HPLC) and spectroscopy, have been extensively used to measure the amount of antibiotics. The spectroscopic method has been used to measure OTC in the pharmaceutical effluent. To acquire a proper method for quantitative analysis of the samples, spectrophotometer (Tiehm et al., 2009; Hasanzadeh et al., 2014), stage injection analysis (Kumar et al., 2011) and high-performance liquid chromatography (Salem and Saleh, 2002; Douˇsa and Hosmanova, 2005) were considered. Eventually, the spectrophotometer method was recognized as the suitable method to analyze the samples. This selection was based on criteria, such as the accuracy of the method, the tools required for the analysis, the operating conditions required, the response range of the methods, the lower cost of this experiment compared to other methods and the relative simplicity of the method.

**Method of Analysis**

The method of Ashuk Kumar et al. was used to analyze the samples (Shaojun et al., 2008). According to this method, pure oxy-tetracycline powder, potassium permanganate, sodium hydroxide and distilled water are required. The basic principle of the experiment for oxidation is manganese-capacity alteration. Manganese ions tend to be greener in alkaline environments and more colorless in neutral environments. This behavior of the permanganate is the main basis of the spectrophotometer method. The role of sodium hydroxide in this method is to create an alkaline environment for the reaction. The resulting material is blue and has a maximum wavelength of 348 nm.

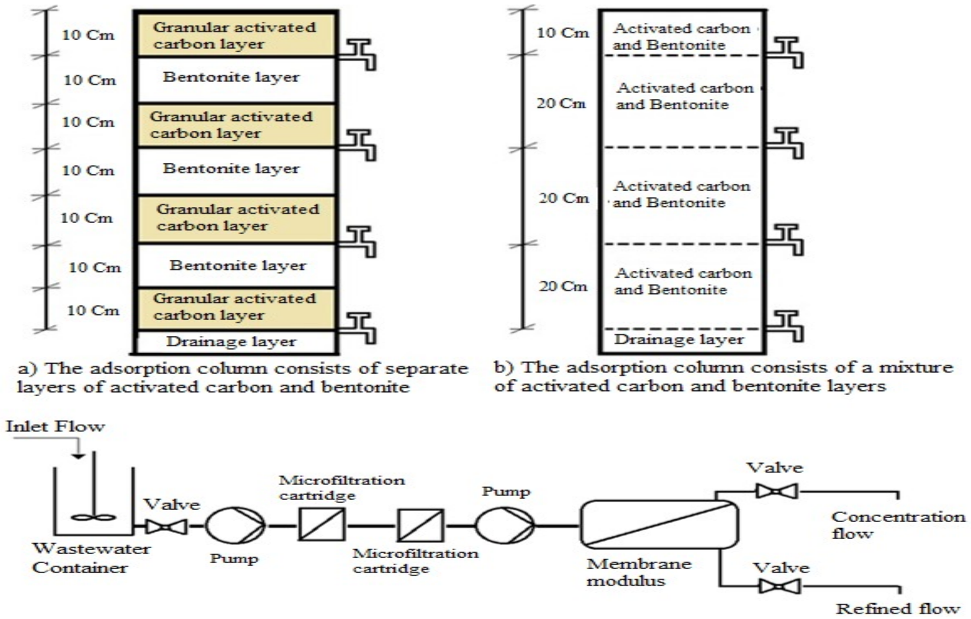
**Making Standard Solutions and Using Reagents**

First, the standard solution was made by using 0.001 g equivalent to 1 mg of OTC powder in 50 ml of distilled water. Then, to make the experiment solution, an amount of 5 ml of the previous solution is added to 50 ml of distilled water. Then, 1M NaOH and 0.01M $KM_nO_4$ solutions are prepared with the appropriate ratios. After making the calibration curve, different volumes of 0.5, 0.7, 1, 1.2 and 1.5 ml of the experiment solution along with 1 ml of 1M NaOH were poured into 1.5 ml of 0.01 M $KM_nO_4$ and the volume of the solution was 10 ml. At the wavelength of 348, the control sample is introduced and then, the other vials are placed in the device after 4 minutes of making the solution. Then, the calibration curve is drawn by obtaining the absorption coefficients. Using this linear equation, the absorbed value can be obtained for each concentration of the sample. All materials in this method have been purchased from the German company Merck.

**Method of Determination of Antibiotic Concentration**

The concentration of OTC was measured by a PC Spectro spectrophotometer. In spectrophotometric methods, due to the passage of light with a specific wavelength and the amount of light absorbed by the solution, the desired concentrations of contaminants in the solution can be measured.

Fig. 1 schematically shows the membranes of the membrane and adsorbent systems.




(c)

**Figure (1): Pilot performance of granular activated carbon adsorbent and bentonite (a) and (b) and (c) the pilot operation of the membrane system (c)**

The RO membrane pilots, along with the membranes, were available in the laboratory of the Khajeh Nasir al-Din Toosi University of Technology and were washed only to clean the membranes. However, due to the clogging of the activated carbon cartridges and microfiltration, these cartridges were renewed. Additional specifications of these cartridges, RO membranes and activated carbon adsorbents are as follows:

Granular activated carbon cartridge for RO pilots: This cartridge removes taste, odor and chlorine from the input solution. The operating temperature of this cartridge ranges from 4.4 to 51.6 degrees Celsius. Microfiltration cartridge for RO pilots: This cartridge removes sand, silt and turbidity from the input solution. The maximum operating temperature of this cartridge is 52 degrees Celsius.

**Pilot Setting-up**

The absorbent pilot should be careful not to take sampling valves with adsorbent materials and the system will work regularly. Other items are common to almost all of the methods discussed.

## RESULTS AND DISCUSSION

The relevant experiments were performed by considering the independent and dependent variables, as well as different input and output conditions, to reduce OTC contamination. Finally, the granular activated carbon adsorption column, as separate and mixed layers, of activated carbon and bentonite, iron (III) chloride coagulation and RO membrane system for removal of OTC antibiotic are compared and the optimal method is selected. The variables of the present study included changes in pH, applied pressure and antibiotic concentrations. Also, in the adsorbent column system, there is no semantic pressure change and this variable is removed. There are also two independent variables affecting the system performance, which are time and temperature, where in this research, they have been omitted from the study. The time variable is considered due to the small effect on removal. However, this effect was examined in the coagulation experiment and after the difference in results was insignificant, it was omitted. The difference in the percentage of removal by membrane filtration systems at 30 minutes and 24h is very small and negligible. Temperature is also not taken into account due to not-so-large changes in wastewater, because in fact, water currents have a relatively constant temperature. In this study, a temperature of a constant value of 25°C was assumed. The research environment in this study was the wastewater of Aras Bazaar pharmaceutical factory located in Amol Industrial town, Iran, which was used under different conditions of RO membranes and granular activated carbon adsorption column and coagulation.

**Adsorption Experiments**

Two different experiments were conducted on the activated carbon adsorption system. The first one is the adsorbent column containing 5-cm layers of granular activated carbon and bentonite, which was poured with a half of each. Unfortunately, the results were by no means satisfactory due to the substantially low infiltration rate, in which more than 60% of the inlet wastewater remained in the column after one week and only about 40% discharged from the adsorption column. Also, the poured layer-by-layer activated carbon and bentonite mud did not provide the desired results, because the water did not leave the bentonite layers. For this reason, this experiment was repeated as a mixture with different portions of granular activated carbon and bentonite.

A variety of pH (3, 5, 6.5 and 9.5) was considered for wastewater samples in both adsorption column systems. Also, the concentration of the antibiotic OTC has changed as well as the membrane systems. The results are shown in Tables 1-4. The optimum proportions of activated carbon to bentonite were obtained from 60 to 40 after repeating the experiments and this ratio was considered in the main set-up. The TDS and OTC removals were about 55% and 70%, respectively. According to the results, as the effluent passes through the underlying layers of the adsorption column, the removal efficiency of OTC and TDS increases. The highest removal occurred at a pH of 6.5 with 77% removal of antibiotics and 55% removal of TDS. Apart from this, the TDS removal process fluctuated at a pH of 9 and did not follow a regular pattern.

**Table 1. Absorption results at pH=3**

| R | Initial TDS | Final TDS | EC | Absorption Coefficient | OTC Removal (%) | TDS Removal (%) |
|---|---|---|---|---|---|---|
| First Valve | 450 | 400 | 590 | 13.2 | 35 | 11 |
| Second Valve | 450 | 378 | 520 | 10.25 | 50.5 | 16 |
| Third Valve | 450 | 389 | 448.3 | 9.7 | 53.3 | 13.5 |
| Fourth Valve | 450 | 320 | 333 | 6.5 | 69.6 | 28 |

**Table 2. Absorption results at pH=5**

| R | Initial TDS | Final TDS | EC | Absorption Coefficient | OTC Removal (%) | TDS Removal (%) |
|---|---|---|---|---|---|---|
| First Valve | 450 | 380 | 634 | 12.54 | 38.8 | 15.55 |
| Second Valve | 450 | 356 | 593 | 10.11 | 51.25 | 20.88 |
| Third Valve | 450 | 318 | 530 | 9.98 | 51.91 | 29.33 |
| Fourth Valve | 450 | 264 | 440 | 7.5 | 64.5 | 41.33 |

**Table 3. Absorption results at pH=6.5**

| R | Initial TDS | Final TDS | EC | Absorption Coefficient | OTC Removal (%) | TDS Removal (%) |
|---|---|---|---|---|---|---|
| First Valve | 450 | 354 | 590 | 10.11 | 51.25 | 21 |
| Second Valve | 450 | 312 | 520 | 7.1 | 66 | 30.6 |
| Third Valve | 450 | 269 | 448.3 | 5.12 | 76.6 | 40.2 |
| Fourth Valve | 450 | 200 | 333 | 4.9 | 77 | 55 |

**Table 4. Absorption results at pH=9.5**

| R | Initial TDS | Final TDS | EC | Absorption Coefficient | OTC Removal (%) | TDS Removal (%) |
|---|---|---|---|---|---|---|
| First Valve | 800 | 554 | 923 | 9.1 | 56 | 30.75 |
| Second Valve | 800 | 587 | 978 | 7.8 | 63 | 26.6 |
| Third Valve | 800 | 526 | 876 | 6.2 | 71.11 | 34.25 |
| Fourth Valve | 800 | 342 | 570 | 5.6 | 74 | 57 |

**Coagulant Experiments**

**Conducting the Jar Test**

To conduct the jar test, the same amount of experimented wastewater was poured into a beaker with a stirrer inside it. The amount of consent coagulant is adjusted to pH values of 3.5, 5.5, 7.5, 9.5 and 11.5, stirred for 120 minutes at a maximum speed of 120 rpm, followed by stirring for 20 minutes at a minimum speed of 25 rpm for flocculation. After that, at the optimal pH, different amounts of coagulants were added to the beakers. The amount of coagulant was ideal with the least amount of consumption, so that the best result is achieved in terms of flocculation. Then, the sample was mixed for one minute at a high speed of 120 rpm, followed by 20 minutes at a low speed of 25 rpm. In the next step, the sample of each beaker was tested for the sedimentation capacity of the formed clots. For instance, the volume of sediment deposited in each sample is determined at the specified time and the resulting changes are plotted *versus* pH to reach the best pH for the coagulation operation.

To perform advanced coagulation and TOC removal, some changes have been made to conventional water-treatment processes, which are designed only to remove turbidity from water. For that reason, some changes were performed on the type and dose of coagulant or polymer, as well as on coagulation conditions (Koyuncu et al., 2008).

The concentration of available antibiotics in the wastewater was 300 mg/l and 40 mg/l iron(III) chloride coagulant was utilized. Table 5 presents the results of OTC removal after 30 minutes. The results showed that TDS increased due to the breakdown of OTC molecules; so, the only focus was on the antibiotic concentration characteristic. The highest TOC removal was at a pH of 7.5. After that, the experiments continued and the results of OTC removal after 12 hours of contact time are represented in Table 6.

**Table 5. Coagulation-experiment results at different pH values with 40 mg/l iron(III) chloride after 30 min**

| Features | PH | Initial TDS (mg/l) | Final TDS | Absorption Rate | Initial Antibiotic Concentration | Final Antibiotic Concentration | Antibiotic Removal (%) |
|---|---|---|---|---|---|---|---|
| 1 | 3.5 | 180 | 225 | 1.201 | 300 | 10.069 | 96.6 |
| 2 | 5.5 | 96 | 130 | 0.99 | 300 | 6.8437 | 97.7 |
| 3 | 7.5 | 120 | 167 | 0.886 | 300 | 5.254 | 98.2 |
| 4 | 9.5 | 200 | 1050 | 2.266 | 300 | 26.35 | 91.2 |
| 5 | 11.5 | 300 | 1750 | 2.33 | 300 | 26.66 | 91 |

**Table 6. Coagulation-experiment results at different pH values with 40 mg/l iron(III) chloride after 12 hours**

| Features | PH | Initial TDS (mg/l) | Final TDS | Absorption Rate | Initial ntibiotic Concentration | Final Antibiotic Concentration | Antibiotic Removal (%) |
|---|---|---|---|---|---|---|---|
| 1 | 3.5 | 180 | 180 | 1.236 | 300 | 10.7565 | 96 |
| 2 | 5.5 | 96 | 140 | 1.337 | 300 | 12.1474 | 95.95 |
| 3 | 7.5 | 120 | 168 | 1.205 | 300 | 10.1298 | 96 |
| 4 | 9.5 | 200 | 840 | 2.233 | 300 | 25.8423 | 92 |
| 5 | 11.5 | 300 | 1530 | 2.34 | 300 | 27.3249 | 91 |

It was concluded that the optimal pH is 7.5. Consequently, different concentrations of iron(III) chloride coagulant; 10, 40, 70, 100 and 200 ppm, were considered for developing the experiment to find out the best concentration for OTC removal. The highest removal of OTC occurred at a dosage of 40 mg/l. Therefore, the coagulation experiment at pH of 5.5 to 7.5 and $FeCl_3$ reagent dosage of 40 to 100 ppm had the highest efficiency. Besides, the parameters were measured again after about 20 hours.

**RO Membrane Filtration Experiment**

While performing RO experiments, severe clogging happened due to the presence of BOD, COD and TSS. In fact, in a real system, the parameters of BOD, COD and TSS are basically removed in the pre-treatment. At a constant pH of 6.5, the optimum pressure was obtained from intermediate pressures of 5, 7, 9 and 11 bars for the RO system. The pH values then were experimented with at the optimum pressures of 3, 5 and 9. Table 7 illustrates the OTC and TDS removals at a pH of 3 (acidic aqueous). The removal efficiency of antibiotics experienced an increase as the pressure increased and the highest TDS removal occurred at a pressure of 7 bars. According to Table 10, increasing the pressure also resulted in an increment in the antibiotic removal and the highest TDS removal happened at a pressure of 9 bars followed by a reduction in TDS removal. As indicated in Table 9, at a pH of 6.5, increasing pressure led to an increasing rate of antibiotic removal and the highest rate of TDS removal occurred at a pressure of 7 bars which was followed by a reduction of TDS removal as the pressure increased. Based on Table 10, at a pH of 9, it can be concluded that by increasing the pressure, the removal efficiency of OTC antibiotic increased, whereas the removal of TDS has an increment only up to 9 and declined after that. Hence, the highest OTC and TDS removals were at the optimal pH of 9.

**Table 7. RO-experiment results at different pressures and pH = 3**

| Features R | Pressure (variable) | Initial TDS (mg/l) | Final TDS | pH | Initial Antibiotic Concentration | Final Antibiotic Concentration | TDS Removal (%) |
|---|---|---|---|---|---|---|---|
| 1 | 5 | 320 | 170 | 3 | 1.98 | 92.6 | 46.8 |
| 2 | 7 | 320 | 140 | 3 | 1.75 | 93.8 | 56.25 |
| 3 | 9 | 320 | 158 | 3 | 1.68 | 94.2 | 50.62 |
| 4 | 11 | 320 | 421 | 3 | 0.923 | 98 | 49.39 |

**Table 8. RO-experiment results at different pressures and pH=5**

| Features | Pressure (variable) | Initial TDS (mg/l) | Final TDS | pH | Initial Antibiotic Concentration | Final Antibiotic Concentration | TDS Removal (%) |
|---|---|---|---|---|---|---|---|
| 1 | 5 | 365 | 27 | 5 | 1.876 | 93.2 | 92.6 |
| 2 | 7 | 365 | 22 | 5 | 1.605 | 94.58 | 93.97 |
| 3 | 9 | 365 | 20 | 5 | 1.276 | 96 | 94.5 |
| 4 | 11 | 365 | 26 | 5 | 1.056 | 95.9 | 92.8 |

**Table 9. RO-experiment results at different pressures and pH = 6.5**

| Features | Pressure (variable) | Initial TDS (mg/l) | Final TDS | pH | Initial Antibiotic Concentration | Final Antibiotic Concentration | TDS Removal (%) |
|---|---|---|---|---|---|---|---|
| 1 | 5 | 220 | 24 | 6.5 | 1.5 | 95.12 | 89 |
| 2 | 7 | 220 | 20 | 6.5 | 1.15 | 96.9 | 91 |
| 3 | 9 | 220 | 20 | 6.5 | 1.132 | 97 | 90 |
| 4 | 11 | 220 | 27 | 6.5 | 0.98 | 97.7 | 87 |

**Table 10. RO-experiment results at different pressures and pH = 9**

| Features | Pressure (variable) | Initial TDS (mg/l) | Final TDS | pH | Initial Antibiotic Concentration | Final Antibiotic Concentration | TDS Removal (%) |
|---|---|---|---|---|---|---|---|
| 1 | 5 | 850 | 205 | 9 | 1.236 | 96.4 | 75.9 |
| 2 | 7 | 850 | 134 | 9 | 0.904 | 98.1 | 84.3 |
| 3 | 9 | 850 | 102 | 9 | 0.909 | 98.12 | 88 |
| 4 | 11 | 850 | 210 | 9 | 0.786 | 98.7 | 75.3 |

The results obtained in this study are consistent with other previous studies. For example, in a study by Nasrollahi et al. (2022), membrane processes with advanced oxidation processes (AOPs), surface adsorption and biological treatments to remove antibiotics were investigated. The results showed that all methods with different percentages have the ability to remove antibiotics, each of which has its own advantages and disadvantages. Also, in another study conducted by de Ilurdoz (2022), the effect of combined processes in removing antibiotics from aqueous solutions was investigated. The results showed that hybrid processes are also mentioned as a good alternative. In this regard, biological methods (biological aeration filter, anaerobic digestion and biologically active carbon filter) and membrane technology (nanofiltration and reverse osmosis) had the best removal percentage (ranging from 80% to 100%), while chemical methods (coagulation-flocculation) and constructed wetlands (constructed wetlands with the horizontal subsurface flow) had the worst results (below 80%).

## CONCLUSIONS

In this work, the removal of OTC from drug effluents through different methods; namely, using activated carbon and bentonite adsorption, iron(III) chloride coagulation and RO membrane method, was investigated and the following conclusions are drawn:

The effects of adsorbent dose, contact time, pH, changes in pH, applied pressure and initial OTC concentration on the OTC removal from aqueous solutions have been investigated. Furthermore, the aforementioned variables have important effects on the process; high concentration of OTC negatively affected the removal efficiency and contact time at high levels caused desorption.

Activated carbon and bentonite adsorption caused a reduction of 55% and 77% of TDS and OTC at a pH of 6.5, respectively. Therefore, this method is only suggested for the pre-treatment section and is not recommended for the main treatment due to the remaining OTC residues, being time-consuming, not breaking the OTC molecules and only separating them, as well as having no financial benefits.

Moreover, a pH of 5.5 to 7.5 and iron(III) chloride coagulant dosage of 40 to 100 mg/l were considered as suitable ranges for the coagulation method. In these ranges, the OTC removal was 98.7% and molecule breaking was observed in the reduction of TDS.

The results for the RO membrane method showed that the operating pressure has a great effect on the performance of removal efficiency; thus, a pressure of 9 bars was determined as a suitable pressure which resulted in a 98.7% reduction of OTC and TDS removal. Apart from this, at high OTC concentrations, the feed concentration polarization was observed which reduced the membrane performance efficiency. For this reason, using RO at a high level of OTC is not recommended. The membranes were functional for more than 4 months, which indicated that a long-term operation would have little effect on the membrane performance. Eventually, RO is considered the most efficient method for OTC removal and adsorption and coagulation could be considered as pre-treatments to increase the work efficiency.

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